Improving Drying Efficiency and Energy Saving for Crumb Natural Rubber Drying with Combined Drying Technologies

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ABSTRACT

Natural rubber (NR) or *cis-1, 4-polyisoprene* is obtained from the latex that is tapped from a tree known as *Hevea Brasiliensis*. It can be in the form of preserved latex concentrates or further processed into sheets, crepes, block rubber that associated with its unique characteristics such as great elongation strength, excellent building tack, low rolling resistance, low heat build-up and good low-temperature performance. These are essential characteristics in dynamic working environment and application especially for vehicle tyre and rubber thread. To produce good quality of technically specified rubber or block rubber from raw natural rubber, drying process plays a vital role in the raw rubber processing. To date, the level of technology involves rubber drying is still inefficient and primitive. Non-uniform drying and the occurrence of wet rubber in the final product are the major drawbacks when using the conventional type of trolley dryer. Also, fuel and electricity usage for a single drying operation is noticeably intense and costly. Indeed, the main limitation of conventional drying method is the drying time that is mainly dependent on temperature gradient inside the product and cannot be sustainably reduced because the product is heat sensitive; whereas drying at high temperature for prolong hours could result in quality deterioration too.

The prime objective of this research was to investigate combined drying as a mean of crumb rubber processing to reduce the drying time, energy consumption and to produce the desirable dried product. In the present study, crumb rubbers were dried by alternative drying strategies whereby additional drying technologies such as hot

air, vacuum, and microwave was integrated into the drying system. Both engineering properties (drying kinetics, effective moisture diffusivity and specific energy consumption) and physicochemical properties (colour, thermal oxidative level, rheological, textural and surface topography) of all dried products were measured and evaluated. The comparisons were made against samples from hot air drying.

The finding shows the drying time of microwave-convective drying (MWHA) was found 89% and 50% shorter than convective hot air drying (HA) and microwave (MW) drying, respectively. In terms of drying rate performance, the maximum drying rate of MWHA was found 455% and 62.5% higher than HA and MW drying, respectively. On the other hand, effective diffusion coefficient (D_{eff}) values of both MW drying and MWHA were ranging from 1.48 x 10^{-8} m^2/s to 5.59 x 10^{-8} m^2/s that is six times higher than HA which has lowest D_{eff} recorded at 0.22 x 10^{-8} m^2/s. It is noteworthy to mention that the energy consumption of MWHA is much lower than other drying strategies due to its excellent time saving and energy efficient features in the drying process. The average specific energy consumption (SEC) values of MWHA's drying strategies were scored below 0.22 MJ/ gH_2O whereby HA drying yields SEC values of 0.36-0.44 MJ/gH_2O and highest was recorded in vacuum assisted drying which scored at 0.75 MJ/ gH_2O. MWHA dried rubber has exhibits lightest colours, minimum total colour change and consistent colour appearance over the range of drying methods. The lesser colour change can be an indication of reduced oxidative reaction and hence better ageing properties of dried rubber. This is corresponding to the high value of activation energy obtained by MWHA's dried

rubber in the thermal degradation study by using the Coats-Redfern methods. Similarly, the rheological properties of MWHA dried rubber including PRI and P_o were found in par and the volatile matter was 28.6% lower in comparison with SMR 20 block rubber. In terms of springiness indices, MWHA dried rubbers gave the highest at 72.23% showing marked performance in mechanical properties. In a nutshell, the potential of combined drying techniques in crumb rubber has been proven through this research and able to improve both engineering and physicochemical properties while the drying time is significantly reduced. Furthermore, these findings will supplement the knowledge on crumb rubber drying process which is beneficial to raw rubber processing industries in having economically feasible drying option to be applied to the existing plants.

PUBLICATIONS

Part of the studies in this dissertation has been submitted for publications.

Book Chapter

1. **Tham, T.C.**, Hii, C.L., Ong, S.P., Abdullah, L.C., and Law, C.L, (2015). Measurement on the Effect of Microwave Energy in Drying-Technical Review. In *Processing of Foods, Vegetables and Fruits: Recent Advances*, Eds. Hii, C.L., Jangam, S.V., Ong, S.P., Show, P.L., Mujumdar, A.S., pp. 61-72.

International Journals

1. **Tham, T. C.**, Hii, C.L., Ong, S.P., Chin, N.L., Abdullah, L.C., and Law, C.L., (2014) 'Technical Review on Crumb Rubber Drying Process and the Potential of Advanced Drying Technique', Agriculture and Agricultural Science Procedia, 2, pp. 26–32.
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2. Ng, M. X., **Tham, T.C.**, Ong, S.P., and Law, C.L., (2015) 'Drying kinetics of technical specified rubber', Information Processing in Agriculture, 2(1), pp. 64–71.
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3. **Tham, T.C.**, Ng, M. X., Ong, S.P., Hii, C.L., and Law, C.L., (2017) 'Application of microwave-assisted drying on specific energy consumption, effective diffusion coefficient and topological changes of crumb natural rubber (*Cis-1, 4- polyisoprene*)', Chemical Engineering and Processing-Process Intensification, 128, pp. 19-35
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Conference Papers

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LIST OF NOTATIONS AND ABBREVIATIONS

Nomenclature

$\bar{x}_1$	Mean of the first set of values
$\bar{x}_2$	Mean of the second set of values
D_{eff}	Effective diffusion coefficient (m^2/s)
$\|E\|$	The magnitude of internal electric field (V/m)
E_a	Activation energy (kJ/mol)
E_t	Total energy consumption (Wh)
F_1	Maximum force (N)
F_2	Force after 60 seconds (N)
L^*	Lightness value (CIE)
L_o^*	Initial L^* value (CIE)
M_i	Initial mass (g)
P_{30}	Rapid plasticity number of the test pellet after ageing at 140 °C for 30 minutes
P_d, P	The density of microwave power absorbed by dielectric material (W/m^3) or (W/g)
P_e	The rating power of equipment(s), W
P_o	Wallace rapid plasticity
R^2	Coefficient of determination
R_{adj}^2	Adjusted coefficient of determination

T_e	End temperature of degradation (°C)
T_o	Onset temperatures of decomposition (°C)
T_r	The temperature of the maximum mass loss rate (°C)
W_d	The weight of dry matter (g)
W_i	The weight of rubber (g) at that specific time t (minute)
X_N	Independent variable with N predictor variables
X_f	Equilibrium moisture content (% db.)
$\hat{Y}$	Dependent variables
a^*	Greenness/Redness value (CIE)
a_o^*	Initial a^* Value (CIE)
b^*	Blueness/Yellowness value (CIE)
b_o^*	Initial b^* Value (CIE)
f_n	Normalize drying rate
k_0	Value of the gradient
n_p	Positive integer
ΔE^*	Total colour change (CIE)
ΔL^*	Change L^* value (CIE)
Δa^*	Change a^* value (CIE)
Δb^*	Change b^* value (CIE)
111	10 minutes drying time each in three different MW power setting
321	15, 10 and 5 minutes drying time each in three different MW power setting

db.	Dry basis
dE_{ind}	Total colour difference with reference to industrial graded dried rubber (SMR 20)
dE_{raw}	The total colour difference with referring to raw crumb rubber (field coagula)
dX/dt	Drying rates (g H_2O/g DM. min)
E_t	Total power consumption in each drying bout, Wh
h	Time (hour)
H_2O	Water
K	Temperature (Kelvin)
kWh	kilowatt hour
mbar	millibar
N_1	Total number of values in the first set
N_2	Total number of values in the second set
R	Ideal gas constant
s_1	The standard deviation of the first set of values
s_2	The standard deviation of the second set of values
t	time (s)
t	t-test value
T_{air}	Ambient air temperature (°C)
W	Watt
wt%	Percentage by weight
x	Magnification level (times)

X_o	Initial moisture content (% d.b)
X_d	Moisture content (% d.b)
X_i	Moisture content at time *t*
A	Frequency factor
N	The number of experimental data points
T	Derivative peak temperature (°C)
f	Frequency (Hz)
n	Order of reaction
p, Sig.	Significant difference
$tan\delta$	Loss tangent coefficient
z	The number of parameters in the model
$\boldsymbol{a}$	Regression constant
$\boldsymbol{b}$	The regression coefficient for each predictor variable

Greek letters

α	The fraction of sample decomposed at time, *t*
ε'	Relative dielectric constant
ε_0	The permittivity of free space (8.86 x 10^{-12} F/m)
μm	Micro-meter
v, V	Air velocity (m/s)
σ	Effective conductivity (Siemens per meter)

$\emptyset$	Moisture ratio (Dimensionless)
ϕ_{exp}	Experimental dimensionless moisture ratio
ϕ_{pre}	Predicted dimensionless moisture ratio
χ^2	Reduced chi-square

Acronyms /Abbreviations

ADS	Air dried sheets
AHP	Analytical Hierarchy Process
ANOVA	One-way analysis of variance
AOAC	Association of Official Analytical Chemists
ASABE	American Society of Agricultural and Biological Engineers
CDC	Characteristic of drying curve
CIE	Commission Internationale de I'Eclairage
CRP	Constant rate drying period
CV	Constant viscosity
DM	Dry matter
DRC	Dry rubber content (%)
DSC	Differential scanning calorimetry
DTG	Derivative thermogravimetric
EMC	Equilibrium moisture content
FD	Freeze drying
FEA	Finite Element Analysis

FRP	Falling rate drying period
GP	General purpose
GRG	Generalized Reduced Gradient
HA	Convective hot air drying
HA/MW	Hot air-microwave drying (Two-stage drying)
LPG	Liquefied petroleum gas
MFD	Microwave-freeze drying
MR	Moisture ratio
MVD	Microwave-vacuum drying
MW	Microwave
MW/HA	Microwave-hot air drying (Two-stage drying)
MW/VD	Microwave-vacuum drying (Two-stage drying)
MWHA	Microwave-convective drying (Hybrid drying)
NMR	Nuclear magnetic resonance
NR	Natural rubber
PRI	Plasticity retention index (%)
RH	Relative humidity (%)
RMSE	Root mean square error
RPA	Rubber processing analyser
RPM	Rotation per minute
RRIM	Rubber Research Institute Malaysia
RSS	Ribbed smoked sheets
SD	Stepping down MW800 to MW600 (MW876)

SEC	Specific energy consumption (MJ/g.H_2O)
SEM	Scanning Electron Microscopy
SIR	Standard Indonesian Rubber
SMR	Standard Malaysian Rubber
STR	Standard Thailand Rubber
SU	Stepping up from MW600 to MW800 (MW678)
TG	Thermogravimetric
TGA	Thermogravimetric analysis
TPA	Texture Profile Analysis
TSR	Technically specified rubber
VD	Vacuum drying
VD/MW	Vacuum-microwave drying (Two-stage drying)
VM	Volatile matter (wt. %)

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CHAPTER 1

INTRODUCTION

1.1 Natural rubber (NR)

Natural rubber (NR) or *cis-1, 4-polyisoprene* is obtained from the latex that is tapped from a tree known as *Hevea Brasiliensis*. Rubber particles vary in size from 0.15-3μ with a molecular weight distribution of 10^5-10^7 g/mol (Baker, Fulton and Updated by, 2000). NR consists of *n* running of monomers of the organic compound isoprene, C_5H_8 to form polymer which makes up about 95% of the cream (water and rubber) and 5% of non-rubbers which include protein, amino acids, glycerides, fatty acids, trace of metal ions, inorganic and organic constituents (Sethu, 1981; Hasma and Othman, 1990; De and White, 2001). The chemical composition of fresh latex is given in Table 1.1.

Table 1.1: Chemical composition of fresh latex (De and White, 2001)

Constituent	**Percentage, %**
Rubber	30-40
Proteins	1-1.5
Resins	1.5-3.0
Minerals	0.7-0.9
Carbohydrates	0.8-1.0
Water	55-60

Latex from the field is coagulated by addition of acid or in some cases involved bacterial reaction in producing acids to promote the coagulation process which known as natural coagulation. NR latex is a colloid with a pH range of 6.5 to 7.0 in which majority of the dispersed phase is rubber whereas dispersion medium is water. The latex usually contains 30-40% of dry rubber by weight and 10-20% of the collected latex is concentrated by creaming or centrifuging and used in its latex form (Simpson, 2002); whilst remaining latex is processed in the form of dried rubber sheets, crepe and bales.

Latex is categorized into two different fields namely field latex and field coagulum. The filed latex is normally collected after four to five hours of tapping before deliver to the rubber processing plant. Unlike fresh latex collected from the plantation, field coagulum comprises of cup lump, tree lace, scraps stored for days are lower quality materials and hence the rubber product processed from these will be of inferior quality (De and White, 2001). Figure 1.1 shows the relationship between different sources of material from latex and respective SMR grading.

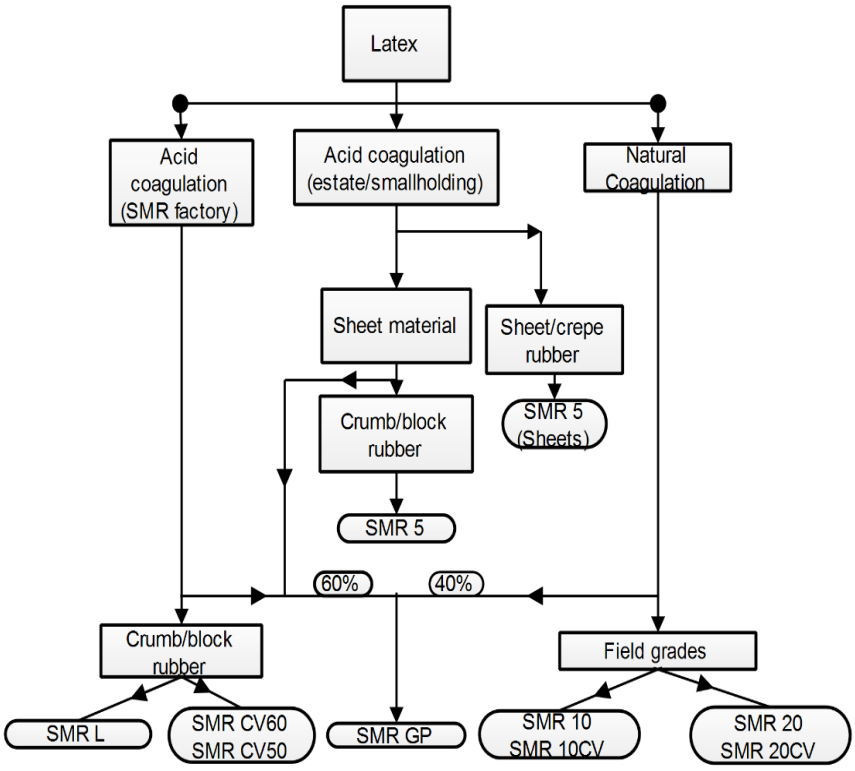


Figure 1.1: Various grades of SMR according to source materials and production processes (Baker, Fulton and Updated by, 2000).

1.2 Research background

The drying process is considering one of the midstream sectors in the rubber industry. The production of SMR rubber is initiated by the rapid coagulation of bulked latex after blended with formic acid to achieve an acidic environment of pH 5.0-5.2 and followed by 16 hours of maturing process. The coagulum is then pressed through stages of soaking, squeezing and shredding for dirt removal and comminution by series of heavy machinery including crusher, cutters, creper and pelletizers to produce small, uniformly granulated crumb rubber. Similar processes as shown in Figure 1.2 are applied to the field coagulum when the crumb is formed

through cleaning and size reduction processes. Granulated crumb rubbers are discrete when wet, but their natural tack returned during drying (Sethu.S, 1967). Both types of rubbers are dried in a trolley dryer at a temperature ranging from 120°C to 130° C to reduce the volatile matter to less than 0.8%. The entire bed of crumb rubber becomes a semi-coherent mass which could be lifted and handled as a block when drying is completed. The drying process is followed by cooling, baling, hydraulic pressing to final form and packed as finished goods.

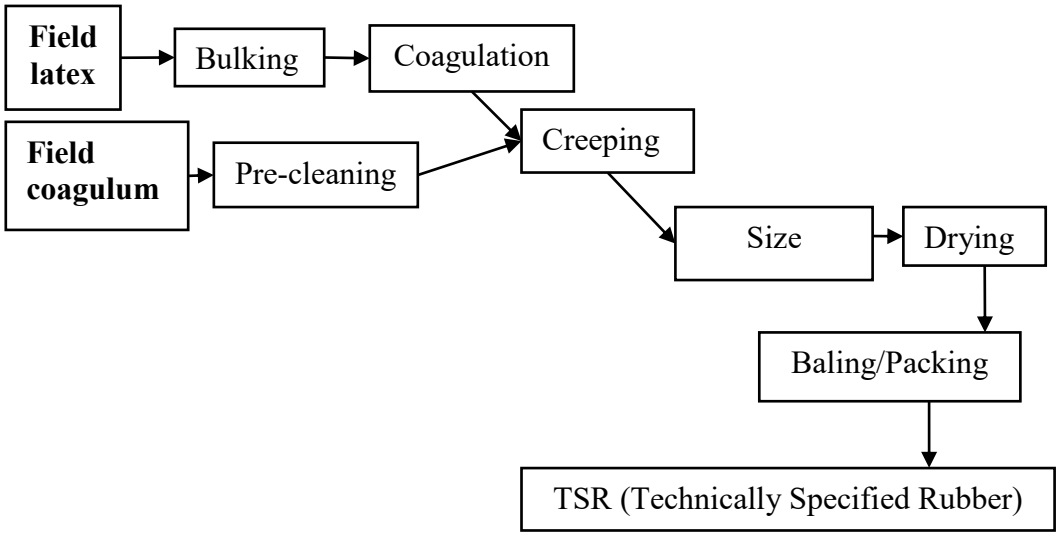


Figure 1.2: The process flow of raw rubber processing

The dryer that is used in this through-circulation drying process is a static bed trolley dryer operated with direct burner fuelled by diesel or gas, steam or thermal oil boiler with a heat exchanger (Golsta Sdn Bhd, 2015). Figure 1.3 shows the trolley dryer and dryer filled with crumb rubber.



Figure 1.3: Dryer configuration (left) and the trolley filled with fresh crumb rubber (right)

To date, the level of technology involved in the drying of rubber is still inefficient and primitive. Non-uniform drying is still the major drawback when using the conventional type of trolley dryer. A non-uniform drying happens when white spots or wet spots are spotted in the block of dry rubber. White spots are white localized patches of high moisture content in raw natural rubber that tend to resist breakdown on mastication and tend not to absorb carbon black as readily as compared to dry rubber on mixing (ISO, 2012).To ensure uniformity of rubber drying, the trolley machine must be equipped and supplied with a huge volume of air flow and operated at high air temperature for at least four to five hours. Apparently, the fuel and electricity cost for a single drying operation is noticeably intense and costly. Indeed, the main limitation of convective drying techniques is that drying time that is mainly dependent on temperature gradient inside the product and cannot be sustainably reduced because the product is heat sensitive. On the other hand, drying at high temperature for prolong hours could result in quality deterioration (Bateman and Sekhar, 1966; Zhang et al, 2012).

In recent dryer market, there are various types of drying technology available that could perform better than the conventional hot air drying. Dielectric heating with a microwave is one of the drying technologies that have been proven to greatly reduce the drying time of many agricultural commodities with the availability of proper control of the drying regime. Majority of the research shows that the dried product with quality attributes equivalent to those of convective dried material can be obtained (Tulasidas, Raghavan and Mujumdar, 1995; Sharma and Prasad, 2006a; Zhang *et al.*, 2006; Dadalı, Apar and Ozbek, 2007). However, some adverse effects of MW drying i.e. thermal runaway could cause thermal damage to the dry product. On the other hand, hybrid drying techniques or combined drying serve as a better alternative than single drying operation in terms of short drying hour, reduced operating cost, setup and quality retention of the dried product (Yongsawatdigul and Gunasekaran, 1996; Sharma and Prasad, 2004; Datta, Geedipalli and Almeida, 2005; Zhang *et al.*, 2006; Setiady *et al.*, 2007). The hypothesis suggested in this study is that it is possible to reduce the drying time and improve the energy saving for rubber drying process by using microwave as the prime source of energy input needed for heat and mass transfer within crumb rubber, whereas the convective air conveys the heat while also removing the moisture from the drying environment.

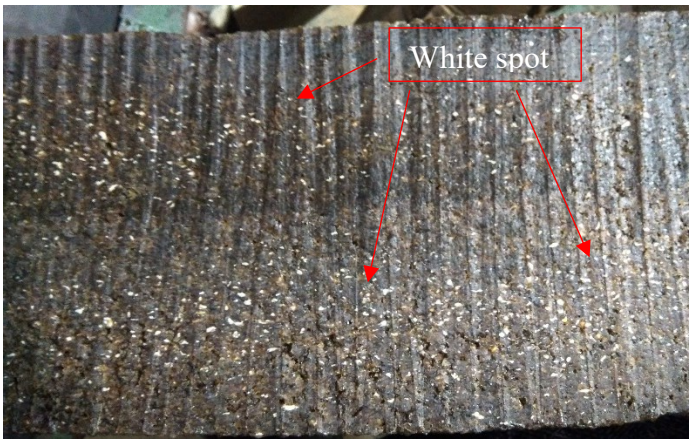
1.3 Challenges in crumb rubber drying process

The main aim of drying applies in the wet rubber processing is to remove the water phase of latex, as effectively and economically as possible and produce rubber in dry form before it is sold to buyers. Recent research and development of dry rubber

merely focused on the vast application of rubber end product and none of the research work has been focusing on the improvement of midstream processing of natural rubber involving machine design and processing technologies (Tasara, Tirawanichakul and Tirawanichakul, 2011). A similar claim was made by Tirawanichakul and Tirawanichakul (2008) where traditional drying method by using hot air is still widely practised in many block rubber factories located in many countries particularly in the Southeast Asian countries like Malaysia, Indonesia, Cambodia and Thailand. Besides, existing drying techniques involve in rubber processing are not energy efficient and pose several key quality issues. For instance, the drying system fails to guarantee uniform and thorough drying for the rubber even though after long cumbersome drying operation. Indeed, rubber is not very permeable to moisture and creating a dry 'skin' on the surface of the crumbs during conventional hot air drying will trap moisture inside. Nonetheless, the volatile matter, VM of the final product must lower than 0.6% to prevent mould growth or malodour (Rama Rao, 1984) while SMR technical specification addressed slightly higher to a maximum allowable VM of 0.8%. Figure 1.4 shows the white spots rubber often found in the dried rubber.



(a) Fresh crumb rubber (b) Dried rubber (SMR 20)



(c) White spot (wet spot) discovered in central of dried rubber

Figure 1.4: Drying of crumb rubber and its challenge

The white spot issue in dried rubber is possibly due to the lack of rigid knowledge in the moisture removal and heat distribution along the dryer zone including the dryer length, height, or volume depends on the dryer configuration. For example, an indefinite amount of moist air introduced over the length of dryer might vary the evaporation rate of the bed of materials and thus affects overall drying efficiency. On the other hand, Sainte-Beuve (2008) claims that white spots in dried rubber are mainly due to crumb heterogeneity on entering the dryer instead of quality or operation of the dryer. Hence, an optimal drying process requires a deep understanding of drying behaviours of crumb rubber, thermos-physical parameters correlated with different drying conditions (Tasara, Tirawanichakul and Tirawanichakul, 2011).

Substantial energy usage in the drying process is nevertheless another imperative factor that should be addressed by the existing drying system. According to Dasappa,

Paul and Kumar (2009), typical fuel consumption (diesel oil) is about 35 litres per ton of dry rubber (29.12 of LPG/ton dry rubber) corresponding to 1.24 GJ/ton dry rubber of heat energy. Therefore, a full day operation of five-ton trolley dryer will consume up 148.8 GJ/ton dry rubber which also equivalent to 41,333 kWh in electricity term. Such huge amount of heat energy is required to heat up the air, raw material, body structure (including container) of the dryer while the rest of the energy is to compensate the latent and sensible heat losses to surrounding (due to poor insulation) and exhausted to the atmosphere with moist air. Also, drying performance in rubber varies with the depth of the bed, whereby the thicker the rubber bed, the higher the energy consumption (Tasara, Tirawanichakul and Tirawanichakul, 2009). An energy survey conducted by Utomo, Hasanudin and Suroso (2010) stated that energy from the diesel fuel is mostly utilized in the drying stage of crumb rubbers which measured at 1.16707 MJ/kg of dry rubber (98.06% from total fuel energy consumption) in the production of low-grade crumb rubber, Specific Indonesian Rubber, SIR 20. Conversely, high-grade rubber, SIR 3 recorded about 97.9% out of total fuel energy consumption i.e. 1.9769 MJ/kg of dry rubber. Noticeably, the conventional hot air-drying process consumes more than 95% energy from total fuel consumption of production and higher-grade rubber requires more energy than low grade. Even with recent advancement in drying technology, hot air type trolley dryer remains the main drying machine used by the producer in producing dry rubber. This is probably due to the lack of approach or details study in advanced drying techniques such as hybrid drying technique which has proven with a good track record in the literature (Gowen, Abu-Ghannam and Frias, 2006; Zhang *et al.*, 2006). Therefore, development of revolutionized drying strategy in

crumb rubber processing is paramount especially in improving the drying rate and energy consumption while maintaining the product quality. The application of combined drying technique is envisaged to give both the advantage in rapid drying and energy saving for the drying of crumb rubber.

1.4 Research objectives

The main objectives of this research were:

- To conduct systematic microwave drying studies with continuous power at different levels of heated air temperature, power density and inlet air velocity
- To design and develop a laboratory microwave-assisted drying apparatus
- To determine the drying kinetics of rubber under various operating conditions by using combined drying techniques
- To compute the specific energy consumption and effective diffusion coefficient for respective drying strategies
- To determine the thermal oxidative resistance of microwave dried rubber and compare it with commercial standard
- To measure the effect of varying drying parameters on drying performance of respective drying strategies

1.5 Research scopes

The crumb rubber was dried by hot air, vacuum, microwave and microwave assisted drying. Fixed bed drying method was applied to various drying strategies comprises a single stage, two- stage and combined stages. The drying rate and drying

characteristics of crumb rubber were determined by moisture content profile. Hot air-drying (100°C to 120°C) was used as reference method. The effective moisture diffusivity of individual drying strategy in different parameters was determined from product moisture based on Fick's second law of diffusion. Ten of different thin layer models were introduced to model the characteristic of drying of crumb natural rubber. An optimal thin layer model was selected to represent majority drying behaviour of crumb rubber in different drying strategies.

Specific energy consumption (SEC) was computed to evaluate and compare the energy usage in the effect of different drying parameters. Comparisons were made against the laboratory hot air dying method and the actual energy involved in the drying process at production line are not considered in this study. On the other hand, the colour change of dried rubber was related to the standard and fresh product. Whereas thermal oxidation of dried rubber in terms of activation energy and degradation temperature recorded from a group of drying strategies were compared with the hot air-drying method. Rheological investigation in dried rubber sample was carried out between a group of drying strategies to compare the performance indices in accordance with SMR 20 standard. Additionally, the springiness of dried rubber obtained in different drying conditions were examined respectively and compared with the hot air-dried sample. Likewise, dried rubber sample was subjected to Scanning Electron Microscopy (SEM) to compare the sample topography obtained from hot air and microwave drying method which can be used to describe the rate of water diffusion. Influence of varying the drying parameter in affecting drying performance of hot air microwave assisted drying was evaluated

and analyses through multiple regression analysis methods where statistical models were produced.

1.6 Significance of the research

In this research, the current issue of uneven drying of crumb natural rubber could be minimized. Additionally, crumb natural rubber can be dried at the shortest period and at minimum energy spend while maintaining the quality of dried rubber. The research findings shall be indispensable in assessing the drying kinetics and energy involved in crumb rubber processed by both conventional and combined drying methods. Furthermore, the focus of study has been identified as a prime yet still open for research in view of the need to better understand of drying mechanism and physicochemical properties of crumb rubber in various drying conditions. The findings will supplement the knowledge on crumb rubber drying process which is beneficial to raw rubber processing industries in having economically feasible drying option to be applied to the existing plants. On the other hand, studies on the hybrid drying of raw rubber can provide a better insight of the drying mechanism and quality control to other biomaterials too.

CHAPTER 2

LITERATURE REVIEW

2.1 Overview of NR industry in Malaysia

NR is one of the essential commodity of Malaysia with a contribution of annual export value at RM6.24 billion and its production volume was 722,122 tons in 2015 (Malaysian Rubber Board, 2016). According to Department of Statistics Malaysia, a large portion of the rubber planting was owned by the smallholders totalling 992,510 hectares and the balance 86,120 hectares owned by estates (Malaysian Rubber Board, 2016). With the title of world's fifth largest producer of NR and world's eighth largest consumer of rubber in 2016, Malaysia remains a global player in rubber, supplying the global market with a varied range of rubber products (Malaysian Rubber Export Promotion Council, 2016). There are various grades of NR's products such as Standard Malaysian Rubber (SMR), latex and compounded rubber are exported to other destinations especially to China. The NR usage would be supported by ongoing expansion in the global vehicle fleet, low prevailing prices as well as the increase in world population. On the other hand, NR is domestically consumed for rubber gloves, tyres and tube, rubber thread and more than 7 out of 10 domestic consumption of NR was rubber gloves industry (Department of

Statistics, 2017). NR is extensively applied in the societal field and industrial due to the high and reversible deformability. NR or general termed as elastomer consists of very high molecular mass molecules that can be cross-linked together to form a network. Due to this unique networking, the molecular segments between cross-links are randomly coiled when rubber is unstressed. In contrast, when a tensile stress is applied, deformation is principally achieved by localised motions of the molecular segments that take on new shapes. This unique characteristic has associated with great elongation strength, excellent building tack, low rolling resistance, low heat build-up and good low-temperature performance which are essential characteristics of dynamic working environment and application. Besides, the ability of NR to dissipate heat that makes tyres containing it so much safer than tyres made entirely with synthetic rubber and the bulk of dry NR produced is thus used in high-performance tyres for aircraft and heavy vehicle industry (Cecil. and Mitchell, 2005).

In general, the NR coagulated from latex can be prepared in crumb (granulated) form or machining as sheet whilst the remains are manufactured as crepe or sold as centrifuged rubber (Cousin, Benet and Auria, 1993). Unlike sheet rubber production, TSR commonly processed with automated machinery which is larger in production scale. Advantages claimed for the TSR over the conventional sheet and crepe grades of rubber are listed as following (Associated Speciality Rubber (Pvt)Ltd, 2012):

- Available in a limited of well-defined grades according to consumer's request

- Mistakes can be avoided through an assessment of the actual content of foreign and volatile matters and thus allow the consumer to decide the actual worth of the material
- Being marketed as compact and polyethene wrapped bales (33.33kg per bale), degradation of the rubber on storage, handling and transportation can be prevented
- Being in standard size and compact, considerable savings can be made in transport through mechanized handling and containerization

Meanwhile, TSR is regarded as a modern form of NR (Associated Speciality Rubber (Pvt)Ltd, 2012), specifically designed to meet the requirement of the rubber consumers; ensure the value of money payable by the consumers and to confirm better uniformity and consistency in the quality of NR. In Malaysia, TSR is categorized in accordance to the Standard Malaysian Rubber (SMR) specification scheme that must be abided by rubber manufacturer and fellow consumer in Malaysia as well as other countries for rubber trading purposes. As shown in Table 2.1, a higher grade of SMR's rubber has both excellent processing and easier mixing characteristics that can considerably minimize the mixing and mastication duration as compared to the lower grade of RSS. The SMR 20 is set as a control sample for entire experimental work in this research study to serve the huge demand for standard block rubber in the tyre industry. In the following section, inclusive review on crumb rubber drying mechanism, quality assessment, advanced drying technology, diverse drying techniques, several of existing NR drying system reported beforehand was presented and discussed.

Table 2.1: Standard Malaysian Rubber (SMR) Specifications Scheme [a] (Malaysian Rubber Board, 1991)

Source of material	**Latex concentrate**			**Sheet material**	**Blend**	**Field-grade material**	
Parameter	**SMR CV 60**	**SMR CV 50**	**SMR L**	**SMR 5** [b]	**SMR GP**	**SMR 10**	**SMR20**
Dirt retained on 44µ aperture (max, %wt)	0.02	0.02	0.02	0.05	0.08	0.08	0.16
Ash content, (max, wt %)	0.50	0.50	0.50	0.60	0.75	0.75	1.00
Nitrogen, (max, wt %)	0.60	0.60	0.60	0.60	0.60	0.60	0.60
Volatile matter, (max, wt %)	0.80	0.80	0.80	0.80	0.80	0.80	0.80
Wallace rapid plasticity, P_0, (min)	-	-	35	30	-	30	30
Plasticity retention index (PRI), (min, %)	60	60	60	60	50	50	40
Mooney viscosity, ML (1'+4"), 100°C	60 (±5)	50(±5)	-	-	65(±7)	c	c
Colour coding marker	Black	Black	Light green	Light green	Blue	Brown [d]	Red [e]
Plastic strip colour	Orange	Orange	Transparent	Opaque white	Opaque white	Opaque white	Opaque white

[a] Mandatory from Oct.1, 1991

[b] Two sub-grades of SMR 5 are SMR 5RSS, which are prepared by direct baling of the ribbed smoked sheet and air-dried sheet (ADS), respectively.

[c] The Mooney viscosities of SMR 10 CV and SMR 20 CV are, at present, not of specification status. They are, however, controlled at the product end to 60 (±7, -5) for SMR 10CV and 65 (+7, -5) for SMR 20CV.

[d] Magenta for SMR 10 CV; [e] Yellow for SMR 20CV

2.2 Fundamental to drying process

Drying is commonly employed in the process of a production to prevent the post-harvest degradation of foodstuff, seafood, fruits and vegetables. Most of bio-material possess high moisture content require pre-treatment and drying process to preserve the inherent quality. Chua and Chou (2003) emphasized the importance of post-harvest methods i.e. drying in converting perishable products into more stabilized products that can be kept under a minimally controlled environment for an extended period. Indeed, the rate of moisture movement is much depending on material characteristics. The drying material can be categorized into two types i.e. hygroscopic and non-hygroscopic. The material that can be dried to zero moisture level is known as non-hygroscopic whereas the hygroscopic material such as food products will often have a residual moisture content. Figure 2.1 displayed the moisture that is categorized into bound moisture or unbound moisture which leftovers in the material. A hygroscopic material will either absorb or desorbs moisture when exposed to air and it depends on the relative humidity of the air. A non-hygroscopic material such as NR, however, is poor in thermal conductivity and may lead to condensation on the surface of the capillary water which in turn leads to rot or mould (Transport Information Service, 2002). The equilibrium moisture content (EMC) of the sample is reached when the vapour pressure of water in the material becomes equal to the partial pressure of water in the surrounding air. The EMC is important in the drying since this is the minimum moisture to which the material or product can be dried under a given set of drying condition (Garg and Prakash, 2000).

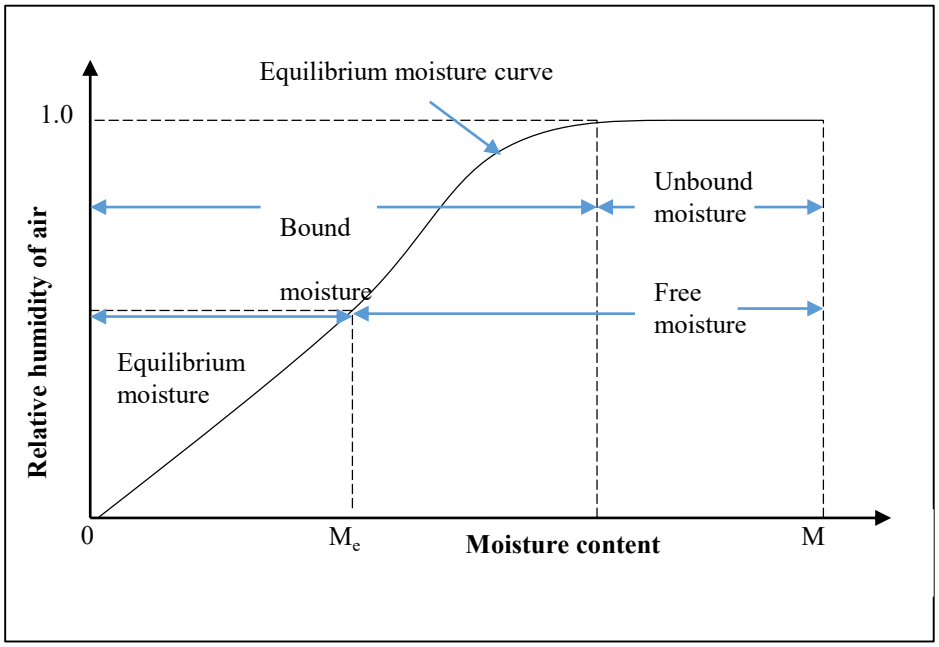


Figure 2.1: Moisture in the drying material (Garg and Prakash, 2000)

A typical drying curve as illustrated in Figure 2.2 can be used to describe the drying characteristic of the dried sample. The drying curve has been divided into different stages including an initial transient stage (from A to B), CRP (from B to C) and FRP (from C to D and E). The differences between CRP and FRP period can be verified through the factor that controls the drying process. For example, the CRP is controlled by external condition i.e. gas film mass transfer whereas active diffusion control is dominated during FRP. The point C or critical moisture content is defined as soon as the drying rate starts to decrease. The drying rate will continuously decrease and cease at point E when the moisture in the product is in the equilibrium state with the surrounding.

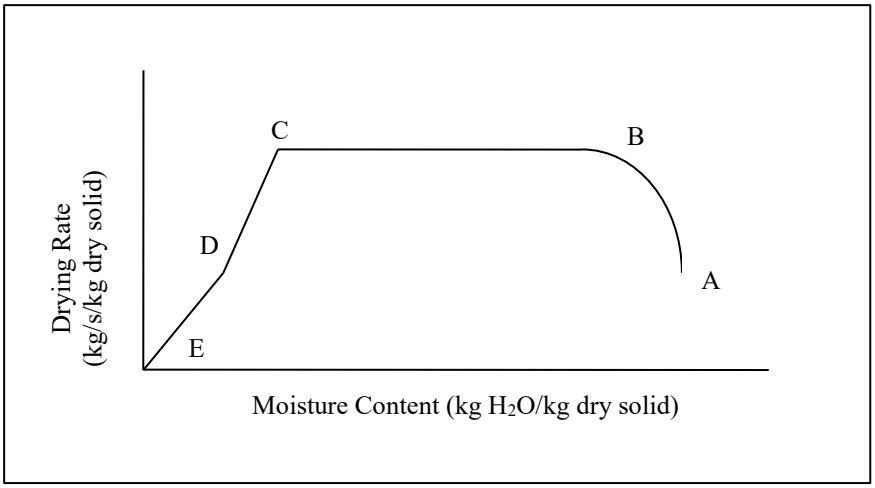


Figure 2.2: General curve for drying rate vs moisture content (Ameen and Bari, 2004)

Study of drying characteristic of a product is deemed important such that specific drying strategy can be incorporated into a certain stage to improve the drying process. It is also noteworthy to mention that drying behaviours of different conditions in the same strategy can be summarized by characteristics of drying curve (CDC) in a single normalized drying rate curve based on van Meel's concept. van Meel (1958) presumed a constant critical moisture content and a modification of the coordinates to organise all experimental drying rate on a single basic curve. Otherwise, a dimensionless drying rate based on pseudo-constant rate was suggested by Keey and Suzuki (1974). Several authors (Belghit, Kouhila and Boutaleb, 2000; Bellagha *et al.*, 2002; Kouhila *et al.*, 2002) have reported the CDC through the fitting of both moisture ratio and normalized drying rate based on the van Meel transformation. However, no application of these models has been found in the literature concerning rubber drying. In this research, the dimensionless drying rate versus a dimensionless moisture content data was regressed to obtain an empirical equation that can be used to describe the CDC of crumb rubber in different drying methods.

2.3 Drying mechanisms of crumb natural rubber

In the block rubber processing, the raw NR is subjected to few times of washing and cleaning process and therefore drying is vital to eliminate the excess water and volatile content from the rubber through the extensive drying process. It is to minimize product degradation upon chemical reaction especially longer storage period is expected. Prior to drying, crumb rubber or term as a granulated form of NR is produced from field coagula through the process of cleaning, multi-stages of size reductions by mechanical means through a combination of pelletizing, shredding and creping process. Crumb rubber is comminuted to a diameter size of 4mm and exhibits as a discrete form of wet rubber but returns to its natural tack along with the drying process. Conventional hot air drying of crumb rubber is generally conducted at temperature 100 ° C or slightly higher to produce dried rubber for SMR usage.

Few mechanisms have governed the drying of crumb rubber in different conditions. At high water content, it is seen a spontaneous oozing of the serum and drips off free water from the wet rubber which known as syneresis. As drying initiated, interstitial water started evaporated from the rubber surface. In low water content, diffusion of water has governed the moisture transferred from inside the crumb particles and its subsequent evaporation (Cecil. and Mitchell, 2005).Cousin, Benet and Auria (1993) claims the drying mechanisms of crumb rubber affected by both factors includes temperature and relative humidity (RH) of both the ambient air and the rubber. The initial transient stage describes rubber layers were saturated with water and temperatures of the ambient air and the rubber granules equalled to wet bulb temperature. Meantime, constant drying

rate present in the second stage of drying in which RH of the air and the moisture content of rubber sharply reduced. This was due to inter-granular heat exchange between phases when wet granulated rubber brought to contact with hot carrier fluid whereby the granular temperature is gradually increased and finally exceed the vaporization temperature which facilitates water vapour evaporates from the surface of the granular. Meanwhile, the air in the dryer is saturated with vapour and forms a thick film around the rubber surface. The process of convective heat and mass transfer shapes the constant rate drying period (CRP) and is dominated by evaporation of water from the surface meanwhile the rate of evaporation is proportional to the surface area and the rate of heat transfer (Rahman, 1985). The complex structure of the rubber and resulting in different binding forces includes bound water and surface water causing the complexity of the drying of natural rubber. Breymayer *et al.* (1993) claimed the removal of surface water is dependent on temperature and air velocity during the CRP. Meanwhile, the subsequent falling rate drying period (FRP) is governed by temperature. As the drying progress, internal moisture diffuses from the centre of the particle to the surface to reclaim for the moisture loss at that region. In another word, the gradient of vapour pressure creates gaseous diffusion towards the surface and evaporation generated inside the porous medium. This capillary movement in capillary-porous biological materials, suggests the drying exposure causes the loss of volatile compounds (Akbarian, Ghasemkhani and Moayedi, 2014).In the mid of drying, the outer layer of crumb might become less permeable and restrict any further migration of water from the inner to the surface due to the case hardening. Meanwhile, the drying rate is decreased in corresponding to a reduction of moisture content. To resolve the issue, a higher drying temperature

is introduced into the system to provide excessive heat energy for internal water to overcome the barrier and transferred to the surface (Cecil. and Mitchell, 2005).

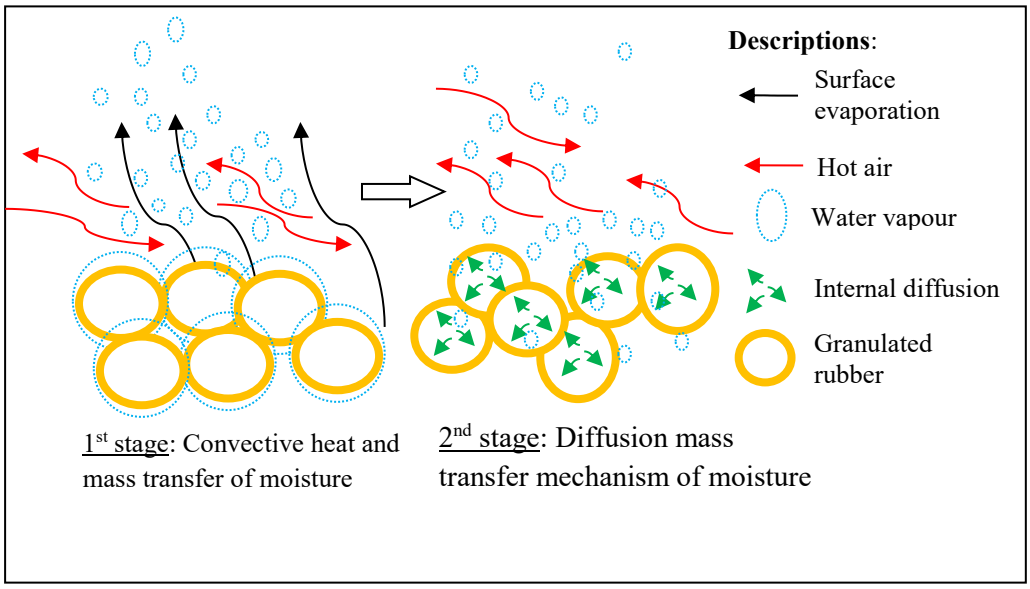


Figure 2.3: Heat and mass transfer at granulated rubber

Besides, sample temperature is almost uniform during FRP due to the additional energy transport by the vapour back diffusion and re-condensation as claimed by Schlunder (2004). Figure 2.3 illustrates the moisture diffusion mechanism inside the granulated rubber which is governed by FRP instead of CRP (Jamaleddine.T.J. and Ray.B.M., 2011). Further evidence was found in Rahman's study (1985) who suggested that overall drying time of crumb rubber is considerably dominated by the diffusion process of water. On the contrary, Azam Ali (2008) proposed that the rate of drying was also dominated by chemical composition and physical structure of the food during the falling rate period.

Another research on drying kinetics of STR 20 by Suwannawong (2004) demonstrated the thin layer drying rate was actually affected by the initial moisture content as well as the hot air temperature which is proportional to the

drying rate. John.C. (2005) suggested the first stage of drying is to be controlled at a relatively low temperature and at high humidity to prevent the formation of a hard skin on the crumb surface which could restrict the permeable of water diffuse. Meanwhile, relatively high temperature and low humidity are suitable for falling rate period of drying.

2.4 Drying techniques

2.4.1 Smokehouse and solar drying of natural rubber sheets

Generally, the raw natural rubber is prepared either in the form of crumb (in large scale production) or sheet form (in small and medium scale production). In smallholder production, the latex after tapping from the farm is coagulated and milled into sheets with vary in thickness, density, moisture and contaminated with foreign residue. Without using dryer or smokehouse, rubber sheets can be dried in the shade or expose to sunlight. Both of methods are indeed slow drying process and dried rubber sheet properties are heavily depending on weather condition. According to Breymayer *et al.* (1993), rubber sheet dried in shade required 10 to 12 days to dry, whereas solar has four to five days shorter drying time than in shade. A humid weather will lead to the growth of fungi whereas overexposure to sunlight will causing the polymer chains scission and deterioration of natural antioxidants (Breymayer *et al.*, 1993). For small to medium production, natural rubber in the form of the sheet is commonly dried in an air-drying tunnel or smokehouse, whereby smoke is introduced to latter case (to produce ribbed smoked sheet) although both methods have similar preparation (Pratoto, Daguenet and Zeghmati, 1997).For information, rubberwood is the prime source of fuel to feed the smokehouse and maintain a

temperature between 45 °C to 60 °C in order to dry and prolong the shelf life of rubber sheets (Breymayer *et al.*, 1993). However, the rubber wood price is getting higher as it is now preferably as raw material for furniture, woodchip, charcoal etc. (Breymayer *et al.*, 1993; Pratoto, Daguenet and Zeghmati, 1997).Moreover, Tekasakul *et al.* (2015) claimed that non-uniform drying of the rubber sheets consequences in a low efficiency of fuel usage. In fact, approximately 1 to 1.5 tons of rubberwood is needed to produce one ton of dried rubber sheets (Breymayer *et al.*, 1993; Tekasakul and Promtong, 2008).To cope with this issue, lots of researchers have proposed several efforts to enhance the drying efficiency, for example, the utilization of solar energy in both direct and indirect, respectively (Dejchanchaiwong *et al.*, 2011; Janjai *et al.*, 2015). For example, a solar dryer integrated with biomass furnace for rubber sheet drying was proposed by Rama Rao (1984). N Radhakrishnan Nair L Verghese, N.M Mathew (1988) designed and fabricated a solar cum smoke drier to reduce the drying time from 8-12 days to 5 days with around 70% saving of firewood in comparison to conventional smoke-drying. Similarly, Breymayer *et al.*(1993) developed solar heated rubber sheet dryer consists of a flat plate solar air heater connected to a drying chamber. A biomass furnace was integrated into the dryer to minimize the impact of ambient or weather conditions on the drying system. A recent study by (Dejchanchaiwong *et al.*, 2016) claimed that the performance of the mixed mode dryer is superior to the indirect solar dryer and thus recommend the use of mixed mode dryer for smallholders in the drying of the natural rubber sheet. Although the drying of rubber sheet by utilizing solar energy seems promising, however, the solar dryer has not been implemented commercially.

2.4.2 Hot air drying for crumb natural rubber

For large scale production, the drying process for crumb natural rubber typically involves more sophisticated machinery consists of both mechanical dewatering and hot air trolley dryer. Firstly, coagulated latex which contains water and volatile compositions undergoes the first stage of dewatering process through vibration screening to rinse off free water accumulated during the cleaning process. Thereafter, coagulated latex is continuously fed to the second stage of dehydration machines such as crepe roller, shredder, pelletizer or hammer milling in which the cutting, pressing and mixing process of the coagulated occurred simultaneously along the dewatering system. Free water removal in the second stage is basically governed by the pressure gradient build up between the latex granule during the mixing, pressing and squeezing process. The involvement of mechanical dewatering prior to drying reduce the water content down to approximate of 85% of dry rubber content (DRC),while further thermal drying at between 100 °C to 130 °C is particularly important to achieve a minimum DRC of 99.2% for dried rubber in accordance to SMR which being introduced since 1965 (Malaysian Rubber Board, 1991). It is well known that volatile matter levels greater than 0.6% can promote the growth of mould and lead to malodour (Rama Rao, 1984).

Drying of crumb rubber is an energy-intensive process whereby hot air (HA) as the typical drying medium is applied to promote the convective heat by removing volatile content and moisture from the wet solid. Heat needed to increase HA temperature may be supplied in the form of direct firing with fuel combustion or indirectly by using a heat exchanger. HA circulated type of dryer is mostly

employed in the drying of a deep bed of rubber. These including chamber dryer with trolleys, a single layer or multilayer tray dryer and apron dryer while they vary in construction, handling and heating system (Sung, Fah and Beng, 1973). Trolley dryer is broadly used in crumb rubber drying being either batch, semi-continuous or continuous. The dryer includes a tunnel through which boxes are pushed or dragged and through which HA is circulated as illustrated in Figure 2.4 (Cecil. and Mitchell, 2005). In comparison to air-dried techniques in ADS, rubber dried faster in through circulation drying process because of the high surface area per weight ratio of granulated rubber combined with high-velocity airflow over each partition.

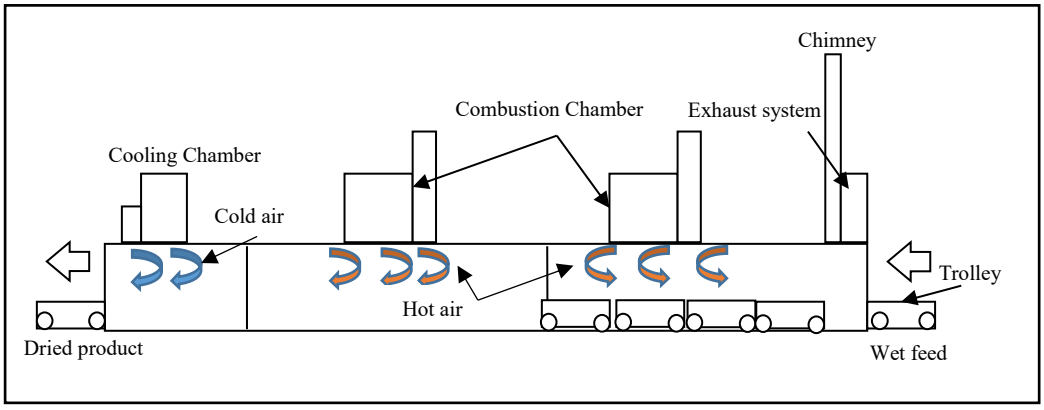


Figure 2.4: Trolley type hot air dryer

The crumb is loaded into the multi-box trolleys which pass through the tunnel one after the other, exposing the wet crumb to different drying zones. Each compartment has perforated bottoms to allow water drip off and passage for hot air circulation in the inter-granular spaces of crumb rubber layers with approximates 30 to 50 cm in height (Naon, Berthomieu, Benet, Saix, *et al.*, 1995). Indeed, John.C. (2005) recommended a practical and economical way of drying crumb rubber in a bed depth of 25-30cm with a circulating hot air at 80°C to 100°C in a forced convection motion from top to downwards through the bed at

an airspeed of 15 to 30 meters per minute. Also, openings with adjustable doors are fitted in the upper and lower parts of the dryer to direct the direction of the airflow reaching the wet product and to regulate its velocity (Berthomieu *et al.*, 1997). Cooling fans are often installed before exits and used to blow ambient air onto the dried crumb to cool it down to a temperature below 60° C to meet packing specification. Table 2.2 summarises the recommended drying temperature at different drying stage for crumb rubber. It is worth to mention that the latex crumb dried at lower temperatures than the field grade rubber. This is mainly due to the structure of latex crumb is more porous and consistent than a crumb of lower grade and therefore, rapid and even drying can be achieved for latex grade.

Table 2.2: Recommended temperature for drying crumb rubber (Cecil. and Mitchell, 2005)

	Temperature range (° C)	
Source of rubber\ Stage of drying	**Latex grade**	**Field grade**
First	100 -105	105-110
Second	105-110	110-120

A convective HA dryer unit operates with an upward air flux was applied by Cousin, Benet and Auria (1993) in experimental drying study of a thick layer of natural crumb rubber. The dryer was restricted from air recycling to ensure at constant temperature and humidity where the evolution of temperature and relative humidity of inter-granular air and water content of crumb were desired. Khongchana *et al.* (2007) claimed that 40 minutes of drying time at 130 °C followed by 180 minutes of drying time at a lower temperature of 110 °C can yield a better quality of dried rubber as compared to other drying conditions. Practically, fresh rubber has a high moisture content in comparison to that of

mature rubber which longer drying time is essential. For instance, fresh crumb rubber with initial moisture content from 40% to 55% dry basis consumed 220 minutes of total drying time (Tirawanichakul and Tirawanichakul, 2008). Wei Y.S. *et al.* (1981) had successfully introduced the conveyor type of HA dryer for continuously drying of standard rubber. Unlike conventional trolley dryer, this invention claims a reduction of drying time from 4 to 8 hours to about 2 hours was achieved without major white spots being observed in the drying process. The study concluded that the maximum temperature for drying latex grade crumbs is 125°C and the ideal bed thickness is 25-28 cm whereas for field coagulum grade crumbs, these will be 115°C and 17-20 cm, respectively (Yushan *et al.*, 1985). By considering different drying parameter e.g. bedding depth of crumb rubber, aeration volume, drying time and temperature, Wei Y.S. *et al.* (1989) revealed that a combination of drying temperature of 125°C and air velocity of 5.3 to 5.4 m^3/s was the optimum conditions in thick rubber drying without contributing much to the oxidation or over cure issue.

Conventional drying methods are indeed reliable and widely adopted in raw rubber processing industry; however, it is rather unfavourable due to its time-consuming process especially in the FRP (Taskini, 2007) which in turn leads to high operating cost and energy consumption. Again, as the oil price hikes higher, the drying cost is increased considerably thereby eroding the profitability of these drying units which probably accounted for more than 50% of total manufacturing cost. In turn, domestic raw rubber manufacturer suffers from this change and most of them are on the verge of closure. Thus, an alternative drying method is imperative to turn around the situation of crumb rubber drying through

the introduction of cheaper energy options to reduce the overall manufacturing cost and minimize off-specification product whilst maintain the production throughput rate.

2.4.3 Vacuum drying

When drying of liquid water present within the material, higher temperature above boiling point is required to generate a sum of the partial pressure of air and vapour of which surpass the external pressure. The existence of pressure gradient drives the internal moisture towards the exchange surfaces. High degree of air temperature can be achieved either through convective drying, contact drying or with an aid of electromagnetic sources. Alternatively, lowering of external pressure in vacuum drying (VD) permits a reduction in water boiling point. As a consequent, water boiled and evaporated at lower temperatures than under atmospheric conditions and therefore the product can be protected from overheating, which in turn, limits quality degradation (C.Pearson, 1920; Yongsawatdigul and Gunasekaran, 1996). Indeed, oxidation reactions are minimized because drying is conducted in the absence of air (Gunasekaran, 1990).In VD, heat is transferred to the product mainly by conduction and sometimes by radiation. Water circulation throughout the wet product intensifies at lower pressures causes greater internal mass transfer under vacuum (Ratti, 2008). However, the absence of drying medium in the VD chamber hinder convective heat transfer but enhances mass transfer at low temperatures.

In fact, VD has been suggested by India Rubber World since in 1905 (C.Pearson, 1920), where the rubber dried in a vacuum shelf dryer is accomplished rapidly;

when low temperature is necessary, a great saving in fuel, space and labour (Wright.H., 1912). However, the effectiveness in VD on crumb rubber much depends on the level of maturity of the vacuum technology being employed by respective processing plant. Saberian, Amooi and Hamidi-Esfahani (2014) observed that the rate of vacuum drying of loquat at 80° C was nearly 1.5 times faster than 70 ° C and 2.5 times faster than 60° C denoted the increment of temperature proportionally improve the drying rate. Another study by Yamsaengsung and Sattho (2008) on superheated steam vacuum drying on rubberwood recorded a reduction of total drying time from 168 hours to less than 20 hours (moisture content reduction from 0.80 to 0.06 dry basis) and physical properties were significantly improved. Unfortunately, the rate at which material temperature can be raised in a vacuum dryer is limited due to the indirect heat drying technique is constraints by the effective surface area available for heat transfer (Warren and Paul, 2004). Therefore use of internal heating media by incorporating radiant heat input, internal heating media or a combined mode of heat transfer in VD such as microwave vacuum drying could effectively diminish the low heat and mass transfer problems occurs and enhancing the drying kinetics in the drying of the wet product (Bondaruk, Markowski and Blaszczak, 2007; Hu *et al.*, 2007; Setiady *et al.*, 2007).

2.4.4 Microwave drying

Undeniably, convective HA drying is the simplest and mature drying technology widely available in the market, however, its effectiveness is underperforming especially when drying materials are poor in thermal conductivity, or high internal resistance to moisture transfer, while minor heat energy is continuously

dissipated to the surrounding during the heating process. Alternatively, electrical volumetric heating specifically on microwave (MW) heating is gaining attention other than HA drying due to its attractive features in effective moisture removal, short processing times and low drying temperature as a factor to secure the products quality to an extent (Cuccurullo, Giodarno and Albanese, 2012). Chen *et al.* (2013) claimed that there is growing application of MW in drying process as well as in other industries with different process material. MW is a form of electromagnetic radiation in the frequency range of 3,000 MHz to 30,000MHz with a commonly used frequency at 2450MHz (Nave, 2012). While 2450MHz is commonly applied in the domestic oven with a wavelength of 12.2 cm; industrial MW ovens often employed 915MHz with wavelength 32.8 cm in which the depth of penetration of microwave energy at 915MHz is about three times as great as that at 2,450MHz (Industrial Microwave Systems, 2012). Within the available frequencies that are not set aside for communication, 2450 MHz was chosen in this study because 915MHz wasn't a band available worldwide, as 2450 MHz was.

The MW heating process involves the transformation of electrical energy into thermal heat either to increase the temperature of wet product -for drying purpose or to a critical level for melting purposes. Nonetheless, it is commonly known that heat dissipated from the MW energy depends on various parameters, for instance, MW power level, MW field distribution, the location of feed ports, dielectric properties and structure of the sample. By comparing to HA drying, MW drying does not solely rely on the conduction of heat through solid but rather through self-excited molecular activity, particularly among polar

molecules due to the absorption of MW energy. Indeed, MW heating depends on the interaction between polar groups in molecules of nonconductive materials and the alternating electric field of oscillating magnetic field. Some example of molecules such as water, fat and sugar can be absorbed by MW and heat up (Wray and Ramaswamy, 2015). During the volumetric heating process, polarity orientation of polar molecule i.e. water changes in line with a variation of external electromagnetic field generate internal resonance effects and creates the collision between water molecules as well as molecular friction, which, in turn, transformed into thermal energy to raise the temperature of the rubber (Chen, Wang, *et al.*, 2012).On the other hand, the evaporative cooling effect on the surface of material due to rapid water evaporation resulted in pressure gradients which facilitate the moisture diffusion and heat transfer expand outward in the same direction. This has greatly enhanced the migration of volatile matter during drying which performed better than conventional HA drying. The evaluation of absorption efficiency by a dielectric material when it is placed in a high-frequency electric field which also termed as local volumetric heat generation in the product itself can be represented by Equation 2.1:

$$P = \sigma|E|^2 = 2\pi f \varepsilon_0 \epsilon^{'} (tan\delta)|E|^2 \quad (2.1)$$

Where P, refers to MW energy absorbed (W/m^3) per unit volume of the sample at any instant of time; σ is effective conductivity; f is frequency; ε_0 is permittivity of free space (8.86 x 10^{-12} F/m); ε' is relative dielectric constant;$|E|$ is the magnitude of internal electric field; $tan\delta$ is the loss tangent coefficient. The equation has clearly derived the relationship between MW energy absorption which is proportional to the frequency of the applied electric field,

dielectric loss factor as well as to the square of local electric field (Rattanadecho *et al.*, 2007). For a drying process, the MW energy can be applied either in (i) the beginning, (ii)drying rate started to fall (iii) the final stage of drying. In the first case, product's temperature quickly heated up after receiving MW energy which in turn creates puffing effect on the porous structure in the sample that facilitates the moisture diffusion to the surface (Zhang *et al.*, 2006). For the second case, the MW energy heats the trapped moisture to evaporation temperature and helps its movement to the outer layer sample. The last case describes the use of MW energy in removing bound water and to overcome the effects of product shrinkage which can heavily slow down the drying rate (Andres, Bilbao and Fito, 2004; Zhang *et al.*, 2006).

2.4.4.1 Application of MW in rubber drying and its effect on rubber quality

A number of studies have been conducted to investigate MW drying process either using a domestic or household MW oven, a single or multimode cavity, as well as few numbers of the belt, typed MW drier (Rattanadecho *et al.*, 2007; Vongpradubchai and Rattanadecho, 2009; Chandrasekaran, Ramanathan and Basak, 2013). The recent development of inexpensive and reliable MW sources has been increasing attraction in the application of rubber drying (Rattanadecho *et al.*, 2007; Lin *et al.*, 2008; Wang *et al.*, 2010). Wang *et al.* (2010) recommended an initial moisture content of 10% to 20% for MW drying to enhance the rate of drying of NR as well as quality control. Besides, crumb rubber had been tested in continuous microwave belt drier (Rattanadecho *et al.*, 2007). In the experiment setup, MW powers were varied from 2.4kW to 11.2kW

by means of compressed air-cooled magnetrons of 800W each. The experiment confirmed that reduction in drying time with an increasing number of the magnetron with the same thickness of the sample was mainly due to an increase of interval of heating. Chen *et al.* (2012)discovered remarkable reduction of drying time by 93.59% to 13.47 minutes (as shown in Figure 2.5)in MW dried rubber and product quality were higher than those of rubber dried by HA. Two essential criteria of rubber process ability i.e. Wallace Plasticity Index (P_0) and Plasticity Retention Index (PRI) was found superior in the rubber dried with MW than that of HA dried rubber as tabulated in Table 2.3.

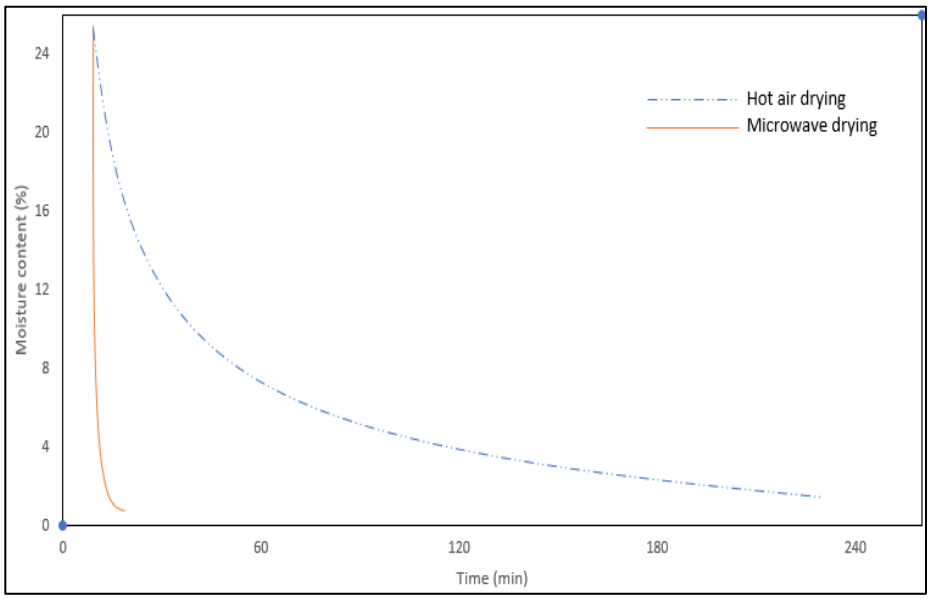


Figure 2.5: Drying kinetics of rubber between MW and HA dried rubber (Chen, Wang, *et al.*, 2012)

Furthermore, Lin *et al.* (2008) showed that MW drying in NR had an average dehydration efficiency of 9.22%, which was 4.7 times greater than the far infrared and 17.7 times higher than HA.

Table 2.3: Properties of natural rubber samples obtained from HA & MW drying (Chen, Wang, *et al.*, 2012)

	Technical Parameters	
Drying Techniques	P_0	PRI
HA	32.2	79.80
MW	44.2	88.20

2.4.4.2 Advantages and Limitations of MW Drying

Chou and Chua (2001) commented on few advantages of MW drying including enhanced diffusion of heat and mass, increased of drying rates without increased surface temperatures and thus better product quality due to lesser oxidation by heat. Besides, the MW drying system is constructed in such a way that heat loss is minimized and to maximize the absorption of MW energy by dielectric materials. This concentrated heating within the sample rather than the cavity when applying the MW energy can reduce the drying temperature, reduce drying defects and shortens drying times (Jindarat *et al.*, 2011). This was proven by Tulasidas, Raghavan and Mujumdar (1995) where MW dried raisin was found superior to HA dried samples in terms of colour, darkness, stickiness and uniformity. Likewise, desirable colour, shrinkage and rehydration properties of the dried carrot were obtained with MW heating (Wang and Xi, 2005).

Although electrical volumetric drying offers various advantages comprises of drying efficiency and product quality, however, understanding of limitations in MW processing is equally important in the selection of drying technique to

achieve an equilibrium in between cost and quality. The MW dryer has limited commercial production due to expensive operating cost, advanced structure, strict airtightness is required and operating difficulties in the drying process of products (Hai-Ming Yu. and Chun-Cheng Zuo., 2015). Some limitations in MW drying including non-uniformity drying due to uneven distribution of electromagnetic energy in MW cavity and non-uniform moisture distribution in the drying material. Non-uniform or excessive MW energy application in the sample will lead to rapid increase in localized temperature and subsequent destruction of charring, together with under dried samples at other locations (Bouraoui, Richard and Fichtali, 1993). Moreover, thermal runaway by an abrupt temperature rise during MW drying must put into consideration too. This phenomenon occurs when minor deviate from MW power causes the temperature to increase rapidly to the melting point of the sample which resulted in quality deterioration if temperature outranged (Vadivambal and Jayas, 2007). Rahman (1985) dissuaded the MW drying in the crumb rubber drying as it is not cost effective yet too capital intensive from economical points of view. In fact, existing applications of MW drying also limited e.g. rubber curing or vulcanization (Ishii, 1995), which infers that MW drying has not been employed extensively on an industrial scale even though MW drying of rubber have been reported in the literature. Unlike HA drying where the product temperature never exceeds the HA temperature; MW drying in which the final product temperature is hard to control often lead to excessive temperature along the corner or edges of drying products results in scorching and product off flavours especially during the final stage of drying (Chandrasekaran, Ramanathan and Basak, 2013).Li, Wang and Kudra (2011) recommended three aspects to improve heating

uniformity in microwave dryers comprise of I) improving the uniformity of electromagnetic field in microwave cavity by complicating the pattern of the electromagnetic field in MW cavity in terms of nodes and anti-nodes; II)improving the uniformity of MW energy absorption by randomly displacing the material or disordered movement of the particulate material in space during drying; III) combination of another drying method in MW drying. First aspects could be resolved by application of mode stirrer or various MW sources with different frequency on the microwave system (Bows, 1999; Plaza-Gonzalez *et al.*, 2005). Bouraoui, Richard and Durance (1994) suggested the application of lower MW power density either by increasing sample size or reducing MW power, which permits for more even heat distribution and reduced charring issue. Additionally, previous works on microwave-based drying of foods (Gunasekaran, 1990; Yongsawatdigul and Gunasekaran, 1996) reported that continuous application of MW energy does not necessarily quicken the rate of water removal once a critical moisture content has been reached and it been revealed that there are no energy or quality advantages of continuous over pulsed application of MW energy. Hence, the alteration of magnetron duty cycle to limit power applied to the sample is recommended in experiments too.

2.4.5 Combined drying

Thus far, many previous works discussed in Section 2.4.1 to 2.4.4 are referring to single stage drying whereby these drying techniques have its own limitations owing to individual drying mechanism. It is commonly accepted that two-thirds of the drying time may be spent removing the last one-third of the moisture content in the final stage of HA drying which makes the process become energy

extensive due to longer drying times (Nadia R. Pereira., Antonio Marsaioli Jr. and Ahrne, 2007). Likewise, vacuum technology is expensive in terms of operating and capital cost whereas MW heating can cause the poor quality of the dried product due to uneven heating (Zhang *et al.*, 2006). To overcome these problems, more rapid, energy saving, and self-controlled drying methods are necessary (Motevali, Minaei and Khoshtagaza, 2011). The limitation of conventional drying techniques has encouraging scientist and industry player to improvise existing techniques through the incorporation of two or more technology in a system. For instance, an effective measure to alleviate some of the limitations of single MW is to associated MW with HA or VD drying technique (Reyes *et al.*, 2007). Kudra (2006) outlined few common characteristics of contemporary technologies being developed recently comprises of combined modes of heat transfer, a combination of conventional drying techniques, a novel interaction between the gas and solid, new techniques of heat generation or utilization, a combination of mechanical and thermal dewatering, employ superheated steam as a drying means.

The idea of combined drying emerges as an alternative to most of the manufacturer who objectively looking for optimal retention of product quality as well as to save the drying cost. A combination of MW and HA drying generates synergistic effect and creates advantages in regards to processing time and quality (Zhang *et al.*, 2006). Substantial studies were carried out with MW-HA combined drying in many agricultural crops such as potatoes (Khraisheh, McMinn and Magee, 2000),banana (Maskan, 2000),orange slices (Díaz *et al.*, 2003),kiwi fruit (Maskan, 2001), carrot (Prabhanjan, Ramaswamy and

Raghavan, 1995), apple (Andres, Bilbao and Fito, 2004) and mushroom (Funebo and Ohlsson, 1998).Sharma and Prasad (2006) conducted MW-convective drying of garlic and obtained a drying time reduction of 80% with superior quality dried garlic when merging MW at 0.4 W/g to HA at 60°C to 70 °C. Correspondingly, examples of hybrid drying process comprising an initial forced-air convective drying followed by MW-final drying has been reported to promote better product quality with an extensive saving of time and energy (Prabhanjan, Ramaswamy and Raghavan, 1995; Feng and Tang, 1998; Maskan, 2000). Similarly, the application of MW in the falling rate period or at a low moisture content in the drying of apple slices and mushroom by using MW-assisted air drying also reported by Funebo and Ohlsson (1998).

Moreover, in the effort of effectively overcomes uneven distribution of the electromagnetic field, MW-vacuum rotary drum dryer was used to move and mix the chilli particulates to achieve uniform and good quality of the product (Kaensup, Wongwises and Chutima, 2002). Raw potatoes dried by the microwave-vacuum system at 60°C for 150 minutes were produced without scorch (Setiady *et al.*, 2007). Regier *et al.* (2005) extended the MW study by comparing the carotenoid retention in the carrot to the drying technique and concluded that MW vacuum outperforms others in comparison with freeze-drying and convection drying.

Meanwhile, the conveying system of a continuous vacuum-microwave drier was designed in a spiral motion to improves the uniformity of microwave absorption for apple slices (Han *et al.*, 2006). Datta, Geedipalli and Almeida (2005), Sumnu,

Turabi and Oztop, (2005), Chou and Chua (2001) demonstrated MW drying combined with others drying techniques such as forced air and infrared radiation to further enhance the process efficiency and cost reduction. Rakesh *et al.* (2009) claimed that heating profile for MW combination heating is more uniform compared with MW only heating. Indeed, MW drying associated with mechanical dewatering or osmotic dehydration could also reduce energy consumption in drying (Kudra and Mujumdar, 2009). Apparently, by gathering the advantages of MW while minimizes the disadvantages inhered with the application of each technique alone, the MW assisted drying shows promising results in both quality and time-saving.

2.5 Mathematical modelling of thin layer drying curve

Drying is a complicated process involving simultaneous heat and mass transfer. A simple way to model this phenomenon is through mathematical modelling which including theoretical model, semi-theoretical and empirical model. Many semi-empirical and empirical models have been used to model the drying behaviour of the dehydrated product. Among them are models derived from Newton's Law of Cooling and Fick's Second Law of Diffusion (Ertekin and Firat, 2017). As can be seen in Table 2.4, drying behaviours of various drying techniques can be described by ten general models that are among the most acceptable in the literature.

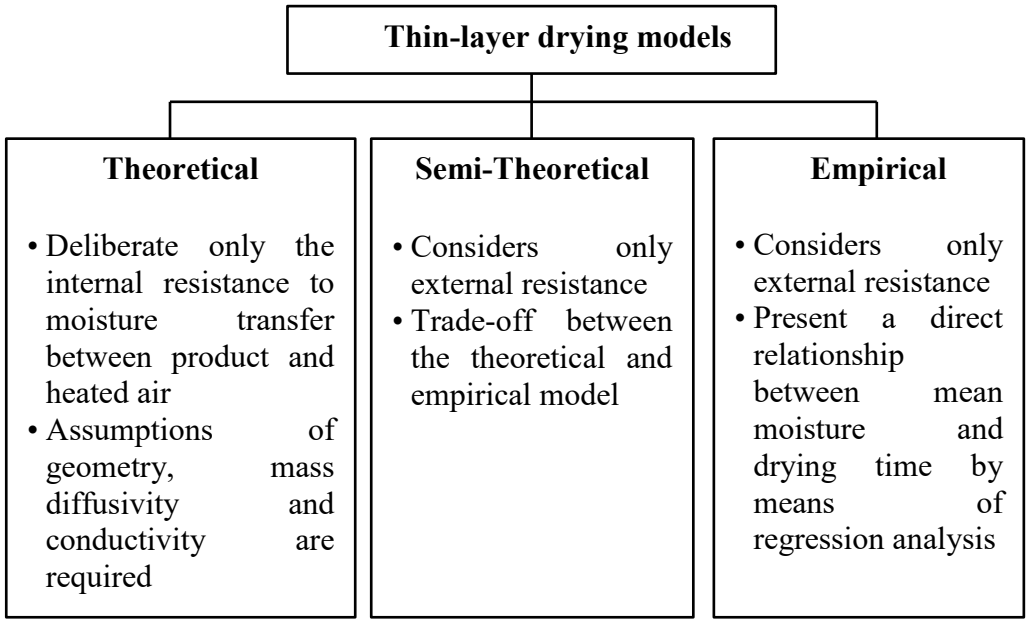


Figure 2.6: Characteristics of thin layer drying models (Wang and Singh, 1978; Demirtas, Ayhan and Kaygusuz, 1998; Midilli, Kucuk and Yapar, 2002; Kumar, Sarkar and Sharma, 2012)

Researchers employed thin layer models to obtain information to interpret the drying behaviour of different food products such as the sultana grapes (Yaldiz, Ertekin and Uzun, 2001), apricot (Toğrul and Pehlivan, 2003), Roselle (Saeed, Sopian and Zainol Abidin, 2008), red pepper (Akpinar, Bicer and Yildiz, 2003), peppermint leaves (Torki-Harchegani *et al.*, 2016) and tomato (Rajkumar *et al.*, 2007). For ADS rubber, various models were proposed to described theirs drying behaviours such as Diffusion Approximation model (Suchonpanit, Tirawanichakul and Tirawanichakul, 2011), Weibull Distribution (Dejchanchaiwong *et al.*, 2011), Modified Henderson and Pabis model (Tasara, Suchonoanit and Tirawanichakul, 2011). Prediction of crumb rubber drying condition via mathematical modelling prepared by Naon *et al.* (1995) found that drying rates changed with drying time, temperature and relative humidity. With

the development of the mathematical model, the thick layer drying rate was predicted by parameter and input collected from thin layer drying. The model is further compared to the semi-automatic trolley dryer used in the industry and results were in good agreement when humidity, temperature, relative humidity and drying energy were considered.

Apart from that, a simulation programme derived from the validated mathematical model was developed by Berthomieu *et al.* (1997) to simulate an industrial convective drier to handle rubber in granule form which also able to predict the properties of the medium at any time and for any point within the layer of rubber. However, for granulated rubber, researchers only focused on convective HA, vacuum heating and obtained the corresponding drying models (Naon, Berthomieu, Benet and Saix, 1995; Tirawanichakul and Tirawanichakul, 2008; Ng *et al.*, 2015). The effects of drying parameters such as drying rate and moisture ratio of MW associated with HA drying in rubber are not researched thoroughly whereby the mathematical model explaining drying characteristics of microwave-convective dried rubber is not found in any of the literature.

Table 2.4: Thin layer drying curve models for variation of moisture ratio (MR) with time (t)

No.	Model name	Model	References
1	Lewis	$MR = e^{-kt}$	(Mujumdar, 2006)
2	Page	$MR = e^{-kt^n}$	(Page, 1949)
3	Henderson and Pabis	$MR = a.e^{-kt}$	(S.M.Henderson and S.Pabis., 1961)
4	Logarithmic	$MR = a.e^{-kt} + c$	(Togrul and Pehlivan, 2002)
5	Two terms	$MR = a.e^{-kt} + c.e^{-gt}$	(Henderson, 1974)
6	Verma	$MR = a.e^{-kt} + (1-a)e^{-gt}$	(Verma *et al.*, 1985)
7	Two-term exponential	$MR = a.e^{-kt} + (1-a)e^{-kbt}$	(Sharaf-Eldeen, Blaisdell and Hamdy, 1980)
8	Modified Henderson and Pabis	$MR = a.e^{-kt} + b.e^{-gt} + c.e^{-ht}$	(Karathanos, 1999)
9	Midilli	$MR = a.e^{-kt^n} + b.t$	(Midilli, Kucuk and Yapar, 2002)
10	Hii and Law	$MR = a.e^{-kt^n} + c.e^{-gt^n}$	(Hii, Law and Cloke, 2009)

Note: *a, b, c, g, h, k* and *n* are empirical constant for respective model

Hence, the present mathematical models are ready to fit the experimental drying data for the prediction of drying characteristic of dried rubber at different hot air temperature, air velocity, pressure and MW power density conditions.

2.5.1 Fick's Law and D_{eff}

Generally, Fick's second law of diffusion is applied to describe the mechanism of moisture transfer during drying by using Equation 2.2.

$$\frac{\partial X}{\partial t} = D_{eff} \nabla^2 X \tag{2.2}$$

On the other hand,D_{eff} of a sample is associated by its inherent mass transport property of moisture which comprises of liquid diffusion, vapour diffusion, molecular diffusion (capillary diffusion) , hydrodynamic flow and other possible mass transport mechanism (Maroulis, Kiranoudis and Marinos-Kouris, 1995).It is also recognised that this propriety is a function of moisture content, temperature and pressure which aid in determining the rate of moisture diffusion (Karel, 1988). General term of the partial differential equation of mass diffusion equation for drying agricultural products with geometrical shape of an infinite slab, an infinite cylinder shape and a spherical shape is derived with following assumptions (Crank, 1975; Vazquez *et al.*, 1999).

I. Moisture transfer in the material is symmetric with respect to the centre
II. Internal heat transfer usually occurs much faster than internal mass transfer which the initial moisture content and temperature are assumed uniform inside the product (Coumans, 2000)
III. Uniform initial moisture distribution
IV. Heat loss is very minimum and can be negligible
V. Moisture movement and heat transfer are unidimensional
VI. Constant moisture diffusivity
VII. Negligible shrinkage

By considering the initial and boundary conditions to Equation 2.2, the general solution for the different shape of the sample with respect to moisture ratio can be obtained as follows:

$$Ø_{slab} = \frac{8}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{(2n_p-1)^2}exp\left[-\frac{\pi^2(2n_p-1)^2}{4L^2}\right]D_{eff}t \quad (2.3)$$

$$Ø_{cylinder} = \sum_{n=1}^{\infty}\frac{1}{\lambda_{n_p}^2}exp\left[-\frac{\lambda_{n_p}^2 D_{eff}t}{R^2}\right] \quad (2.4)$$

$$Ø_{sphere} = \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{{n_p}^2}exp\left[-\frac{\pi^2 n^2 D_{eff}t}{R^2}\right] \quad (2.5)$$

Where Ø is moisture ratio; $8/\pi^2$ is shape factor for infinite slab; *t*, the drying time (s); L, the characteristic half thickness of sample (m); n_p, is a positive integer and D_{eff}, effective diffusion coefficient (m^2/s), λ is empirical constant

For long drying times, only the first term $(n = 1)$ in the series expansion of the above equation can give a good estimate of the solution (Tutuncu and Labuza, 1996). Therefore, Equation 2.3 to 2.5 are transformed in terms of ln (Ø) versus drying time (*t*) and the $D_{eff,}$ was determined from k_0 , the gradient value obtained from the slope method.

2.6 Criteria for selection of drying technique and quality variations

Selection of optimal drying techniques is crucial such that the process must be done both efficiently and economically. In fact, the demand for energy efficient and rapid drying process never ceases by either end user or manufacturer in tandem with concurrent technology and advanced technical knowledge. Besides

the improvement on the existing dryer through part modification; evolution of special drying techniques has been introduced to meet special applications in a specific industrial sector. Among the criteria to fulfil by an efficient dryer shall include enhanced product quality, reduced energy consumption and downtime, higher productivity, easier of control, enhanced flexibility and ability to combine other operations in the same unit. Anyhow, a trade-off is required between capitals and operating cost should be considered as a field to examine the suitability of the new drying system.

2.6.1 Changes in microstructure

Studies revealed that the method of drying has a huge impact on the final microstructure of the samples such as rheology, glass transition and moisture sorption (Jaya, 2009). Aguilera (2005) deduced that the attractive qualities of dried food materials such as colour, texture and flavour are attributed to the microstructure. A comparison of the microstructure of low-fat durian chips that dried via HA drying and superheated steam drying was investigated. Results showed superheated steam dried product had less uniform, fewer but larger pores than HA dried products (Fazaeli, Tahmasebi and Djomeh, 2012). The sharp increase of superheated steam temperature to 100°C caused the water inside durian to boil rapidly and evolution of steam resulting in large pores formed within the material (Jamradloedluk *et al.*, 2007). An investigation by (Bondaruk, Markowski and Blaszczak, 2007) on potato cubes discovered that vacuum-MW processing causes a more serious collapse in the microstructure of dried potato cubes as compared to hot-air drying. This was due to higher vacuum level lowered boiling temperature of the material and lead to melting of starch

granules which weakens the starch-protein matrix. It was suggested that vacuum-MW dried samples are characterized by good reconstitution ability during rehydration.

Likewise, Huang *et al.* (2011) compared the microstructure of four different drying techniques which are microwave freeze drying (MFD), freeze drying (FD), microwave-vacuum drying (MVD) and vacuum drying (VD) on mixed potato with apple chips. MFD chips showed the best quality and these products were preferred by consumers. Arash Tahmasebi. *et al.* (2011) highlighted different pore structures have great influence on moisture transport mechanism and MW absorption characteristics. In terms of drying temperature, higher drying temperature provides a substantial amount of heat energy which leads to a more violent evaporation of water as a result of a rapid collision between water molecules in facilitating the moisture diffusion process. This has raised the hydrostatic pressure gradients of moisture between internal and outer of the dried specimens and causes the formation of large voids, resulting in the crispy product (Fazaeli, Tahmasebi and Djomeh, 2012). The effect of drying temperature had been discovered in the study of tomato dried with microwave assisted drying method where a significant level of Scanning Electron Microscopy, SEM image disruption was observed in proportion with increasing of drying temperature or MW power level (Izli and Isik, 2015).

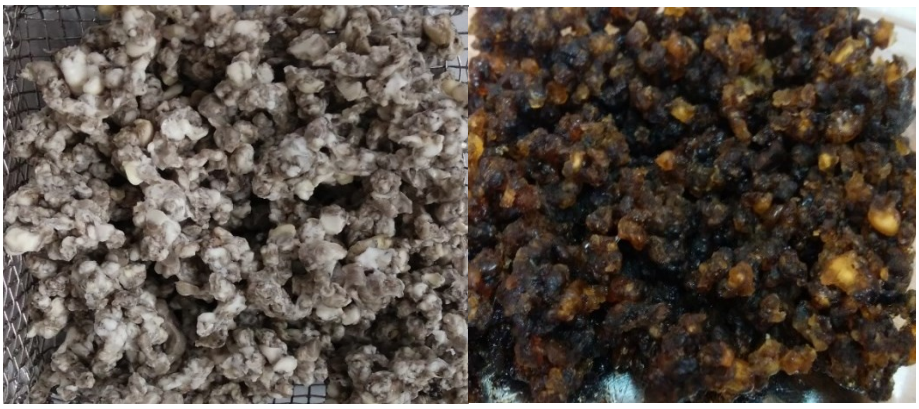
Substantial amounts of research work have been done on foodstuff and yet microstructure examination is also adapted in NR to effectively characterize its physical properties change as a result of different drying treatments. Chen *et al.*

(2012) worked on wet NR granules drying and, SEM was utilized to study the morphological structures of NR dried by HA and MW, respectively. The granules of NR dried by MW were of rough surfaces and the cutting traces produced during the processing of wet NR remained on the natural rubber surface; whereas, the NR granules dried by HA were of smooth surfaces, and the cutting traces produced during the processing of wet granules disappeared. There were a lot of big air-pores on the sponge-like surface of MW dried NR granules, and the interface between the air-pores and the NR was very distinct for these big air-pores, which did not adhere to each other. On the contrary, the air-pores on NR granules dried by HA were discrete and adhered with each other and the interface between the air-pore and NR was very fuzzy. This can be explained by the melting effect of the high drying temperature and long drying time of HA drying which linked the granules surface and small rubber threads; whereas the rough surface of MW-dried rubber resulted from outward migration of interior moisture to the exterior of NR granules when volumetric drying facilitates internal heat generation of water molecules. This observation of microstructure's change in dried rubber reveals the fact that the drying rate of MW dried rubber is superior to that of HA drying as the internal moisture can easily diffuse towards the surface with leaving of many air-pores at the rubber surface. Hence, a proper selection of drying technique able to mitigate adverse effects such as damage to cell membranes, discoloration and deterioration of wall structure, shrinkage which plays a decisive role not only on the absolute value of moisture diffusivity but also on the moisture content, bulk density, non-hygroscopic property in influencing the drying kinetics of a material.

Besides, Energy Dispersed X-ray, EDX through SEM system was used to inspect any metallic residue contaminated with the dried rubber. Sekhar (1958) stated that rubber contains metallic contaminants or if naturally occurring pro-oxidants are activated will reinforce the oxidation effect too. It is believed that the occurrence of a metallic compound such as copper, manganese and iron that significantly contribute to oxidative degradation. Bateman and Sekhar (1966) discovered that copper is a potent oxidant followed by iron and manganese has lesser oxidative effect in comparison to the other two. Therefore, discovering of any metallic residue inside the dried rubber will reduce the PRI and affecting the overall quality of rubber.

2.6.2 Drying temperature

In the rubber processing, careful selection of drying practice e.g. temperature is crucial to achieving a balanced system without sacrifice the rubber's quality. As shown in Figure 2.7 (b), under-heating in dried rubber caused the formation of white spots due to surfaces of granules have been dried out while some residue moisture still trapped inside. In contrary, the dried rubber will become darken and may lead to textural changes including shrinkage, case hardening, as a consequence of overheating at constant high drying temperature for long drying time (Cecil. and Mitchell, 2005). Hence, it is important to ensure consistency of drying conditions because even slight overheating can cause the rise of the rate of oxidation much more than the rate of diffusion and thus hardening (cross-linking);whereas gross overheating results in continuous chain breakdown(chain scission) and the hard surface turn softens and sticky (Chin Peng Sung, 1969).



(a) **(b)**

Figure 2.7: (a) fresh crumb rubber; (b) Rubber dried with HA oven at 100°C for more than 3 hours

In fact, viscosity and oxidation resistance are two major rubber properties that influenced by drying temperature. Therefore, overheating and holding the rubber crumb at high temperature for long hours should be avoided to minimize the oxidation (Sekhar, 1971; Cecil. and Mitchell, 2005). The similar finding suggested the higher drying temperatures lead to oxidation of the rubber which ruptures the molecular chain and affects the physical properties (Breymayer *et al.*, 1993). Excessive drying at elevated temperature affects mechanical processing ability of rubber (Imprastitichai, Thanawan and Suchiva, 2009) in two conditions. Firstly, the cross-linking of the rubber chains making the rubber harder as an effect of aldehydic condensation (cross-linking happened between carbonyl group, aldehydic [–CH=O] randomly distributed on the rubber chain and amino acids present among the non-rubber constituents of the latex). Secondly, the chain scission reaction which long rubber chain break soften the rubber as a result of chain breakage (Chindaprasert, 1997). As suggested by Pike and Watson (1952) and Watson (1953), the main causes to oxidation are (i)

exposure to sunlight; (ii) high temperatures; and (iii) excessive mechanical working. Oxidised rubber is lower molecular weight rubber and it has a close relationship with poorer physical and technological properties. Molecular size is the essential characteristic of an elastic polymer-its reduction in any way acts against the recognition of the best rubbery properties (Bateman and Sekhar, 1966). Zhang *et al.* (2014) commented relative excellent comprehensive properties, for instance, good strength, ageing resistance, wear resistance could be achieved if the raw natural rubber was dried under an appropriate drying time and drying temperature.

In fact, the majority of the industrial rubber dryer operates deep bed system with air flow through the bed temperature above 100 °C to maximize the product throughput, while striving to stay above the standard to do business (Sekhar, 1971; Yushan *et al.*, 1985; Tirawanichakul, Tirawanichakul and Tasara, 2011). Yet, it is concern that NR drying at low to medium temperature is still rare in literature and emphasis on innovative drying method for rubber is particularly needed especially with the availability of various advanced drying technology available at the market.

2.6.3 Thermal stability of dried rubber

Unlike other bio-origin product, however, raw NR is a thermally sensitive material that specifically susceptible to atmospheric oxidation due to the basic units of this polymer contain C-H and C=C groups which are vulnerable to attack by alkylperoxyl radicals (Howard, 1974). Even though natural antioxidants exist in NR latex such as phospholipids, amino acids, phenols, tocotrienols and

betaines (Nadarajah *et al.*, 1971; Prasertsit, Rattanawan and Ratanapisit, 2011), yet, oxidation can be manifested during collection by bacteria effects (Santos K. *et al.*, 2005), the processing operation which characterised by high temperature, thermal treatment and mechanical shear but only for a comparatively short time. The study of thermal oxidative degradation behaviours is one of the key essential element in understating the influence of heat in the processing and fabrication procedures in a polymeric system. Knowledge of degradation or mode of decomposition of NR can be useful in understanding the NR physical-chemical properties. Thermogravimetric analysis (TGA) is a well-known technique for defining the thermal stability of NR by study the range of service temperature with referring to a simple thermogram. Thermogravimetric data is useful by providing thermal stability information on thermal decomposition, mass loss of the material in different stages, threshold temperature, and the natural behaviour of the degradation of the material (Asaletha, Kumaran and Thomas, 1998; Satheesh Kumar, Yaakob and Siddaramaiah, 2009). Thermal stability analysis of dried rubber can reveal the effect of reaction temperature and time on the decomposition characteristics of NR during the drying process. A nuclear magnetic resonance (NMR) spectroscopy study of analysis of damage of NR suggested a high temperature may be the key factor promoting the oxidative degradation of commercial NR (Thuong *et al.*, 2016). This was confirmed by highest epoxidation level detected in SMR 20, a low grades rubbers processed at a higher drying temperature. Chain scission occurred when at high temperature due to more thermal energy transferred to NR cause the easy alignment of NR chains in the direction of the flow (Vega *et al.*, 2012).

2.6.4 Colour change

Colour assessment has been applied in the quality inspection of bio-origin material covering fruits, vegetables, herbs and spice and extensively reported in the literature. For instance, the effect of drying strategies and treatment in colour change of apple cubes (Mothibe *et al.*, 2014), blueberries (Zielinska and Michalska, 2016), herbs (Gan *et al.*, 2017; Tham *et al.*, 2017), salak fruit (Ong and Law, 2011) , pomegranate (Tontul and Topuz, 2017), tomato (Izli and Isik, 2015) were studied. Yet, in rubber processing, colour inspection is used to grade the crepe rubber, RSS (ribbed smoked sheets) and TSR according to a specific scheme. As for RSS and crepe natural rubber products, visually inspection is based on purity, elastic, and colour. RSS 1 to RSS 3 is higher graded RSS mainly applied for the manufacture of products for pharmaceutical and engineering use. The lower grades of RSS 4 and 5 are generally utilized for the manufacture of automobile tyres and general products (Thomsons Group, 2011). Prior to grading, these sheets are separated for visual inspection. Oxidized spots, heated, under cured, over smoked, burnt sheets are not permissible (Transport Information Service, 2002). The decomposition or degradation caused by oxygen and which alters the properties of elastomer is usually related to change of colour which causes to rejection. From Figure 2.8, ADS displayed the lightest colour when compared to the rest of the rubber sheet. The pale-yellow colour of ADS and RSS 1 turned darker when descending to grade RSS 3 and followed by RSS 5. The colour change from light yellow to black is depending on the extent of degradation because NR is easily oxidized when exposed to environmental conditions like oxygen, UV radiation and undergo different types of degradation such as oxidation degradation, thermal degradation and photo-degradation

(Veith, 1957). Chin Peng Sung (1969) reported that factors of discolouration of rubber could due to (I) drying temperature over 100 °C, (II) increase of ammonia preservation of the field latex, (III) excessive addition of chemicals such as sodium sulphite and sodium metabisulphite. Besides, some had reported the existence of polyphenolic compounds in NR may darken its colour too (Yapa, 1976; Wititsuwannakul *et al.*, 2002; Suwimon Siriwong. *et al.*, 2015).

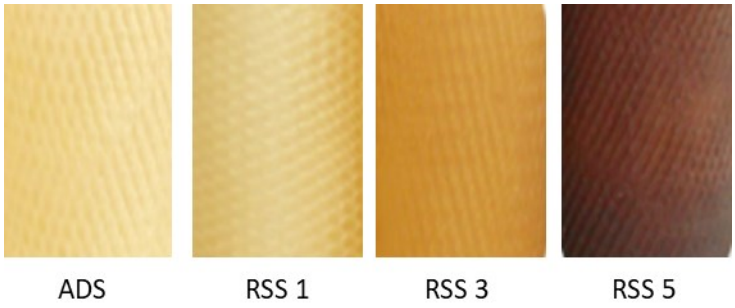


Figure 2.8: Colour grading based on the type of rubber and quality

Unlike RSS or crepe rubber products, TSR does not necessarily apply visual assessment for grading except for TSR-L rubber. TSR-L is light in colour with its cleanliness and excellent heat ageing resistance. Conversely, TSR-WF is similar to TSR-L but is a darker colour that fails to pass the Lovibond colour test by comparing to TSR-L. Although colour assessment has limited function in grading TSR-products, however, full understanding is still required in the impact of drying strategies in influencing the colour change of the product and its associated thermal quality.

2.6.5 Specific energy consumption (SEC)

An important measure of dryer efficiency is the specific energy consumption (SEC), which is defined as the heat input to the dryer per unit mass of evaporated water (Kudra, 2012).SEC values mainly depend on the drying methods in terms

of heat transfer, mixing of the bed material, physical and chemical proprieties as well as the operating conditions (Tarhan *et al.*, 2011). For instance, in the drying of peppermint leaves, the SEC values of the contact dryer were found lower than those of the HA dryers with ranged between 6.12 to 8.89 MJ/ kg.H_2O (Tarhan *et al.*, 2011). For industrial paddy drying system, the single stage drying involving only inclined bed dryer consumed 5.07 to 6.38 MJ / kg.H_2O while two-stage drying using fluidized bed dryer followed by inclined bed dryer used up 5.95 to 8.02 MJ/ kg.H_2O (Sarker *et al.*, 2013).On the other hand, an increase of the drying air temperature from 40 to 70° C has brought down the SEC values from 164.33 to 85.45 MJ/ kg H_2O for convective garlic clove drying in a laboratory dryer (Sharma and Prasad, 2006b). The same paper concluded that SEC in the microwave-convective drying process could be as low as 90% as compared to the convective drying process. Jindarat, Sungsoontorn and Rattanadecho (2015) reported the SEC values of combined microwave-hot air spouted bed is consistently lower than the hot air spouted bed drying method considered at the same hot air temperature and coffee bean initial weight. Similarly, J.Varith. *et al.* (2007) discussed the influence of moisture content on SEC for peel longan dried with combined microwave-hot air. These have demonstrated the combined drying method has an advantageous effect in lowering the SEC value. It was interesting to note that SEC of rubber has been reported only by several researchers in using the HA as a drying medium. In the experimental study of fixed-bed HA drying of crumb rubber, Tirawanichakul and Tirawanichakul (2008) concluded that air flow rates and initial moisture content are greatly affected the SEC and quality of rubber. The experiment yielded SEC ranging in between 20.58 to 38.06 MJ/kg H_2O whereby volumetric air flow rate acted as

the dominant effect on the SEC value. Likewise, Ekphon *et al.* (2013) reported that high drying temperature has lower SEC values compared to drying with low temperature such that the SEC values of ADS dried in 40 to 70° C was in ranges of 5.31 to 12.10 MJ/kg H_2O Various case studies have been reported in the combined drying process for drying purposes, however, there are none of the published studies relevant to combined drying of rubber which is deemed important to enrich the energy consumption database between industry and laboratory drying.

2.6.6 Plasticity retention indices (PRI), Wallace Plasticity (P_o) and volatile matter

The degradation reaction varies in rate and extent depending on the chemical and physical structures of the rubber, initial molecular weight, the presence of chemical and physical impurities and the nature of the external environment to which they are exposed (Satoudeh, 1993). Scott (1969) reiterated the most important accelerating environmental influences apart from oxygen, which is deleterious to polymer stability are heat, light, mechanical deformation, radiation, contamination by metal ions and ozone. Chain scission is mostly occurred in NR degradation and resulting in a weak softened stock, often showing surface tackiness (De and White, 2001; Wong, 2004). It is worth to mention that oxidation effects on a molecular level can be either cross-linking or breaking molecular chains. The cross-linking of the rubber chains making the rubber harder as an effect of aldehydic condensation whilst the breakage of the chains softens it (Zhang et al, 2012; Zhang *et al.*, 2014a). To measure the resistance of raw rubber to oxidative degradation, plasticity retention index (PRI) is applied

in dried rubber. A rubber with higher values of PRI indicates a less rapid break down and high-temperature ageing resistance in comparison to rubber with a low value (Baker, Fulton and Updated by, 2000). Meanwhile, higher P_0 suggests the rubber molecule is more intact means less chain scission which commonly leads to harder processing for this type of rubber in the factory. The latex grades generally have PRI values above 60%. For PRI values below 60%, PRI is correlated with the heat build-up and ageing of vulcanizates and with reversion during cure (Subramaniam, 2013). For a dry product, the higher the PRI, the better the ageing resistance. Whereas lower PRI rubber has inferior quality together with greater variability in this property. This index is a mechanical property that widely referred by manufacturers, mainly in tire industry that is one of the major NR users. The importance of PRI can be categorized into two: Firstly, it acts as a quantitative index for improving or controlling the quality of raw rubber; Secondly, exercising consistency in process control of the manufacturing industry. PRI provides a useful and numerical indication of an inherent quality of raw rubber and its processing characteristic. It possesses a close relationship with technological properties, notably to tensile strength, resilience and thus to heat build-up. Table 2.5 shows the variance of PRI in influencing the technological properties as reported by Bateman and Sekhar (1966).

Apparently, the PRI is directly proportional to both tensile strength and resilience meanwhile inversely proportional to the drying temperature. Higher temperature at prolonging drying hours could be deteriorated and affecting the PRI indices. PRI scored below 40% shows poorer rubber properties in

comparison to 60% and 80%. This standard is further governed by the SMR scheme whereby raw rubber with PRI above 40% is commonly accepted in the market.

Table 2.5: Technological properties of dry rubber in different of PRI (Bateman and Sekhar, 1966)

Plasticity retention index %	Tensile strength MPa	Rebound Resilience (Dunlop), %	Temperature at the 3rd hour °C
$20 < PRI < 40$	16.54-19.99	63-74	143.33-116.67
$40 < PRI < 60$	19.99-22.75	74-78	116.67-114.44
$60 < PRI < 80$	22.75-24.13	78-79	114.44-113.33

With concern of P_0 less than 30 as reject lots, many TSR factories prefer 34 or 35 ± 2 on P_0 because of easier processability. This standard could be accomplished by controlling the dryer temperature and residence time inside the dryer where both higher drying temperature and long residence in the dryers lowers the P_0 of dried rubber (Vare, 2003). This statement is further supported by Loke's study (Loke, 1974) on the effect of heat during and after drying on PRI for processed rubber, Loke has demonstrated that the increase of temperature with 40 °C difference (from 60°C to 100 °C) during the drying of raw NR does not necessarily weaken PRI, but the same application of heat to the rubber after drying reduced PRI remarkably. This suggests profound damage and degradation of natural rubber molecular chains when the thermal energy and moisture travelled along the opposite direction during the drying process.

Nevertheless, a small variation of the design of the heating oven, air circulation in the oven and temperature control of the oven also could have a significant impact on the PRI values (Vare, 2003). It is important that the information of the PRI in combination with the P_o of the raw material allows the manufacturer obtaining an equilibrium between ease of processing and optimum final viscosity and consequently satisfactory vulcanize properties (Loke, 1974). Although the difference, yet it does not bring any harm to the final product as the growth of mould can be prevented when volatile matter content is controlled below 0.6 wt.% (Rama Rao, 1984).

2.6.7 Texture Profile Analysis

Texture profile analysis (TPA) is one the prevalent compression test for determining the textural properties of foods besides some other industries such as pharmaceuticals, gels and personal care product (Texture Technologies Corp., 2017a). The quality of the products in several textural attributes such as hardness and springiness can be affected by process modifications (Herrero *et al.*, 2008). Currently, the most commonly used instrumental method is the TPA (for compression test) and the Warner-Bratzler (for shear test). A simple textural test with TPA would provide data about compression such as hardness, adhesiveness, springiness and cohesiveness. The compression parameters obtained with TPA have been utilized by many researchers to evaluate meat samples (Visessanguan *et al.*, 2004; Houben and van't Hooft, 2005) which possess approximate textural properties in comparison with rubber. Besides, rheological behaviour of rubber related products has been extensively studied by researchers by using the rubber processing analyser (RPA) (Escócio *et al.*, 2008; Zhang *et al.*, 2014b; Gao *et al.*,

2015; Honorato *et al.*, 2016). The physical test to the rubber can often relate to stress relaxation test to quantify the viscoelastic response of rubber. It involves both physical and chemical processes which occur simultaneously in them. Upon the action stress, chemical reaction happened by rearrangement of chemical bonds in the chains and lateral chemical bond whereas physical stress relaxation involves a restructuring of various structural elements which are removed from the equilibrium state by external forces and occur in the field of force of intermolecular forces (Vega *et al.*, 2012). However, there is little information about the application of TPA in the determination of textural properties i.e. springiness of dried rubber. This method is based on the ability of samples to recover from the deformation at the instantaneous relief of stress from the product. Certainly enough, the more a product is destroyed during the drying process, the less springiness it will exhibit (Texture Technologies Corp., 2017b). The springiness test has the advantage of simplicity and furthermore, the obtained results can be compared with the mechanical test performed with industrial standard i.e. SMR. This can improve the existing knowledge about the rheological behaviour of the dried rubber products by using texture profile analyser.

CHAPTER 3

MATERIALS AND METHODS

3.1 Sample Preparation-crumb rubber

Crumb rubber, a semi-processed raw natural rubber (*Hevea brasiliensis*) was supplied by Kuala Pilah Rubber Factory Sdn Bhd, located at Kuala Pilah, Negeri Sembilan, Malaysia (2.7490° N, 102.2502° E). Crumb rubber is a processed field coagulum produced by stages of the processes as described in Figure 1.2. Sizes of rubber coagulum were reduced to remove serum which followed by cleaning with water (to eliminate internal dirt) and subsequently shredded into crumb rubber. Figure 3.1 shows the image of crumb rubber in the form of a spongy mass of easily-separated granules. In this study, crumb rubbers were randomly selected from the bulk and its size was measured with Vernier callipers (530, Mitutoyo, Japan, with an accuracy of ±0.02 mm). Crumb rubbers with an average diameter size of 4mm were chosen for the drying experiments and the sample size was controlled at 50 ± 0.5 g. According to industrial practice, approximately 35% of water is removed from fresh crumb rubbers since the rubber has around 65% of dry rubber content (DRC) before it is fed to a trolley dryer (Tirawanichakul and Tirawanichakul, 2008; Ng and Tham, 2015). To determine the water loss from the crumb rubber, the moisture content was measured by ASABE (1999) method. The initial moisture content of crumb

rubber was in the range of 21% to 48% dry-basis (db.). The initial moisture content of crumb rubber was found to be varied because of the pre-treatment conditions of rubber prior to the drying process as well as latex preparation and functionality of the processing machine (Gale, 1959; Chen, Wang, *et al.*, 2012).



Figure 3.1: Crumb natural rubber

3.2 Dryers specifications and operating conditions

3.2.1 Convective hot air-drying oven

A laboratory scale convective hot air-drying oven (UFB 500, Memmert, Germany, range: 30°C to 220°C with an accuracy of 0.5°C) was used as a primary dryer for convective hot air drying (HA). Isothermal drying of crumb rubber was carried out at 100°C, 110°C and 120°C respectively with a constantly forced circulation of air at 1.0 ± 0.2 m/s. HA was used as a reference and it represents the conventional crumb rubber drying method. Drying sample was placed on top of a wire mesh tray to ensure a good circulation of hot air flow around the sample. The rating watt energy consumption of oven under different setting was recorded at 214Whr (100°C), 244Whr (110°C) and 274Whr (120°C), respectively. The oven inlet air humidity was measured at 64% to 68% and the outlet air humidity has a range of 9% to 18% by using a portable handheld

thermos hygrometer (LOGPROBE 31, E+E Elektronik GES.M.B.H, Austria; working range: 0…100% RH, accuracy ±2% RH).

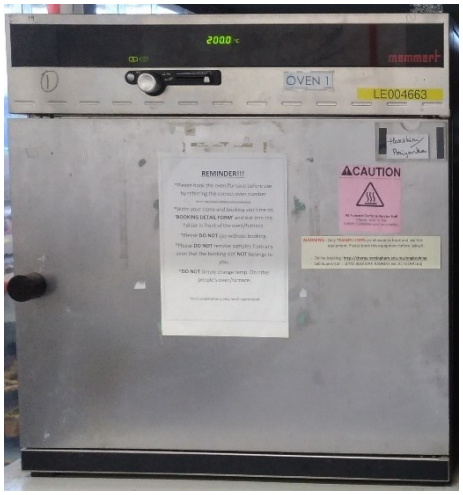


Figure 3.2: Convective hot air-drying oven

3.2.2 Vacuum Oven

As shown in Figure 3.3, laboratory scale of vacuum oven (VO 400, Memmert, Germany, Temperature range: 20°C to 200°C (accuracy: $\leq \pm 0.3$K); Pressure range: 10mbar to 1100mbar) was used to conduct vacuum drying. Unlike the conventional heating system of the interior walls, this oven applies direct heating mechanism with thermos-shelves to allow direct contact between the heating system and the sample which is claimed to reduce 75% of the process time (Memmert, 2015). In the experiment, the temperature of thermos-shelves was fixed at 80°C whereas operating pressure was controlled at 100mbar to examine the drying of crumb natural rubber under vacuum conditions. For information, an operating pressure of 100mbar has a reduced water boiling point at 45.82 °C (Haar, Gallagher and Kell, 1984) and 80°C is already sufficient for this drying purposes. The vacuum drying system consists of a separate vacuum pump

(MD4CNT, Vacuubrand, Germany, operating ranges: 1.5m bar at max. 3.8 m^3/h) and it has a power consumption of around 2421W when in operation.



Figure 3.3: Vacuum oven

3.2.3 Microwave Oven

For dielectric heating or microwave drying, a domestic microwave oven (ME711K, Samsung, Malaysia, power output ranges: 100W to 800W) was used in the experiments. The 20 litres oven has an operating frequency of 2450MHz and maximum power consumption of 1150 W. Magnetron of the oven is cooled down by cooling fan to minimize overheating during the heating process. The oven is equipped with a turntable rotated at 6 RPM to ensure a uniform heating over the fresh sample (Samsung, 2012). The power setting can be adjusted individually until 800W steps. Three power level i.e. 600W, 700W and 800W were selected for rubber drying.



Figure 3.4: Microwave oven

3.2.4 Microwave-convective dryer

A laboratory scale tabletop microwave-convective dryer was designed to perform hybrid drying which allows two drying methods operates simultaneously. This prototype dryer was fabricated by I-Lab Instrument Sdn. Bhd. (Selangor, Malaysia). It is designed as an open loop system whereby the drying air is constantly circulated within the drying system while allowing some atmospheric air to enter the drying system to displace humid air. It consists of a domestic microwave oven (ME711K, Samsung, Malaysia, power output range: 100W to 800W), a 500W tube type heating element covered with aluminium fin and a regenerative blower fan (CR 129-m, 0.25kW, CR electric, China). The microwave oven has the similar features as described in Sections 3.2.3 and a heater is installed in the drying chamber to further increase the drying air temperature. Ambient air is drawn from the environment and channelled to stainless steel hot air duct which is connected to the bottom of the microwave oven. The oven is designed with air change rate between 38 and 70 times per minute and the drying air flows in a parallel direction to the sample. The magnitude of air flow is regulated by a control valve. Overall built-up dimension of the dryer is 0.7 m (length) x 0.5 m (width) x 0.51m (height) with a drying chamber sized of 0.02 m^3. Microwave leakage tests were carried out using

microwave leakage detector (CEM DT-2G, India) to ensure that microwave leakage remains well below the recommended safe maximum value of 5 mW per square cm at 5 cm from the oven surface as recommended by Food and Drugs Administration (FDA), (2017). Figure 3.5 and Figure 3.6 show the actual view and schematic diagram of the microwave-convective dryer.



Figure 3.5: Microwave-convective dryer

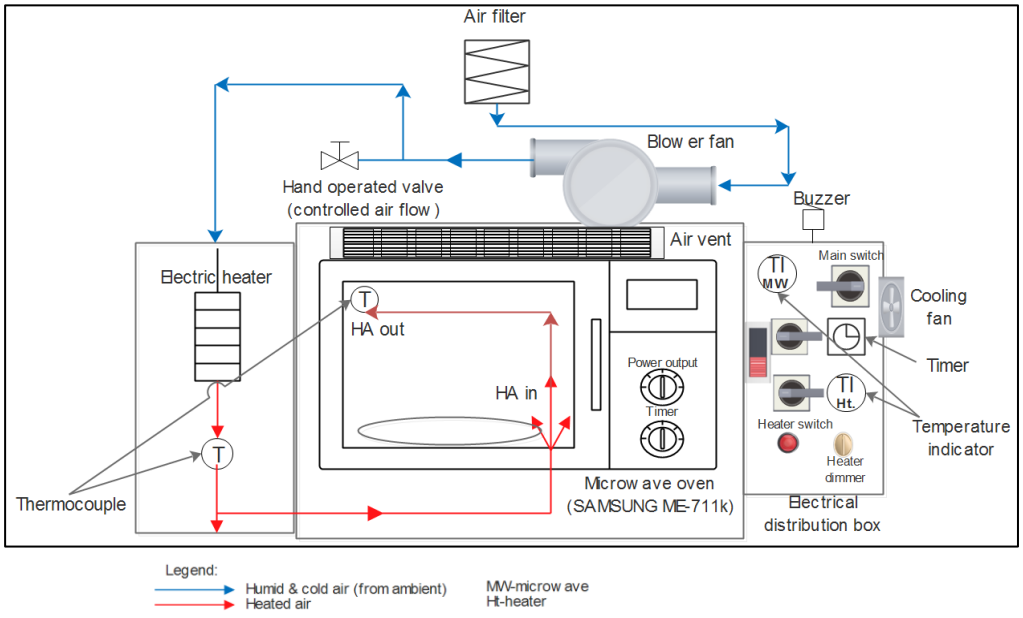


Figure 3.6: Schematic diagram of microwave-convective dryer

An E+E air velocity probe (LOGPROBE 60, E+E Elektronik, Austria, measuring range: 0 - 20 m/s ±0.2m/s) was used to measure the real-time air velocity (See APPENDICES Figure B1) inside the drying chamber prior to the drying process. As shown in Figure 3.6, an air inlet opening is adjacently placed near to the glass plate whereas air outlet opening (air vent) is located on the top side of the oven. The dryer operates without air recycling within the chamber, this maintains the process air at constant temperature and humidity conditions. Drying air circulated from bottom to top via the inlet vent and exit through the vent hole because of positive pressure built up in the chamber during drying which forces the moist air to be discharged into the surrounding. It is noteworthy to mention that air vent was designed in homogenisation grids to evenly distribute the air flow within the chamber.

3.3 Drying methodology

3.3.1 Overview of drying strategies

Drying strategies applied in this study were HA, MW, HA/MW, MW/HA, VD/MW, MW/VD and MWHA as illustrated in Figure 3.7. All drying equipment is placed indoor with room temperature controlled between 18 °C and 20 °C by air-conditioner system to minimize the effect of ambient humidity and air temperature to the operating condition.

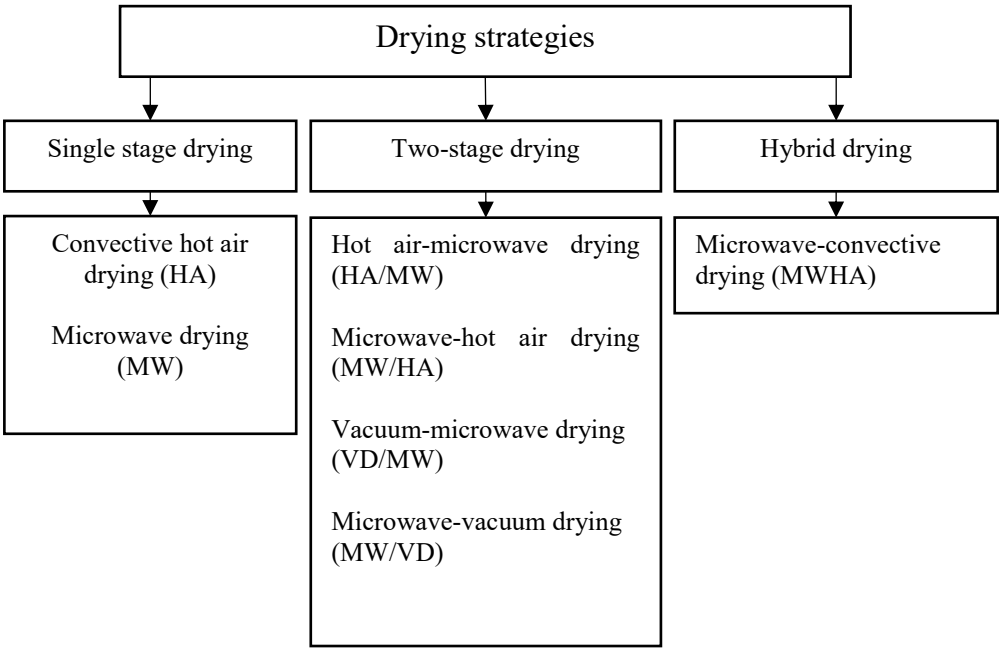


Figure 3.7: Single-stage, two-stage and hybrid drying

3.3.2 Single stage drying

3.3.2.1 Convective hot air drying (HA)

About 50 ± 0.5 g of crumb rubber was spread thinly on a tray fabricated with woven stainless cloth (mesh size:8 x 8; wire diameter: 0.71 mm) which was inserted into the convective hot air-drying oven. The air temperature was controlled at 100°C, 110°C and 120°C respectively. Crumb rubbers were drawn out and weighted using a mini digital platform scale (I-2000, Guangdong, China, capacity 500g with accuracy ± 0.01 g) from a minute interval to hourly interval until equilibrium moisture content (EMC) was achieved. The EMCs were determined by extending the drying process until the change in sample mass between two successive reading is less than 0.01 g in each drying experiment (Digvir, 2003).In most cases, the weighing took less than 20 seconds and the interruption of the steady conditions of drying by cooling and the time lost was found to be insignificant (Madamba, Driscoll and Buckle, 1996; Wongwises and

Thongprasert, 2000; Ong and Law, 2011). All drying samples were triplicate and the variance of results was analysed via statistical software.

3.3.2.2 Microwave drying (MW)

In microwave drying, 50 ± 0.5 g of crumb rubbers were placed inside a 500 ml Pyrex® beaker instead of metal wire mesh. By using glassware, arcing or fire in oven cavity can be prevented during the drying process (Samsung, 2012). The sample was placed in the centre of the turntable which rotated at 6 rpm inside the microwave-convective dryer. The presence of rotating turntable is necessary to prevent local overheating while drying. In this study, the drying power was regulated in the range of 600W, 700W and 800W respectively for each set of experiment. Also, microwave power was regulated in step-up and step-down order e.g. from 16 *W/g* reduced to 14 *W/g* and finally 12 *W/g* with an effective drying time of five minutes (1), ten minutes (2) and 15 minutes (3), correspondingly. The procedure of weighing was similar to Section 3.3.2.1 but at a different interval which started from one minute, two minutes, four minutes and five minutes until the EMC was reached by the product. Special attention was paid to ensure that the sample was not charred.

3.3.3 Two-stage drying

3.3.3.1 Hot air-microwave drying (HA/MW and MW/HA)

In contrast to a single stage drying, hot air-microwave drying employed two drying methods i.e. HA and MW at a different stage of the drying cycle. The two-stage drying intermediate moisture content was decided based on the drying curves of single stage MW and HA drying. Crumb rubbers were dried in

convective hot air dryer for the first 120 minutes. Then, the sample was taken out and dried in a microwave oven for another 30 minutes. Similar drying period was also applied to the reversal of HA/MW whereby the drying started with MW oven for 30 minutes and finished by HA dryer for 120 minutes. For HA, the air temperature was controlled between 100°C and 110°C, respectively. In MW, the power of microwave was regulated at 600W, 700W and 800W for the respective experiment. Weight loss was monitored offline periodically until the EMC was achieved.

3.3.3.2 Vacuum-microwave drying (VD/MW and MW/VD)

In VD/MW, vacuum oven was used to pre-dry the crumb rubber at first one hour and followed by post drying in microwave oven until EMC was reached. Vacuum oven was operated at constant drying temperature: 80 °C; with a partial vacuum pressure at 100 millibar (10kPa), where boiling point of water is reduced to 45.4 °C while MW was carried out in power level ranging from 600 W to 800 W. Similarly, a reversal of the two-stage drying was carried out where MW was introduced in the first stage for 15 minutes followed by VD for about one hour which has the same total drying times as VD/MW. Weight loss in MW was measured in five-minute interval whereas VD had longer drying interval i.e. 10 minutes per weighing session until constant dried rubber weight is reached.

3.3.4 Hybrid drying

3.3.4.1 Microwave-convective drying (MWHA)

Microwave and hot air-drying method were carried out simultaneously in the microwave-convective dryer. Crumb rubbers were placed inside beakers before

putting it on the turntable. The rotation of the turntable and forced air circulation (hot air) are necessary for this system to avoid local overheating of the sample and to facilitate the removal of excess moisture from the dryer while drying. Drying was carried out based on the setting as indicated in Table 3.1. It was necessary to operate the heater for 20 to 30 minutes before the experiment to allow the hot air temperature inside the microwave cavity reaches the pre-set temperature and stabilizes at the set value. Different settings were employed, namely (1) the heater capacity was regulated to its maximum (50° C) and the air vent passage was fully sealed by hand valve to allow maximum air flows to the drying chamber;(2) the air vent was opened to reduce air flow speed by a factor of two while the heater capacity was unchanged; (3) the heater capacity was reduced to its minimum (37° C) and air velocity remained the same as (2). Figure B2 (See APPENDICES) shows the process flowchart of MWHA. For all settings, samples were weighed until EMC was reached by three consecutive readings. Instantaneous product temperature was measured by inserting a digital probe-type thermometer (PDT 550, range: -50 °C to 300°C, accuracy: ±1.0°C, UEi Test Instruments, UK) into the product centre and manually recorded.

Table 3.1: Operating conditions used in the microwave-convective dryer

Group	Drying system	Microwave power level (W)	T_{air} (°C)	v (m/s)
1	MW600HA50V5.2	600	50	5.2
	MW700HA50V5.2	700		
	MW800HA50V5.2	800		
2	MW600HA37V2.8	600	37	2.8
	MW700HA37V2.8	700		
	MW800HA37V2.8	800		
3	MW600HA50V2.8	600	50	2.8
	MW700HA50V2.8	700		
	MW800HA50V2.8	800		

3.4 Drying Kinetics

3.4.1 Moisture content

The moisture content of the drying product in dry basis (db.), X_d was determined periodically based on the weight loss of the sample in accordance to AOAC (1996):

$$X_d = \frac{W_i - W_d}{W_d} \times 100 \quad (3.1)$$

Where moisture content measures the inherent liquid or volatile percentage within the wet product; W_i is the weight of rubber (g) at that specific time t (minute); W_d is the weight of dry matter in the product. Meanwhile, the dimensionless moisture ratio, $\emptyset$ of crumb rubber during drying experiments was computed by following Fick's second law (Crank, 1975) in the thin layer drying model:

$$\emptyset = \frac{X_i - X_f}{X_o - X_f} \quad (3.2)$$

X_o, represents the original moisture content, X_i is the moisture content at time t, and X_f is the equilibrium moisture content.

3.4.2 Drying rates and characteristic drying curve

Drying rates, dX/dt (g H_2O/g DM. min) were calculated from the difference in moisture content between the product being dried at a given time. It was

determined by approximation of the derivatives to finite differences as shown in Equations 3.3,3.4 and 3.5 (Guiné *et al.*, 2006; Hii *et al.*, 2009).

*First order forward finite difference at $t = t_0$ (first point),

$$\frac{dX}{dt} = \frac{X_1 - X_0}{t_1 - t_0} \tag{3.3}$$

*Second order centred finite difference at $t = t_n; n = 1, \dots, n-1$ (intermediate point),

$$\frac{dX}{dt} = \frac{X_{n+1} - X_{n-1}}{t_{n+1} - t_{n-1}} \tag{3.4}$$

*First order backward finite difference at $t = t_f$ (final point),

$$\frac{dX}{dt} = \frac{X_f - X_{f-1}}{t_f - t_{f-1}} \tag{3.5}$$

Where t is time (minute) and X_o and X_f are the moisture content (g H_2O /g DM) at initial and equilibrium conditions, respectively. Characteristic drying curve is derived based on the Van Meel transformation, consisted of $\emptyset$ and normalize drying rate,f_n (Equation 3.6).

$$f_n = \frac{\left(-\frac{dX}{dt}\right)_t}{\left(-\frac{dX}{dt}\right)_o} \tag{3.6}$$

Where f_n , the dimensionless drying rate. The general form of characteristic drying curve is given by $f = f(\emptyset)$,where $0 \leq \emptyset \leq 1$.Goodness of fit of the characteristic drying curves was determined with adjusted R-square values which were generated by using Curve Fitting Toolbox 3.3 (MATLAB® R2016a, The MathWorks, Inc., USA)

3.4.3 Effective moisture diffusivity

In the single layer model, effective moisture diffusivity of crumb rubber was estimated from a one-dimensional universal solution of Fick's diffusional model for the slab geometry (Tirawanichakul and Tirawanichakul, 2008). The equation assumes moisture transfer in the material is symmetric with respect to the centre, uniform initial moisture distribution, negligible shrinkage and constant moisture diffusivity (Crank, 1975; Vazquez *et al.*, 1999).

The following assumptions were made when using this model namely:

I. The crumb rubber is assumed in slab shape composed of series of thin layers
II. Internal heat transfer usually occurs much faster than internal mass transfer which the initial moisture content and temperature are assumed uniform inside the product (Coumans, 2000)
III. Surface moisture content is in equilibrium with the air during the drying
IV. Heat loss is very minimum and can be negligible
V. Moisture movement and heat transfer are unidimensional
VI. The existence of a thermal equilibrium between grain and drying air and the outlet drying air condition from the first thin layer is used as the input drying air condition for the next layer

VII. Uniform distribution of air throughout the dryer

VIII. The material is homogeneous and non-deformable so that shrinkage is negligible

The general solution for an infinite slab shape of moisture ratio can be referred to Equation 2.3. The equation is simplified into Equation 3.7 and the D_{eff} can be determined by the graphical method by plotting ln (Ø) versus drying time (*t*) and calculated by using Equation 3.8.

$$\ln(\emptyset) = \ln\frac{8}{\pi^2} - \left(\frac{\pi^2}{4L^2}D_{eff}t\right) \tag{3.7}$$

$$k_0 = \frac{\pi^2 D_{eff}}{4L^2} \tag{3.8}$$

Assessments between the experimental and predicted data by using different thin layer drying curve models were carried out using least squares nonlinear regression analysis with Generalized Reduced Gradient (GRG) algorithm (Excel® Solver version 2016, Microsoft Corp., USA). The terms that were used to evaluate the best fit of the tested models to the experimental data by comparing the moisture ratio values are the coefficient of determination(R^2); root mean square error (RMSE) and the reduced chi-square(χ^2), which are given by Equation 3.9, 3.10 and 3.11.

$$R^2 = 1 - \frac{\sum_{i=1}^{N}(\phi_{pre,i}-\phi)^2}{\sum_{i=1}^{N}(\overline{\phi_{pre,i}}-\phi_{exp,i})^2} \tag{3.9}$$

$$\chi^2 = \frac{\sum_{i=1}^{N}(\phi_{pre,i}-\phi_{exp,i})^2}{N-z} \quad (3.10)$$

$$RMSE = \left(\frac{\sum_{i=1}^{N}(\phi_{pre,i}-\phi_{exp,i})^2}{N}\right)^{\frac{1}{2}} \quad (3.11)$$

Where ϕ_{exp} is the experimental dimensionless moisture ratio; ϕ_{pre} is the predicted dimensionless moisture ratio; N is the number of experimental data points, and z is the number of parameters in the model. A good fit of a model is characterized by the highest R^2 ,the lowest RMSE and χ^2.

3.5 Specific Energy Consumption (SEC)

The drying of crumb rubber, a process of simultaneous heat and mass transfer, represents an energy-intensive operation of some industrial significance. In this context, energy consumption was both measured and calculated. Energy consumption for each drying method was directly measured by using a digital clamp multi-meter (UT-203, UNI-T, China; accuracy: $\pm 2\%$) and the power consumption was calculated by Equation 3.12.

$$E_t = P_e \frac{t}{3600} \quad (3.12)$$

Where E_t represented total energy consumption (Wh); P_e is the rating power of equipment (W) and t is the drying time in seconds. The power level of microwave with respect to sample size can be expressed in terms of power output per unit of mass of the substance as shown in Equation 3.13.

$$P_d = \frac{P_e}{M_i} \tag{3.13}$$

Where P_d is the power density(W/g); P_e= power output from microwave; and M_i= initial mass of fresh crumb rubber(g).

In this research, the efficiency of drying was evaluated by specific energy consumption (SEC). SEC is defined as the energy required to evaporate a unit mass of water in the drying of product from its initial moisture content to the final moisture content. The total energy utilization rate was estimated with reference to Equation 3.14 (Yongsawatdigul and Gunasekaran, 1996):

$$SEC = \frac{t \times P \times \left(1 - X_f\right) \times 10^{-6}}{M_i \times \left(X_o - X_f\right)} \tag{3.14}$$

Where SEC (MJ /g H_2O);t is the total effective drying time (s) when power is on; P, total power input (W);M_i, initial mass (g); X_o, initial moisture content (fraction) and X_f, final moisture content (fraction).

3.6 Physicochemical analysis

3.6.1 Colour changes

Colour changes of dried rubber were measured using a handheld colour meter (LC100, The Tintometer Ltd, England). Three samples of rubber crumb were picked from the experiments and arranged in a Petri dish to form a thin layer sheet that could cover the nosecone of the colour meter sensor. Images of the single sample were captured in three replications at different point of interest on the samples to obtain an average value. Equations of colour changes namely

L^*, a^*, b^* are given in Table 3.2. They were used to compute the change in different colour parameters.

Table 3.2: Colour parameters (CIE)

Parameters	Colour indications	Equations
Lightness,L^*	0 (black) to 100 (white)	$\Delta L^* = L^* - L_o^*$ (3.15)
Greenness/Redness, a^*	- 60 (greenness); + 60 (redness)	$\Delta a^* = a^* - a_o^*$ (3.16)
Blueness/Yellowness, b^*	- 60 (blueness); + 60 (yellowness)	$\Delta b^* = b^* - b_o^*$ (3.17)

The total colour change of the dry product was determined using Equation 3.18 (Chong *et al.*, 2009; Ong and Law, 2011).

$$\Delta E^* = [(L^* - L_o^*)^2 + (a^* - a_o^*)^2 + (b^* - b_o^*)^2]^{\frac{1}{2}} \quad (3.18)$$

3.6.2 Texture Profile Analysis

A single column texture analyser (TA. XT plus, Stable Micro Systems, UK) was used to determine the mechanical properties i.e. springiness of dried rubber. The test requires that the surface of the sample is smaller than the diameter of the probe. Hence, a 75-mm aluminium compression platen probe (P/75, Stable Micro Systems, UK) was employed to compress dried rubber sample of 10.0± 2 mm (length) x 5.0 ± 1.0 mm (width) x 5.0 ± 1.0 mm (height). First, the samples were placed in the middle of texture analyser platform and the probe was

positioned 20 mm above the surface of the sample. The probe was slowly descending and it compressed the sample until 20% of the product height was lowered, then the probe held at this distance for 60 seconds before it was withdrawn from the sample and returned to its starting position. As shown in Figure 3.8 and Equation 3.19, the springiness is measured by dividing the force,F_2 after 60 seconds with the maximum force F_1 , and multiply by 100%. Hence the highest value of springiness is 100%. The higher the springiness, the more elastic the product is. Each set of the test has three replications.

$$Springiness\ (\%) = \frac{F_2}{F_1} x100 \tag{3.19}$$

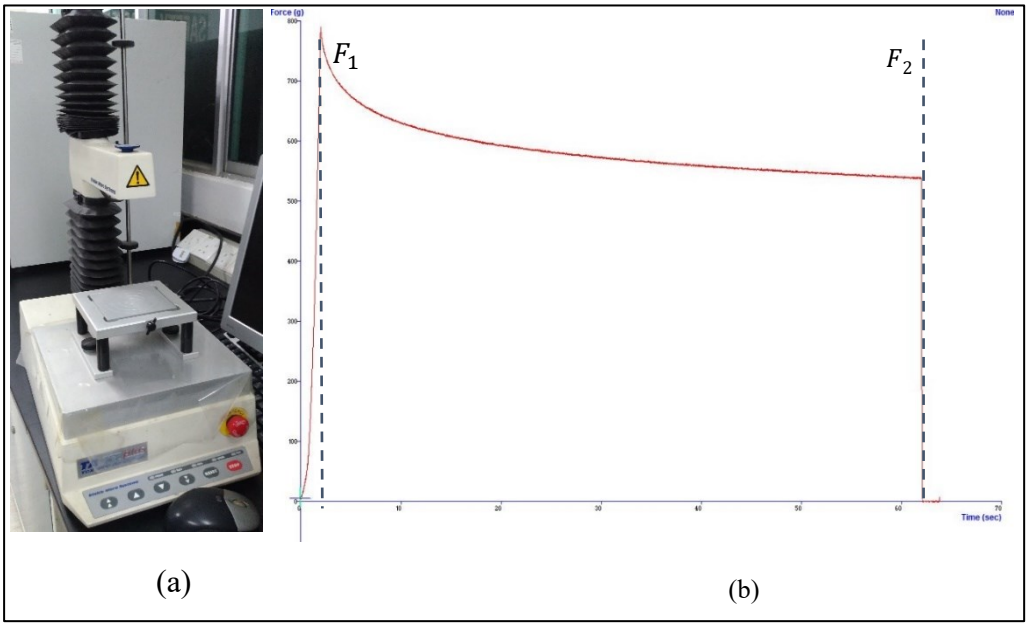


Figure 3.8: (a) Texture profile analyzer and (b) Characteristic plots of load vs. time- springiness

3.6.3 Thermo-Gravimetric Analysis (TGA)

A Mettler Toledo TGA/DSC thermogravimetric analyser (TGA/DSC 1, Mettler Toledo, USA) was applied for the thermal oxidative decomposition measurement. TGA measures the mass loss of a sample as a function of temperature. As natural rubber has low thermal stability, the polymer chain is susceptible to degradation upon the heat treatment such as drying. Thus, the characterization and evaluation of polymer thermal stability are determined via TGA (Loadman, 1998; Ashraf, Ahmad and Riaz, 2009; Rohana Yahya, Azura and Ahmad, 2011). A dried sample weighed about 10mg was chosen randomly from each group of drying strategies to perform TGA test. The test begins with heating started from 100 to 600 °C at a heating rate of 10°C/min under a nitrogen atmosphere with a flowing rate of 50 mL/min. With the help of a built-in software (STARe, Mettler Toledo, USA), the thermogravimetric (TG) curves were interpreted as percentage weight loss as a function of temperature. Meanwhile, the decomposition stages of dried rubber were analysed using a derivative of the TG curves (DTG).

3.6.3.1 Kinetic studies of TGA

The kinetic parameters were employed to evaluate the degradation processes by integration methods for the kinetic analysis of the single thermogravimetric curve data. Several approaches such as Coats and Redfern, Horowitz-Metzger have been reported in the literature for the integral of the Arrhenius equation to obtain the activation energy from experimental data (Shih and Chieh, 2007; Cai and Bi, 2008; Galiani *et al.*, 2013). Pérez-Maqueda, Sánchez-Jiménez and Criado (2005) recommended the Coats and Redfern approach in comparison to

others due to smaller error for the entire range of E_a/RT .In this research, the activation energy of decomposition, E_a has been calculated from the slopes of the best fit straight lines. The Coats Redfern's equation is given by Equation 3.20.

$$\log_{10}\left[\frac{1-(1-\alpha)^{1-n}}{T^2(1-n)}\right] = \log_{10}\frac{AR}{aE_a}\left[1-\frac{2RT}{E_a}\right]-\frac{E_a}{2.3RT} \quad (3.20)$$

for all values of n except $n = 1$

Where α is the fraction of sample decomposed at time, *t*; T is the derivative peak temperature; n is the order of reaction;A is the frequency factor;R is the ideal gas constant $\left(8.3145\frac{J}{mol.K}\right)$ and E_a is the activation energy of the decomposition. The degradation temperature at weight conversion rate of 10%, 30%,50%,70%,90% and T_o , T_r and T_e was chosen to plot $\log_{10}\left[\frac{1-(1-\alpha)^{1-n}}{T^2(1-n)}\right]$ as a function of $\frac{1}{T}$ for $n = \frac{2}{3}$ (Coats and Redfern, 1964). Straight line obtained from least square regression method which gives the best fit slope was used to determine the E_a.

3.6.4 Scanning Electron Microscopy (SEM) Analysis

Surface topography of dried rubber was observed with SEM (SWN; Quanta 400F, FEI, Hillsboro, OR) at an accelerating voltage of 10 kV. The fractured surface was obtained by splitting of the quenched rubber sample in liquid nitrogen(Chen, Zhang, *et al.*, 2012). A sputter was used to pre-coat a layer of conductive gold onto the surface and fractured surface of dried rubber samples obtained from different drying strategies before SEM observation. Three images

with a resolution of 0.97 Mega Pixels (1,024 x 943 pixels) were captured in each region, resulting in nine images (3 images x 3 regions) for each specimen.

3.6.5 Determination of Plasticity Retention Index (PRI) and Volatile Matter (VM)

The analysis of block rubber properties in accordance with Standard Malaysian Rubber (SMR) scheme is widely accepted in rubber industry for its standardization purpose and serves as an essential criterion for quality assessment for rubber processing. The PRI is a mechanical test that provides a rapid assessment of the susceptibility of NR to thermal oxidative degradation (Bateman and Sekhar, 1966). The test comprises of measurements of the plasticity of NR samples before and after heating in an air oven for 30 ± 0.25 minutes at 140°C±0.5°C applying the parallel plate plastimeter with a platen 10 mm in diameter (RRIM, 1992b). The PRI is given in Equation 3.21.

$$PRI = \frac{P_{30}}{P_0} \times 100 \tag{3.21}$$

Where P_0 is the plasticity number of the unaged test pellet; P_{30} is the rapid plasticity number of the test pellet after ageing at 140 °C for 30 minutes. On the other hand, VM is a chemical test that is used to determine residue moisture or matter which is volatile at 100 °C in dried rubber. The lower the VM, the lesser the residue moisture and also lowered the possibility of contamination by solvents in dried rubber (RRIM, 1992a). The volatile matter can be determined by Equation 3.22.

$$Volatile\ matter(\%) = \frac{A-B}{A} \times 100 \quad (3.22)$$

Where A is the weight of test portion before drying; B is the weight of test portion after drying. Both PRI and VM tests were carried out by Mech Chem Laboratory Sdn. Bhd., Klang, Malaysia in accordance with standard test methods recommended by Rubber Research Institute Malaysia, RRIM viz. VM: SMR Bulletin No.7, B5,1992 and PRI: SMR Bulletin No.7, B8, 1992, respectively.

3.7 Statistical Analysis

3.7.1 Multiple regression analysis

The multiple regression tests were applied to identify the relationship between two or more independent variables in affecting a dependent variable. In this research, the independent variable (X) is defined as the predictor variable which includes microwave power level, degree of hot air temperature as well as air velocity. Meanwhile, the dependent variables which includes effective diffusion coefficient and specific energy consumption, which is also known as the criterion variable ($\hat{Y}$). The criterion variable score is predicted using n predictor variables (X_1, X_2, X_3 ... and X_N), where N >2 (Chua, 2013). The multiple regression equation is presented as below.

$$\hat{Y} = b_1X_1 + b_2X_2 + b_3X_3 + \cdots + b_NX_N + a \quad (3.23)$$

Where b is the regression coefficient for each predictor variable; a is the regression constant. All the independent values were recorded into the SPSS Statistics® program (Version 22, IBM, USA). Significance of a model in regression analysis was examined by the coefficient of determination (R^2) as

well as the *t*-test value. In multiple regression analysis, the R^2 is the square of the correlation between the response values and the predicted response which has the similar definition as defined in simple regression analysis as shown in Equation 3.9 (Anonymous, 1997). The R^2 value determine how well data fit to a statistical model i.e. the closer R^2 to 1, the better the fitting of the model to the experimental data. On the other hand, the *t*-test uses means and standard deviations of two samples to make a comparison. The formula for *t*-test is given by Equation 3.24.

$$t = \frac{\bar{x}_1 - \bar{x}_2}{\sqrt{\frac{s_1^2}{N_1} + \frac{s_2^2}{N_2}}} \tag{3.24}$$

Where, $\bar{x}_1$= mean of the first set of values; $\bar{x}_2$= mean of the second set of values; s_1= Standard deviation of the first set of values; s_2= Standard deviation of the second set of values; N_1= Total number of values in the first set; N_2= Total number of values in the second set. The lower *t*-test value indicates a higher level of the influences of independent variables toward the dependent variable.

3.7.2 ANOVA

In this study, dried rubber characteristics in terms of colour changes, SEC and effective diffusion coefficient were statistically evaluated. All statistical analyses were performed using the SPSS Statistics® program (Version 22, IBM, USA). One-way analysis of variance (ANOVA) and Duncan's multiple range tests were used to evaluate the significant difference of the data at $p < 0.05$. The data was expressed as mean ± standard deviation.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Introduction

The drying kinetics and physicochemical changes of crumb rubber processed by different drying methods are presented in this chapter. Results of drying kinetic of crumb rubber including the moisture removal rate and effective diffusion coefficient are presented in Section 4.2. Whereas Section 4.6 elaborates the physicochemical change of crumb rubber including colour changes, thermal oxidation level as well as crumb rubber surface morphology.

4.2 Drying Characteristics

4.2.1 Single stage drying: HA

HA experiments were conducted in universal lab oven with temperature ranging from 100 °C to 120 °C. The set temperature was in accordance with the operating temperature commonly applied in the industrial processing of crumb rubber. Drying curves of HA dried crumb rubber with respect to drying times are shown in Figure 4.1. For information, the average initial moisture content of crumb rubber from the respective experiment is inconsistent with discrepancies lies

between 16% and 51%, therefore the instantaneous moisture content was normalized to moisture ratio. In Figure 4.1, moisture ratio of crumb rubber dropped considerably in the first 80 minutes due to the free water removal from the surface and the water at the surface is easy to remove. As drying continues, internal moisture starts to diffuse out to the surface at a slower pace and drying rate is gradually decreased. Similar findings have been reported by Auria *et al.* (1991) and Sethu.S (1967) where drying speed of rubber sheet and crumb rubber decreases considerably when the water content is less than 10%. At this stage, the surface is dried while the core is still wet, whereby the dried layer become resistance to the mass transfer, resulting in a reduction of the evaporation rate and non-uniform heating. Drying characteristic curve as shown in Figure 4.1 is in fact in agreement with the simulated results obtained by Tirawanichakul and Tirawanichakul (2008) who carried out one stage crumb rubber drying at 100 °C at air flow rate 2.2 m/s.

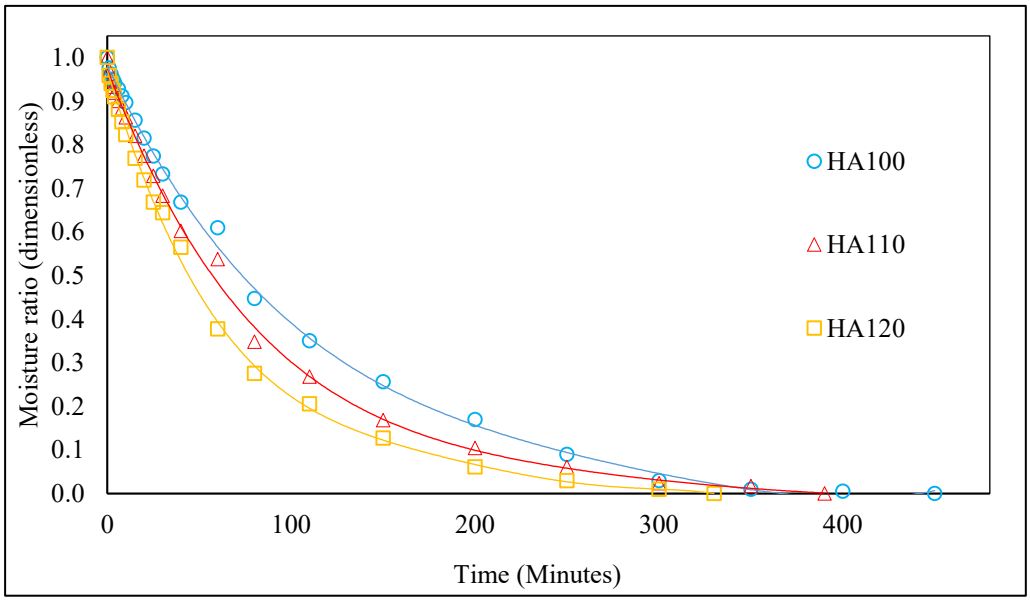


Figure 4.1: Moisture ratio of dried rubber with drying time by using hot air drying (HA)

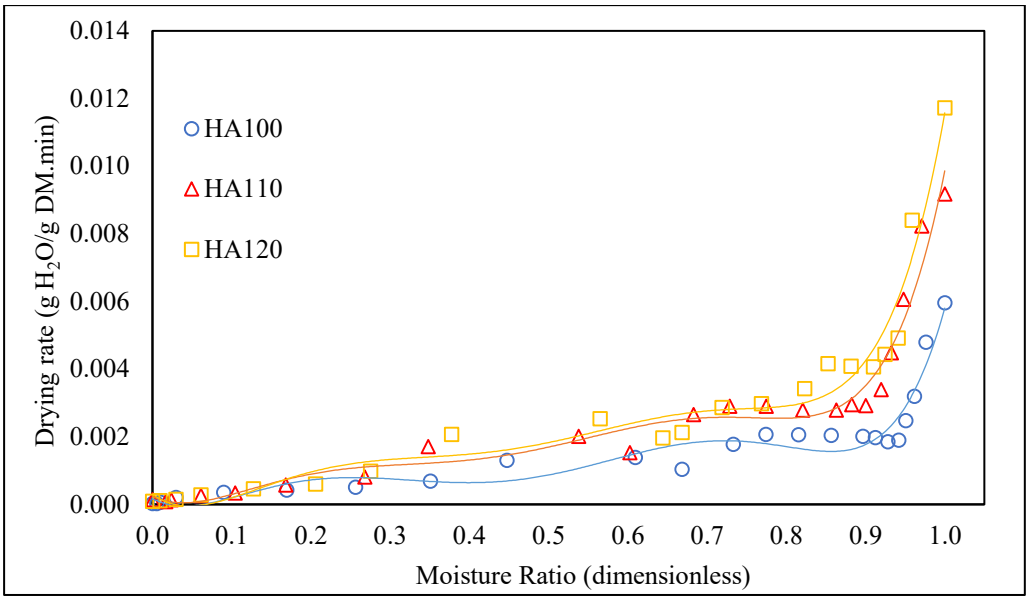


Figure 4.2: Drying rate curve (HA)

Figure 4.2 reveals that drying of crumb rubber does not have a constant drying rate period at the initial but exhibits falling rate period (FRP). In contrast to this finding, Sethu.S (1967) concluded that the drying of crumb rubber only occurred in two stages namely CRP, the first and second FRPs with about 10% of critical moisture content. In this study, the falling rate period consists of two distinct periods, namely first stage falling rate period followed by the second stage at a much slower FRP in between 0.9 to 0.6. However, some constant drying rate was observed in moisture ratio range in between 0.3 to 0.6. This phenomenon can be explained by the change of inter-granular structure of rubber which amplified the moisture diffusion rate at that certain period. Besides, the gas film mass transfer can be applied to explain the similar constant drying rate of rubber (Sethu.S, 1967). Slowest FRP detected at 0.3 when the rubber surface is completely dried and approaching EMC. The plane of evaporation slowly recedes from the surface and moisture transport was dominated by an activated diffusion mechanism. In this period, the amount of moisture removed and drying

rate is relatively small but the drying time is increased. Maskan (2001) suggested the existence of more than one FRP periods may be due to case hardening which acts as a barrier to moisture diffusion. It is an evidence of surface crusting whereby the heat transfer rate becomes slower and take time to reach wetly granulated rubber at the centre of the sample. Whilst, the drying rate curve also shows that HA at 110 °C and 120 °C gave comparable drying rate profile but HA at 100°C gave a lower drying rate as compared with 110 °C and 120 °C ($100^{\circ}\mathrm{C} \leq T_{air} \leq 120^{\circ}\mathrm{C}$).Findings demonstrated that HA120 had highest drying rate i.e. 0.0117 g H_2O/g DM. min by showing a greater reduction in moisture content in comparison with others HA drying method. Figure 4.3 shows a characteristic drying curve obtained from HA at three different temperatures ($27.09\% \leq X_d \leq 32.83$) where 2nd terms exponential equation (Equation 4.1) was found best fit the experimental data. Besides, the fitted curve depicted that all the moisture ratio, MR and dimensionless drying rate, f_n obtained from the different tested conditions, are equally close together, indicating that the effect of disparity in various conditions is small over the range tested.

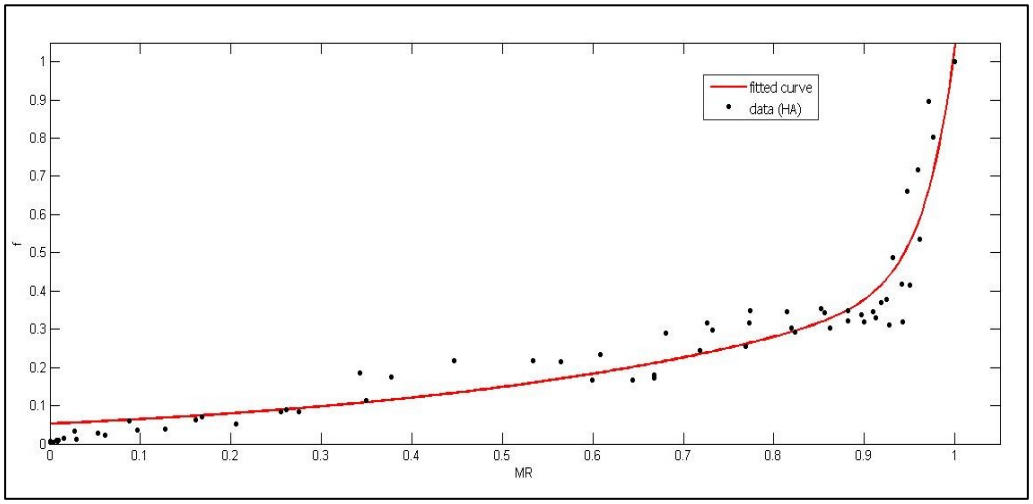


Figure 4.3: Characteristic drying curve of HA

$$f_n(\emptyset) = 2.602 \times 10^{-6} e^{(10.56\emptyset)} + 0.1718 e^{(0.767\emptyset)} \qquad R^2_{adj} = 0.9297 \qquad (4.1)$$

4.2.2 Single stage drying: MW drying

The variation of moisture ratio of MW with drying time was depicted in Figure 4.4. An obvious drying time saving can be observed in the first 10 to 15 minutes when the moisture content drastically reduced by more than 80% from its initial stage. In contrast, HA consumes ten folds of extra drying time to reach the similar moisture content. Owing to the drying mechanism of the MW, intramolecular gains energy by vibration and heat is generated internally. The difference in vapour pressure forces the water to migrate from the inside to the outside. During the first stage of the drying period, most of the MW energy supply is used to heat the sample. The temperature of the sample is rapidly raised up beyond 80°C in few minutes as can be seen from Figure 4.7. Hence, the moisture profile of MW dried- sample decreases faster than HA as shown in Figure 4.4. Meanwhile, MW600 has longer drying time due to lower MW power as compared to MW700 and MW800. The moisture content decreases faster in the case of higher MW power level due to the dipole substances in the specimen absorbed a great amount of MW energy. Besides, the drying rate of MW is higher than HA. As shown in Figure 4.5, the drying pattern follows the typical drying rate curve with maximum drying rate, 0.04 g H_2O/g DM. min. was achieved by 800W. Unlike HA, an initial transient period was observed in the beginning MW drying and followed by FRP thereafter. With a substantial amount of moisture content in the initial stage, these dipole molecules vigorously vibrate when it absorbs substantial microwave energy which generates heat inside the rubber causing a positive pressure that rapidly displaces the water to

the surface. The internal resistance of the rubber was lowered due to high energy content owned by the water molecules.

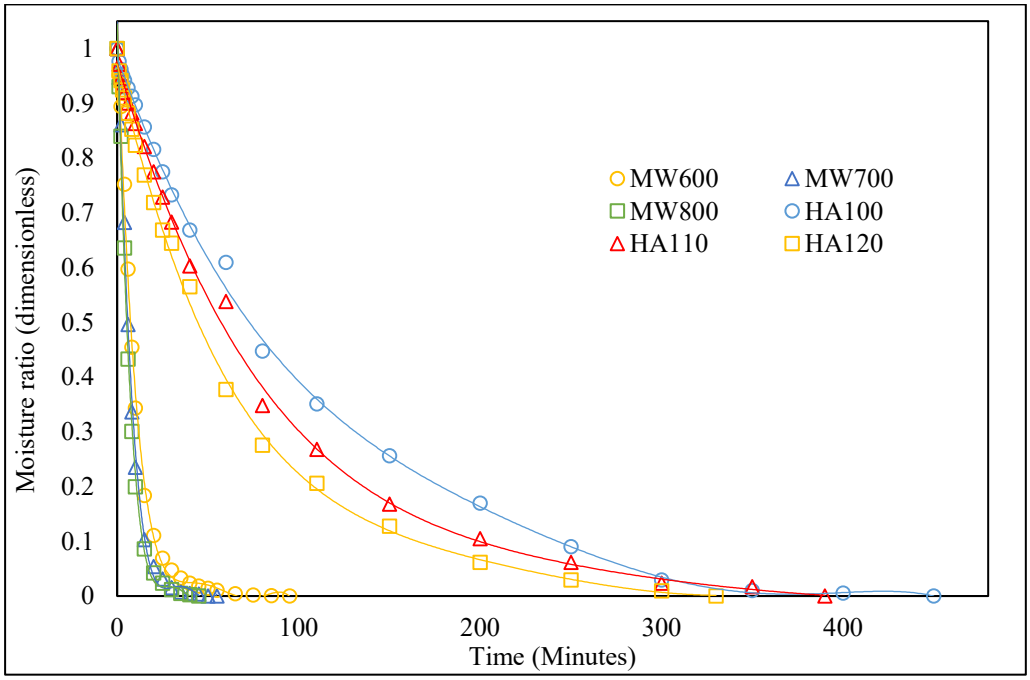


Figure 4.4: Moisture profile between HA and MW drying

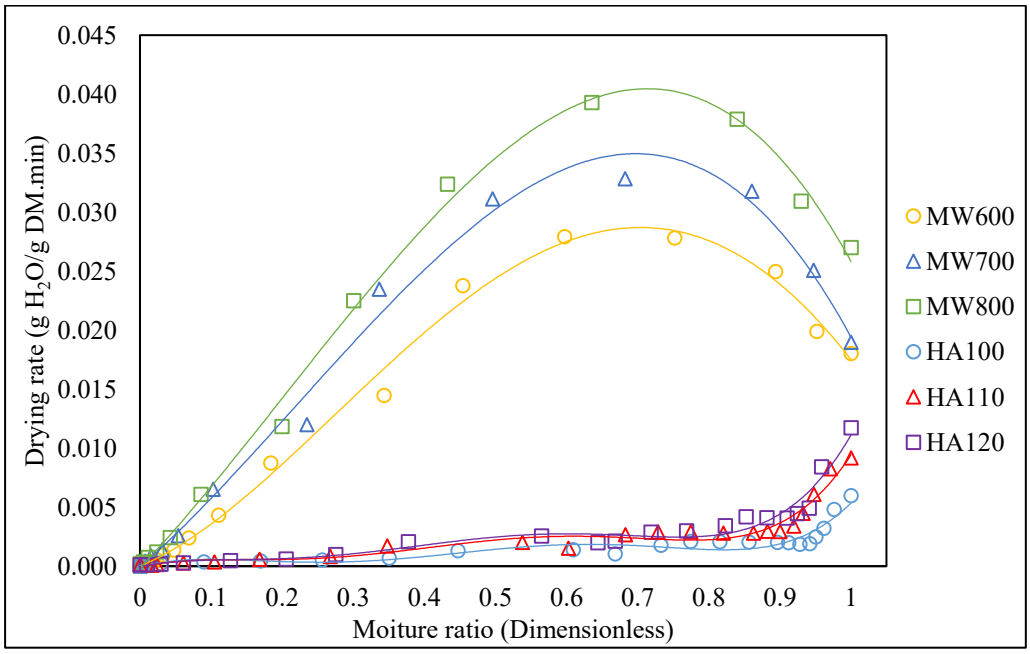


Figure 4.5: Drying rate curve (HA and MW drying)

Figure 4.5 shows that the FRP starts at MR of 0.70 which indicates the water transport is governed by diffusion. When the process nearly reaches the equilibrium moisture content (EMC), the reduction of water availability leads to a decrease of the absorbed MW power (Somsak Vongpradubchai, 2010). By comparing with HA, the average falling rate is higher in MW and this gives a significant drying rate that can save huge drying hours. Within the moisture content $41.45\% \leq X_d \leq 41.67$ the characteristic drying curve of MW can best be described by 3rd order polynomial model as shown in Equation 4.2

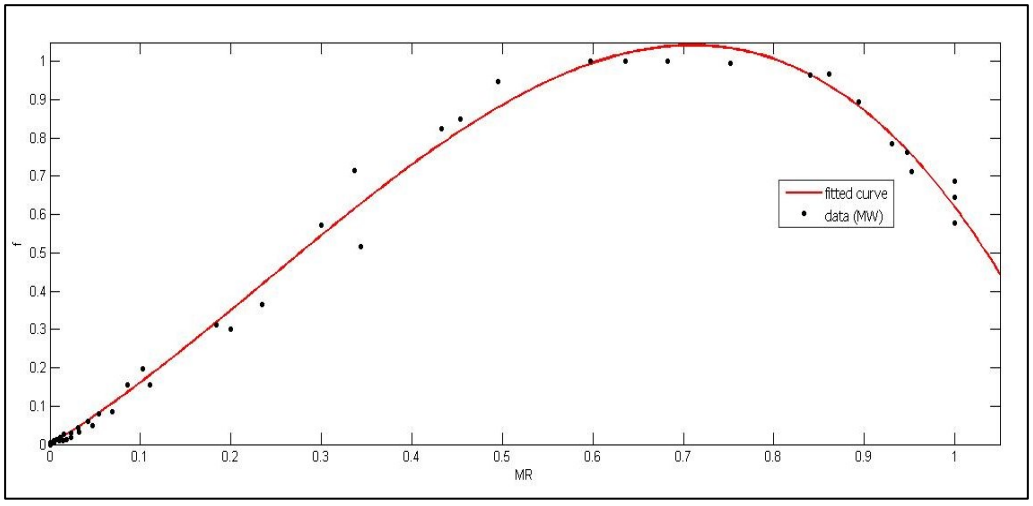


Figure 4.6: Characteristic drying curve of MW drying

$$f_n(\emptyset) = -2.957\emptyset^3 + 2.124\emptyset^2 + 1.455\emptyset - 0.002727 \quad R^2_{adj} = 0.9937 \qquad (4.2)$$

Noticeably, the MW drying times drastically reduced compared to HA, from 300 minutes for HA to 30 minutes for MW. This shows that MW drying can save a huge amount of drying time. In the case of HA, the surface dried faster than the interior which leads to the formation of a dry layer that creates resistance to the heat and mass transfer, hence resulting in a reduction of the drying rate. On the

other hand, thermal runaway might happen if the dipole molecules (e.g. water molecules) gain excessive energy and thus, the core temperature of the rubber may rise exponentially. Figure 4.7 shows the temperature profile during MW drying of crumb rubber. The temperature of the sample rose swiftly in the initial stage of drying, due to greater absorption of microwave energy, as the sample is loose at higher moisture content (Sharma, Prasad and Chahar, 2009). This has led to a higher drying rate due to higher moisture diffusion.

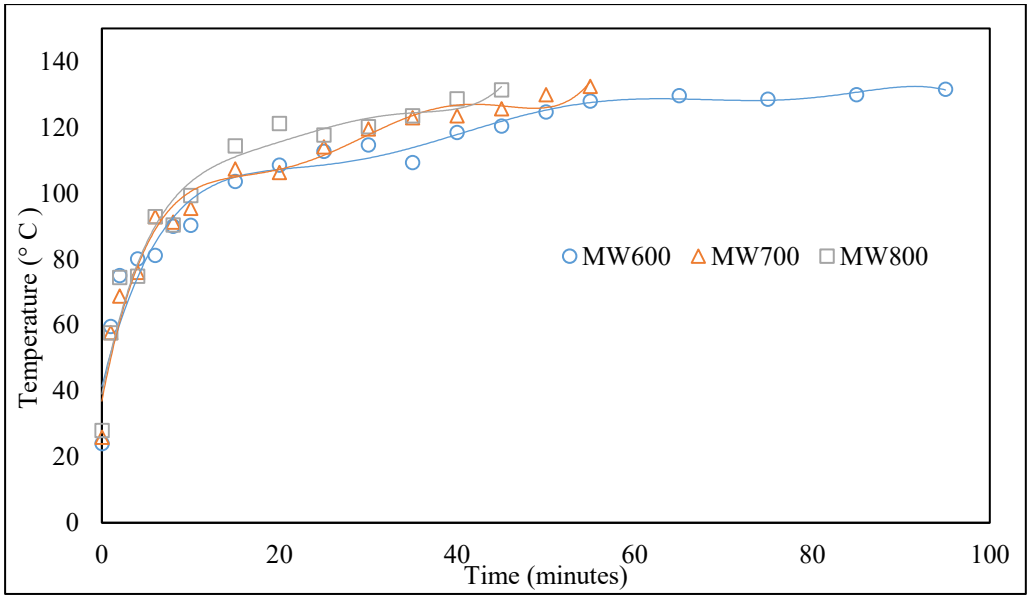


Figure 4.7: Variation of the core temperature of crumb rubber with drying time (MW drying)

Thermal runaway was detected at around 30 minutes during the experiment in which the sample temperature rose exponentially. This phenomenon occurred especially toward the end of the drying may be due to ionic losses becoming dominant over dipolar losses (Sharma, Prasad and Chahar, 2009). The dried material is prone to gelatinization if this uneven heating is persisting in the last stage of drying. Hence, the MW energy supplied must be reduced or stopped to

avoid deterioration of the product quality by causing charring (Nadia R. Pereira., Antonio Marsaioli Jr. and Ahrne, 2007). Besides, an increase in the product temperature also indicates the end of the drying process if the dry product has a high loss factor (Land, 2012).In fact, MW power is selectively reduced during drying to protect the product quality and equipment as well costs saving in energy. For this purpose, crumb rubbers were dried in step up (MW678) and step down (MW876) by varying MW power and drying times (5 to 15 minutes) and the result is depicted in Figure 4.8 and Figure 4.9, respectively.

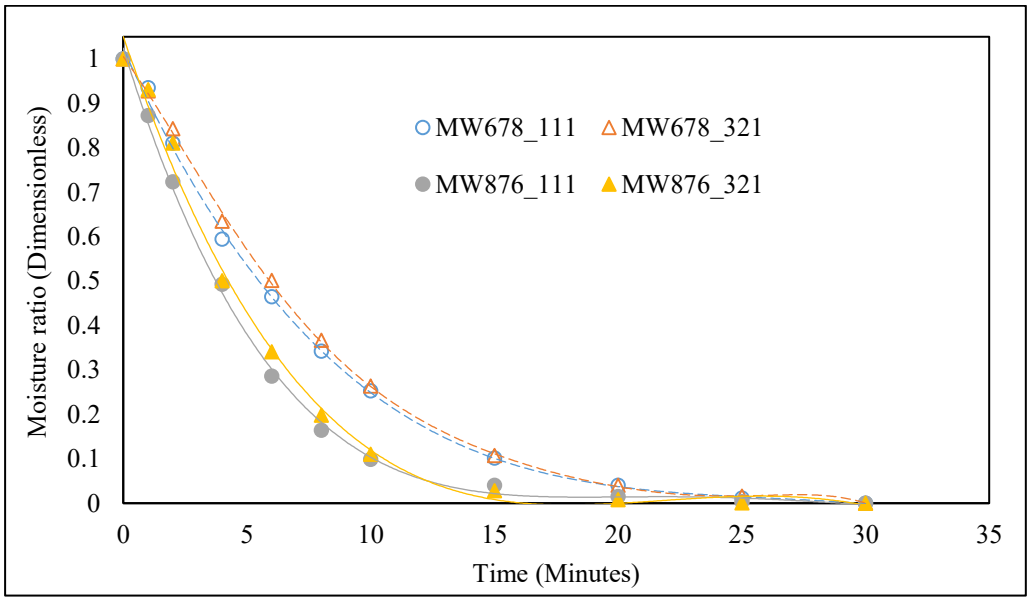


Figure 4.8: Variation of moisture ratio with drying time (MW_SU/SD)

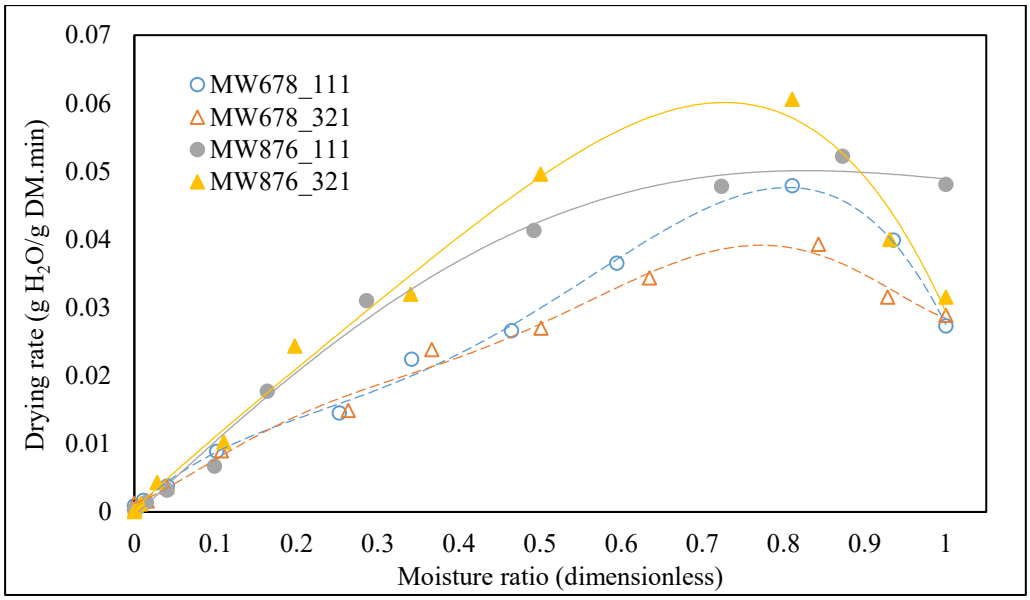


Figure 4.9: Variation of drying rate with moisture ratio (MW_SU/SD)

It was found that the moisture reduction rate of sample underwent step down MW was more prevalent than the step-up MW sample. A higher moisture removal at the first 15 minutes was contributed by MW800 which the sample gained extra energy than MW600 in aiding free moisture removal from the samples. Further, a longer drying period (15 minutes viz. 3 for the first stage) at higher power (MW876_321) significantly reduced the moisture ratio in the third falling rate period (after 15 minutes) in comparison to the MW876_111 which share similar drying period (10 minutes at each stage). The staging of MW power and drying interval could have influenced the rate of moisture removal. Figure 4.9 shows the variation of drying rate of applied drying methods. The finding revealed that the first 15 minutes of MW800 treatment in MW876_321 gave higher drying rate by 13.3% in compared to ten minutes of MW800 treatment in MW876_111. The extra drying time at high MW level i.e. MW876_321 resulted in sustainably high drying in the range of 0.049 -0.06 g H_2O/g DM. min during the first half of drying period. Meanwhile, both steps up drying of MW678_321

and MW678_111 exhibited lower drying rate. With consideration of similar power consumption and drying interval in both drying strategies, the step-down drying, MW876 is highly recommended over step up drying, MW678 due to its exceptional drying rate in producing dry product at a shorter duration.

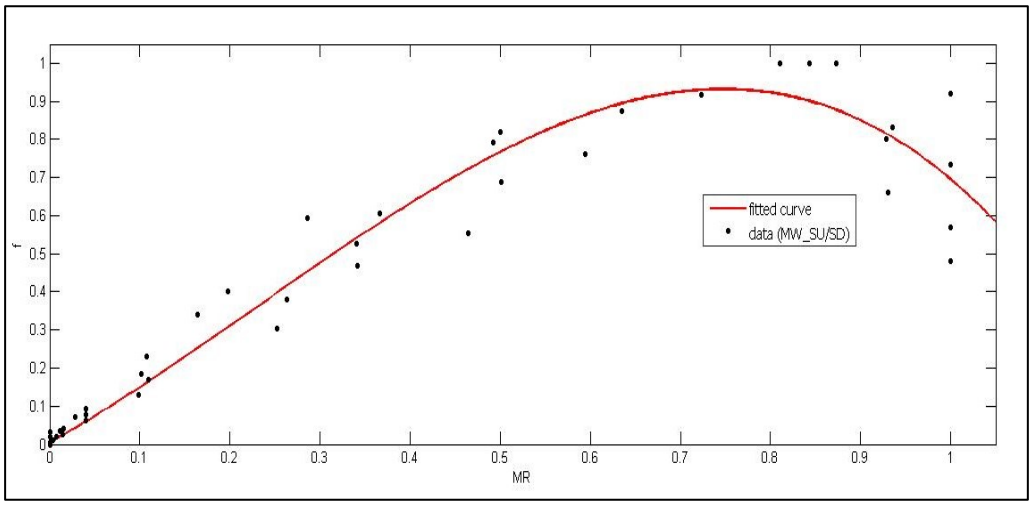


Figure 4.10: Characteristic drying curve of MW_SU/SD

Figure 4.10 shows the 3rd-degree polynomial model of MW with a step up/down function which was applied for rubber drying with moisture content ranges $38.38\% \leq X_d \leq 42.48$. It is apparent that the drying process of MW dried rubber involved two to three periods including accelerating period and FRP.

$$f_n(\emptyset) = -2.065\emptyset^3 + 1.428\emptyset^2 + 1.328\emptyset + 0.004181 \quad R^2_{adj} = 0.9714 \qquad (4.3)$$

4.2.3 Two-stage drying: HA/MW and MW/HA

Characteristics of two-stage drying involving HA and MW at different stage has been explained in the following Figure 4.11 and Figure 4.12, respectively. The

drying trend shows 3 distinguish stages of falling rate period. The sharp gradient in the first 15 minutes was due to the evaporation of free water from the surface. From 20 until 120 minutes, moisture was gradually eliminated from the samples. The mechanism in this phase involving the diffusional and capillary where moisture diffuse at a slower pace than before.

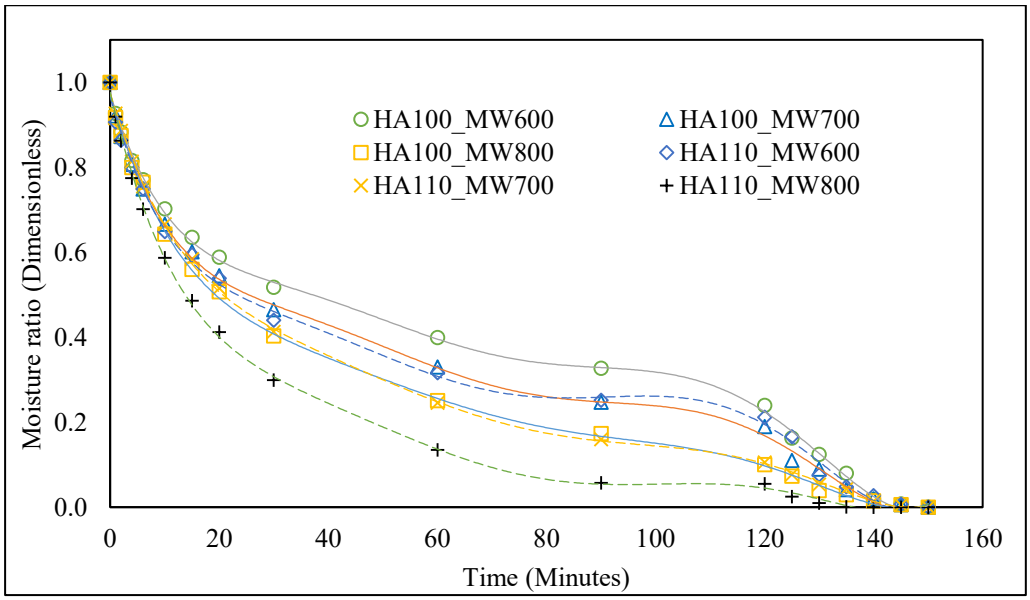


Figure 4.11: Variation of moisture ratio with drying time (HA/MW)

Indeed, HA only managed to remove 76.1% -94.5% of moisture even after two hours of drying. A sharp decrease of moisture started at 120 minutes when MW drying was brought into the drying system right after the HA drying. The change of drying mechanism induces rapid drying and this is obvious in the HA100_MW600 when moisture reduced from 0.239 to 0.20 in five minutes. Whereby HA110_MW800 reached EMC in 20 minutes earlier before the end of the drying. Maskan (2000) reported 64.3% of drying time-saving (from 482 to 172 minutes) when banana sample dried by microwave finish drying. This proves the effectiveness of MW when drying time is reduced and continues to decrease as the MW power increases.

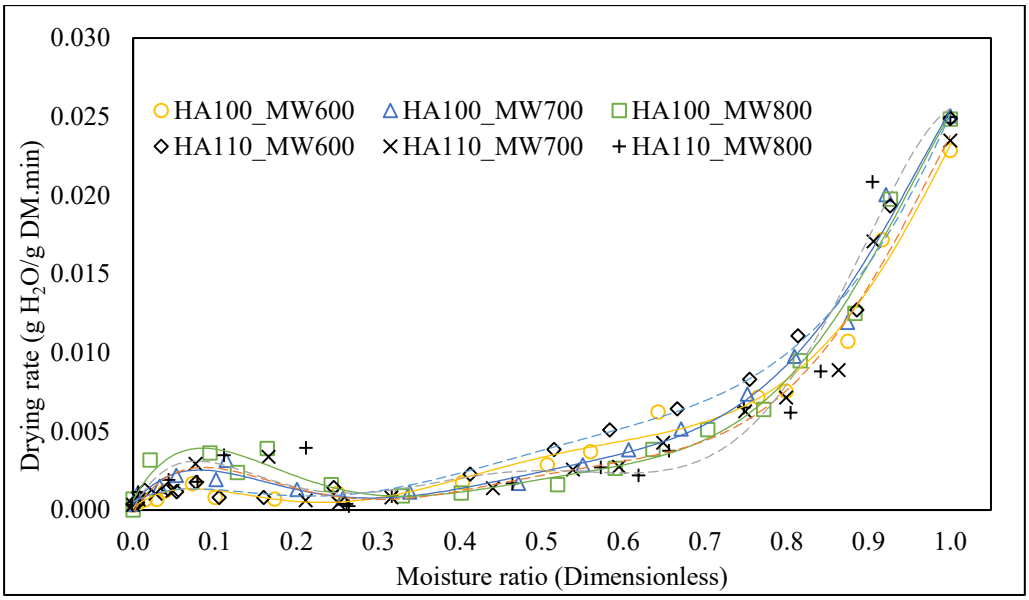


Figure 4.12: Variation of drying rate with moisture ratio (HA/MW)

Obviously, MW assisted drying technique has initiate higher drying rate in comparison with HA initiated drying strategy. Due to the poor heat and mass transfer of HA, the drying rate gradually decreased down the moisture ratio. The second peak occurred after the FRP in the region below 0.25 was due to the introduction of MW which generates sufficient heat for removal of excessive moisture trapped inside the rubber core. As can be seen from the Figure 4.12, highest drying rate recorded during the HA was at 0.025 g H_20/g DM. min, whereas 0.00394 g H_20/g DM. min was obtained by in MW drying region. Additionally, the characteristics drying curve of the HA/MW can be summarized in Figure 4.13. The Equation 4.4 was generated to best fit HA/MW samples with moisture content ranging from $25.72\% \leq X_d \leq 34.47$;$600\text{W} \leq MW \leq 800\text{W}$ and $100^{\circ}\text{C} \leq T_{air} \leq 110^{\circ}\text{C}$.

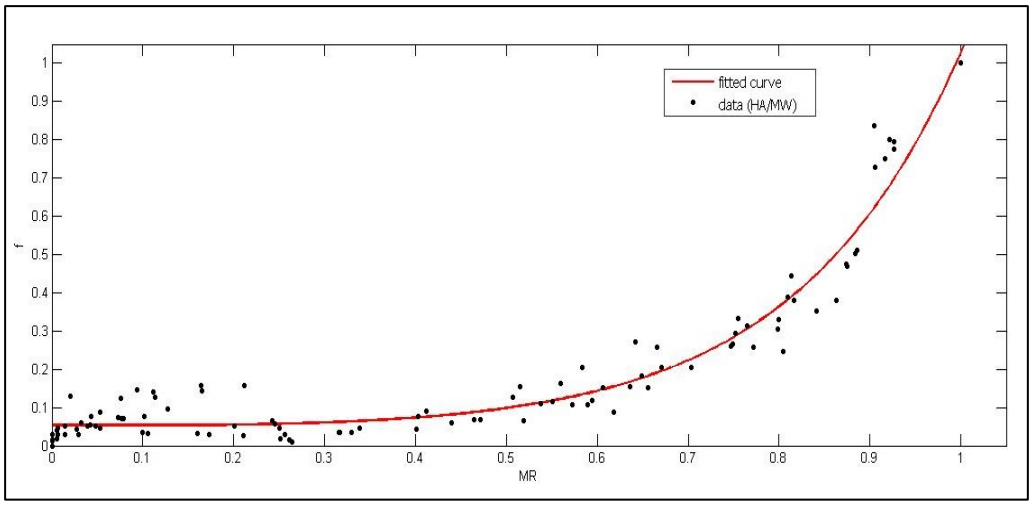


Figure 4.13: Characteristic drying curve of HA/MW

$$f_n(\emptyset) = 0.04961e^{(-0.8495\emptyset)} + 0.004258e^{(5.467\emptyset)} \qquad R^2_{adj} = 0.9611 \qquad (4.4)$$

Second drying technique was initiated with MW drying for 30 minutes and followed by 120 minutes of HA drying. Figure 4.14 depicted similar pattern as compared to Figure 4.4 in which steep gradients of moisture reduction were observed. Tirawanichakul *et al.* (2011) reported similar finding such that the cashew nut dried with MW following by HA has relatively short drying time comparing to the drying with HA alone. It is observed that 95.2%- 99.8% of moisture content was removed in the MW drying (first stage) which showing 5.6%-25.1% enhancement in moisture removal as compared to HA/MW. Nevertheless, the introduction of HA after MW has insignificant influence toward the moisture ratio partly because most of the moisture has been removed during the first stage drying. It was shown that some of the crumb rubber samples dried with MW800 almost reaching EMC even before the introduction of HA. Hence, it can be implied that almost 99% of the moisture was removed in MW phase while rest of 0.1% was drawn by HA.

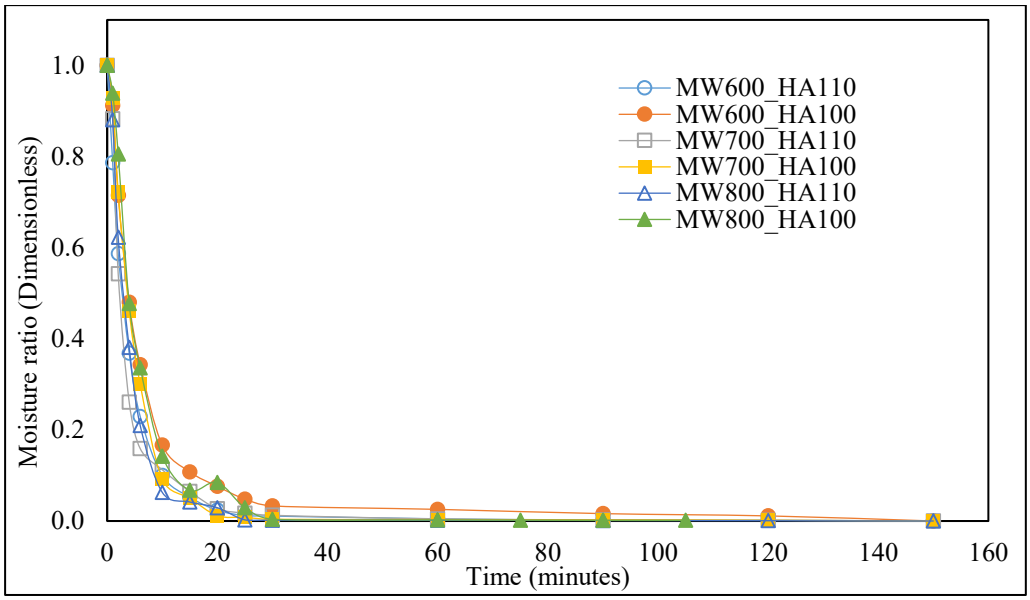


Figure 4.14: Variation of moisture ratio with drying time (MW/HA)

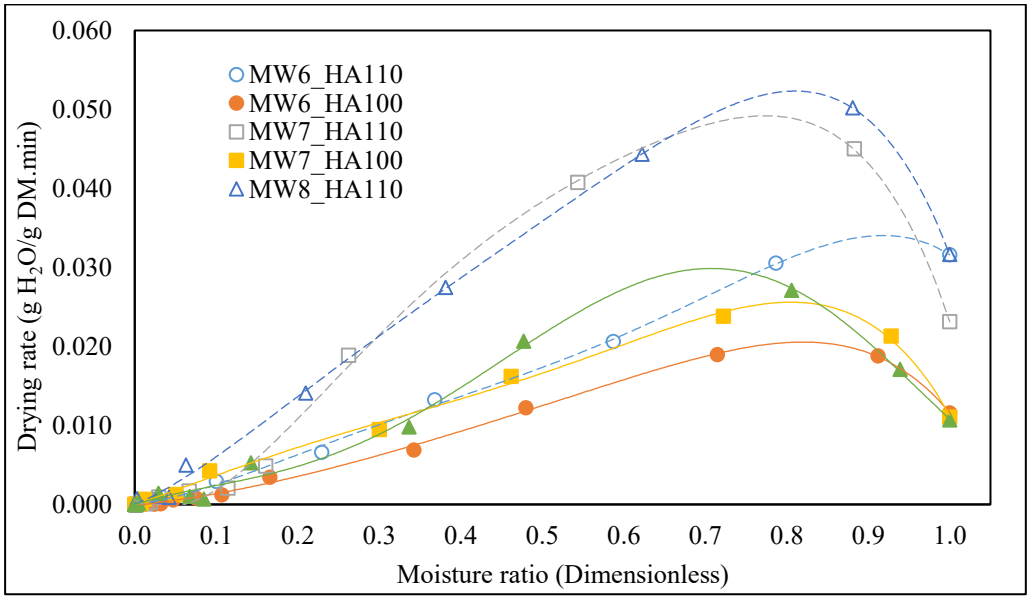


Figure 4.15: Variation of drying rate with moisture ratio (MW/HA)

By comparing to HA/MW, drying process begins with MW demonstrated significant improvement of drying rate. Two stages of drying rate are confirmed in MW/HA including a transient period at$(0.9 < \mathrm{MR} < 1.0)$, and ended with FRP. Meanwhile, drying rate curve in Figure 4.15 implied that no obvious

driving force in increasing the drying rate during the post-drying process by HA. Figure 4.16 shows the drying curve of MW/HA for the sample with moisture ratio $21.30\% \leq X_d \leq 26.81$, , $600\text{W} \leq MW \leq 800\text{W}$ and $100^{\circ}\text{C} \leq T_{air} \leq 110^{\circ}\text{C}$. Besides, 4th polynomial model as shown in Equation 4.5 was found to fit the best the dimensionless experimental data for MW/HA.

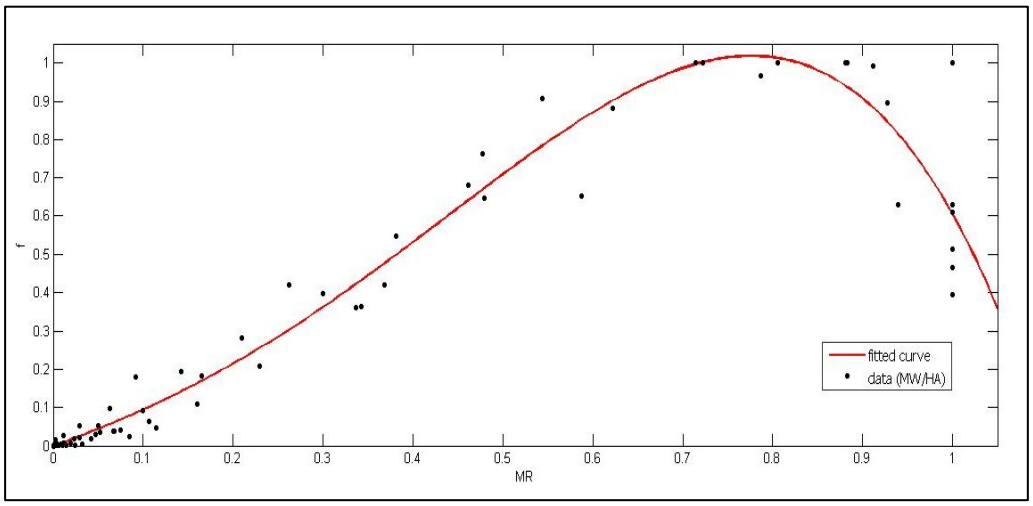


Figure 4.16: Characteristic drying curve of MW/HA

$$f_n(\emptyset) = -3.665\emptyset^4 + 2.78\emptyset^3 + 0.6153\emptyset^2 + 0.8754\emptyset - 0.001953 \quad R^2_{adj} = 0.9615 \qquad (4.5)$$

4.2.4 Two-stage drying: VD/MW and MW/VD

By referring to Figure 4.17, VD removed 69%-77% of its initial moisture for the first hour of drying when VD/MW was applied. Further to post drying by MW drying which rapidly displaces the moisture from the sample causing a sharp fall of moisture from a maximum 0.3 to 0.02 within five minutes. For MW/VD, MW drying has removed 98% of moisture in the first ten minutes and subsequently by VD until EMC reached. The MW drying contributes the most moisture removal in comparison with VD which demonstrates insignificant moisture

removal in the post-drying period. The similar finding was reported in 4.2.3 when MW drying is more dominant in moisture removal than HA in MW/HA.

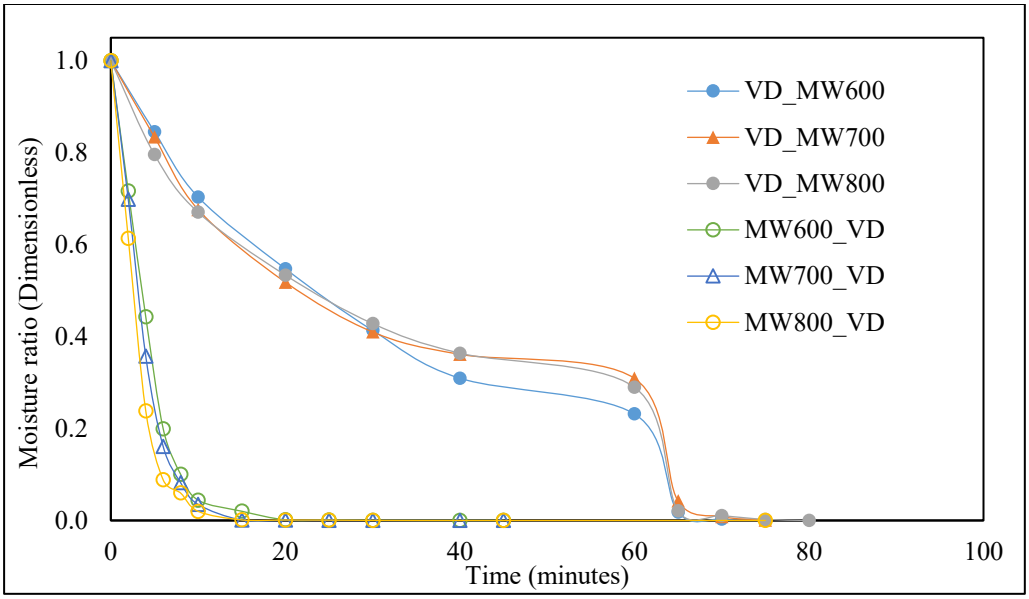


Figure 4.17: Variation of moisture ratio with drying time (VD/MW and MW/VD)

Figure 4.18 shows the drying rate of VD (in VD/MW) is gradually reduced in the range of 0.02 to 0.002 g H_2O/g DM. min (from 1.0 to 0.4) and it surges up as MW energy is introduced. It is clearly demonstrated that the drying rate of MW/VD surpassed VD/MW at moisture ratio between 1.0 and 0.1. The superimpose of MW and VD below the 0.2 regions explained the effectiveness of MW drying in retaining its drying momentum whereas VD has poorer drying performance. Clearly, VD is not recommended for single stage drying process due to its relatively weak drying efficiency unless it is assisted by other drying techniques such as MW or HA (Zhang *et al.*, 2006; Zielinska and Michalska, 2016).

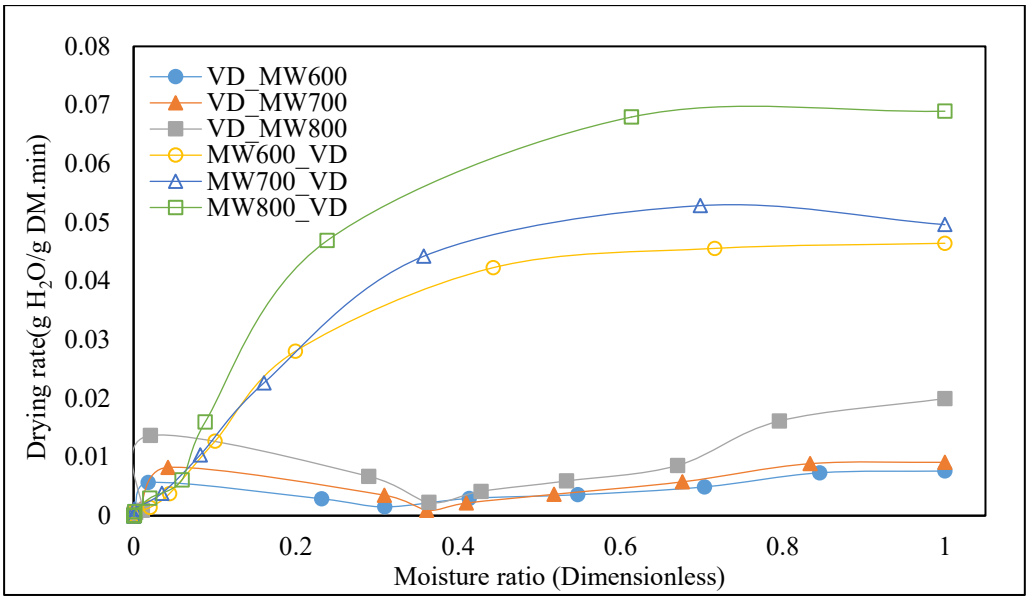


Figure 4.18: Variation of drying rate with moisture ratio (VD/MW and MW/VD)

Figure 4.19 and Figure 4.20 depicted the characteristic drying curve of VD/MW and MW/VD respectively. Experimental data obtained from VD/MW best fitted the 3rd order polynomial model (Equation 4.6) with a moisture content between $26.21\% \leq X_d \leq 49.90$; while MW/VD drying curve best fitted 2nd order polynomial model (Equation 4.7) for moisture content ranging from $35.21\% \leq X_d \leq 37.10$.

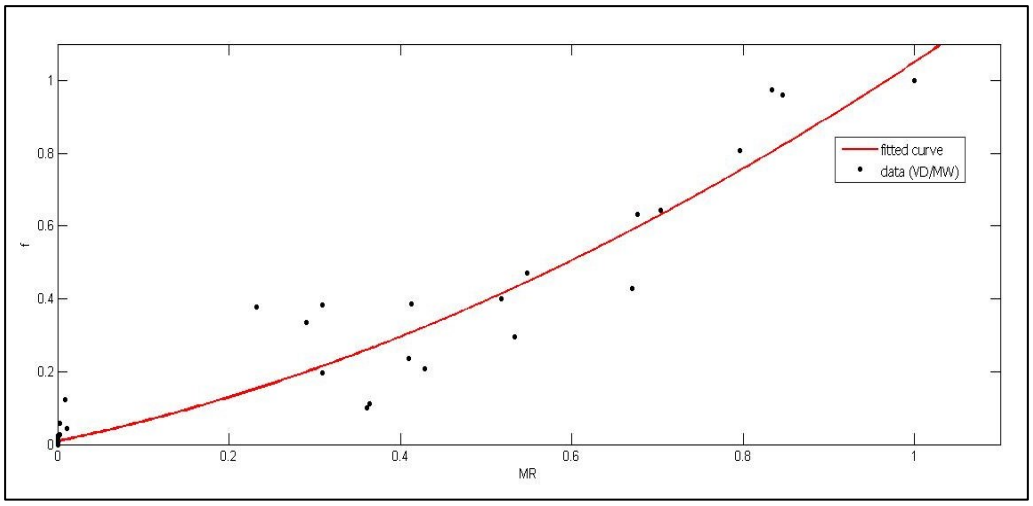


Figure 4.19: Characteristic drying curve of VD/MW

$$f_n(\emptyset) = -0.03572\emptyset^3 + 0.5884\emptyset^2 + 0.4883\emptyset + 0.008501 \qquad R^2_{adj} = 0.9468 \qquad (4.6)$$

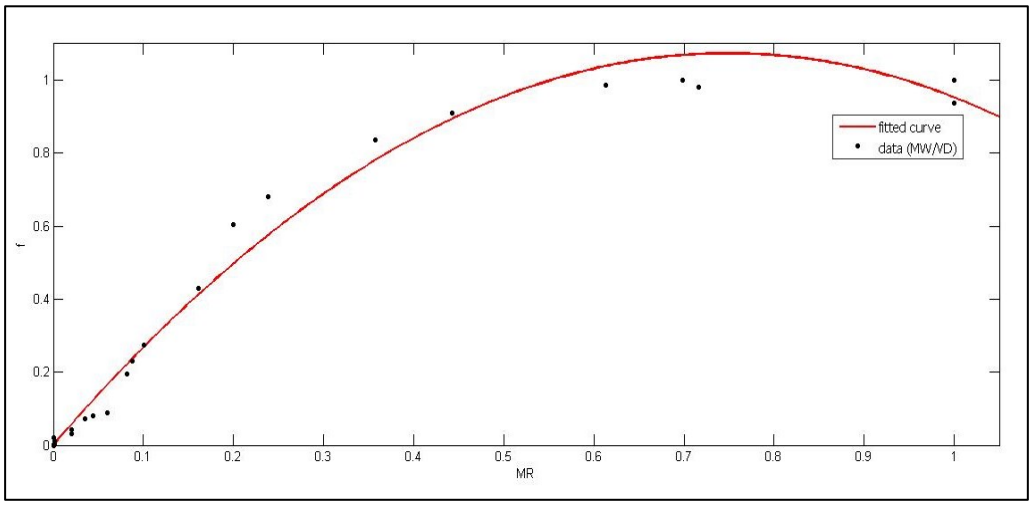


Figure 4.20: Characteristic drying curve of MW/VD

$$f_n(\emptyset) = -1.916\emptyset^2 + 2.869\emptyset - 0.0001793 \qquad R^2_{adj} = 0.9923 \qquad (4.7)$$

4.2.5 Hybrid drying: MWHA

The drying duration for MWHA methods is even shorter when compared with HA (- 89%) and MW (- 50%), respectively. This investigation revealed that MWHA can yield considerable gain in drying time by a factor of five or more. Several other researchers (Sharma and Prasad, 2006b; Vadivambal and Jayas, 2007; Jindarat *et al.*, 2011; Senapati *et al.*, 2014) have reported similar findings whereby time-saving can be achieved by introduction of air flow and MW energy in accelerating the water evaporation and diffusion through the water capillary. Figure 4.21 shows MW800HA 50V5.2 has faster moisture reduction than MW700HA50V5.2 and MW600HA50V5.2 due to the difference in power intensity.

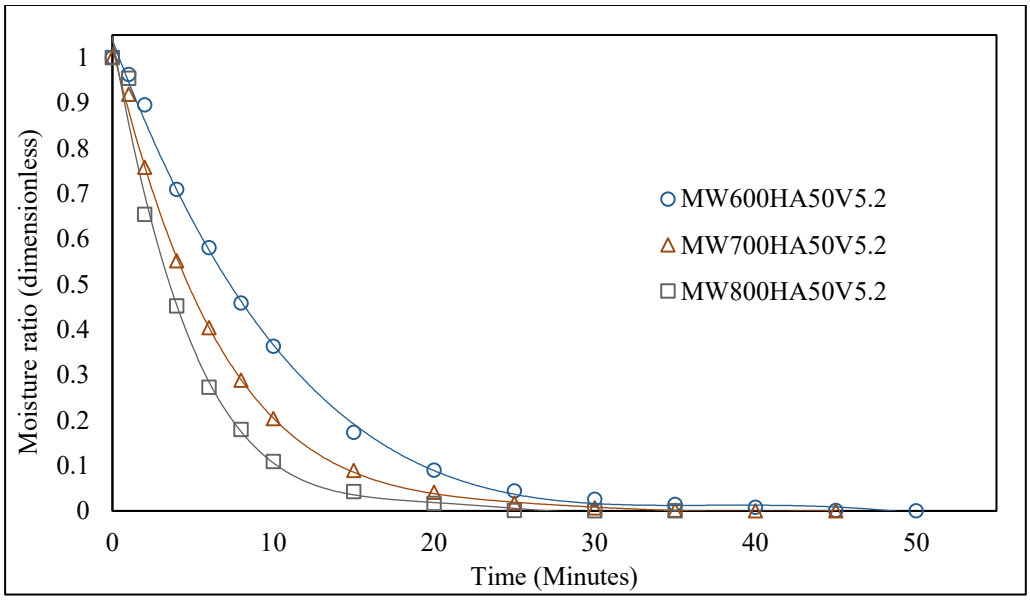


Figure 4.21: Variation of moisture ratio with time (MWHA50V5.2)

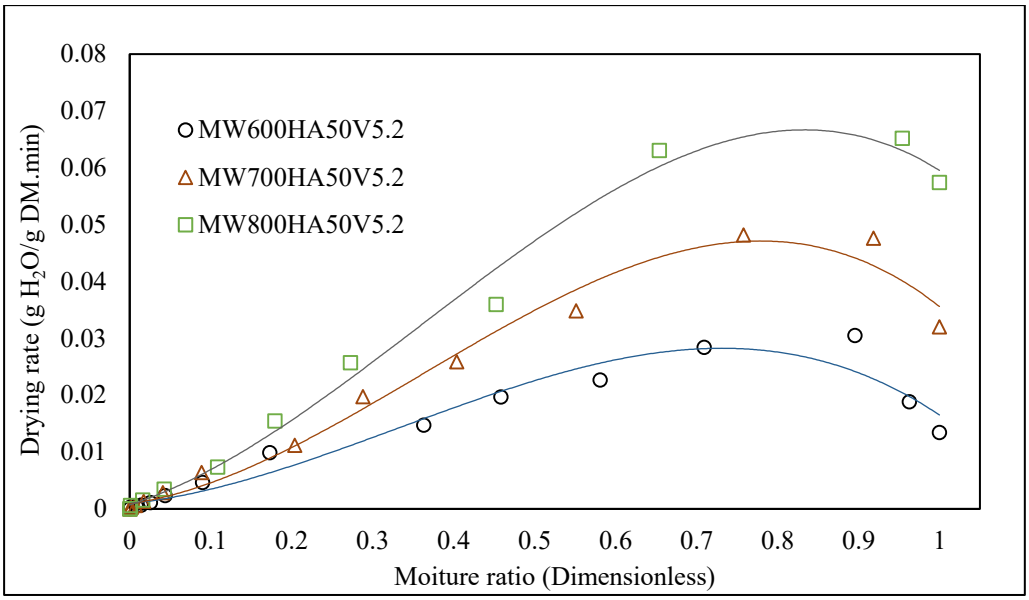


Figure 4.22: Changes in drying rate with using MWHA50V5.2

By referring to Figure 4.22, the drying process is initiated by transient period and followed by FRP whereby the rate of evaporation changes very little as the moisture content is reducing. The drying rate of MR between 0.9 to 0.7 was as high as 0.065 g H_2O/g DM. min by MW800HA50V5.2 and followed by second highest by MW700HA50V5.2 i.e. 0.047 g H_2O/g DM. min and lastly was 0.030 g H_2O/g DM. min by MW600HA50V5.2. FRP started at 0.65 and below in which migration of moisture from the inner interstices of each particle to the outer surface becomes the limiting factors that reduce the drying rate.

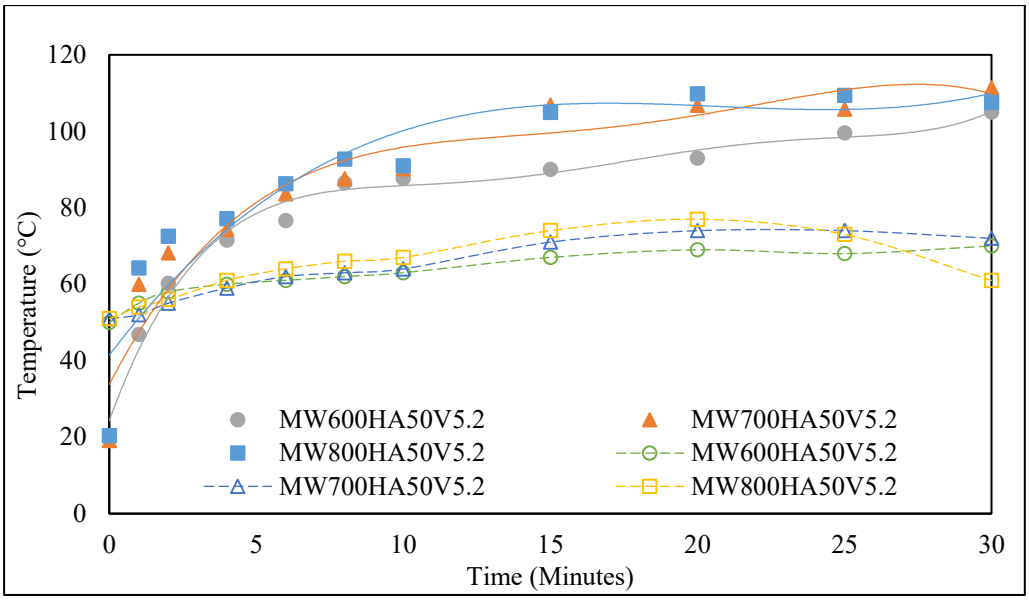


Figure 4.23: Changes in temperature of rubber sample (bolded line) and heater ambient temperature (dashed line) in the first 30 minutes by using the microwave-convective dryer.

The temperature profile of rubber samples underwent three different conditions were presented in Figure 4.23. Temperature gradient rose from 20-22° C to 47-65 ° C in first minutes of drying hour and gradually increase thereafter and surpassed 80 ° C after ten minutes. This temperature profile is supported by Figure 4.7, however with a lower core temperature in samples due to the rapid surface cooling by air flow introduced to the chamber. Additionally, a gradual increment of drying temperature of sample leads to enhancement of the drying rate as depicted in Figure 4.22. Both rubber samples of MW700HA50V5.2 and MW800HA50V5.2 have reached 100° C showed a significant dropped of moisture content in 15 minutes, however, MW600HA50V5.2 depicted relatively slow moisture removal due to a slow increase of temperature and hence lower drying rate. The temperature gauge of the heater has recorded a steady increase from 50 ° C to average of 70 ° C during the drying process. The increase in

temperature is due to the excessive heat released from the sample in the form of vapour which saturated the surrounding of the chamber. Figure 4.24 shows the characteristic of drying curve of MWHA50V5.2 dried samples with a moisture content between $41.33\% \leq X_d \leq 44.90$, can be estimated by using Equation 4.8. which obtained from 4th order polynomial model.

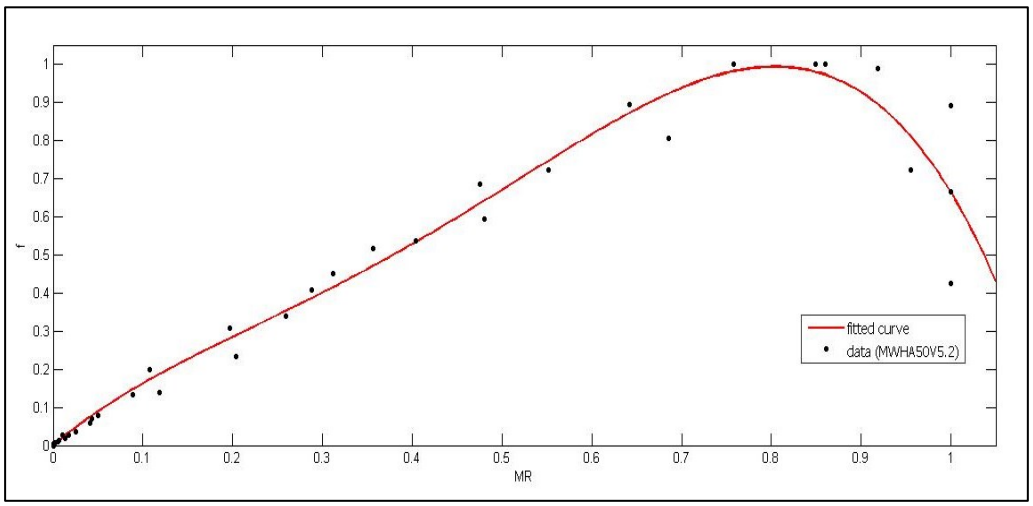


Figure 4.24: Characteristic drying curve of MWHA50V5.2

$$f_n(\emptyset) = -6.308\emptyset^4 + 9.38\emptyset^3 - 4.387\emptyset^2 + 1.978\emptyset - 0.000495 \qquad R^2_{adj} = 0.9798 \qquad (4.8)$$

4.2.5.1 Effect of air velocity in the drying characteristic of MWHA

During MW drying, the convective hot air aids as a carrier of the volatile compounds which continuously emitted from the being dried matter. In the first case study, the drying kinetics of MWHA50V5.2 were compared with MWHA50V2.8.Figure 4.25 revealed that both MWHA50V5.2 and MWHA50V2.8 exhibited a similar trend of moisture reduction in the first ten minutes of drying time. Soon after ten minutes, the moisture removal rate of

MWHA50V2.8 was found quicker than MWHA50V5.2. This can be explained by the reduced air velocity of 2.8 m/s which allow more heat build-up within the sample before it was removed from the air change cycle in the system to reduce the relative humidity inside the oven. Existing researchers reported that the drying rate can be improved by lowering the relative humidity of the convective air (Hongjie *et al.*, 2016). Likewise, if the hot air velocity is too large, the temperature of the surface of rubber will be reduced as well as the water evaporation and thus lower drying rate. An introduction of substantial air into the cavity lowers the surface temperature due to the evaporative and convective cooling effect (Jumah and Raghavan, 2001). Likewise, Sharma, Prasad and Chahar (2009) explained that high value of heat-transfer coefficient at higher velocity could resulting in quicker heat removal from the product which is at a higher temperature than the air. Figure 4.26 shows lower air velocity (2.8 m/s) yield higher temperature of the sample than the sample of higher velocity (5.2 m/s).

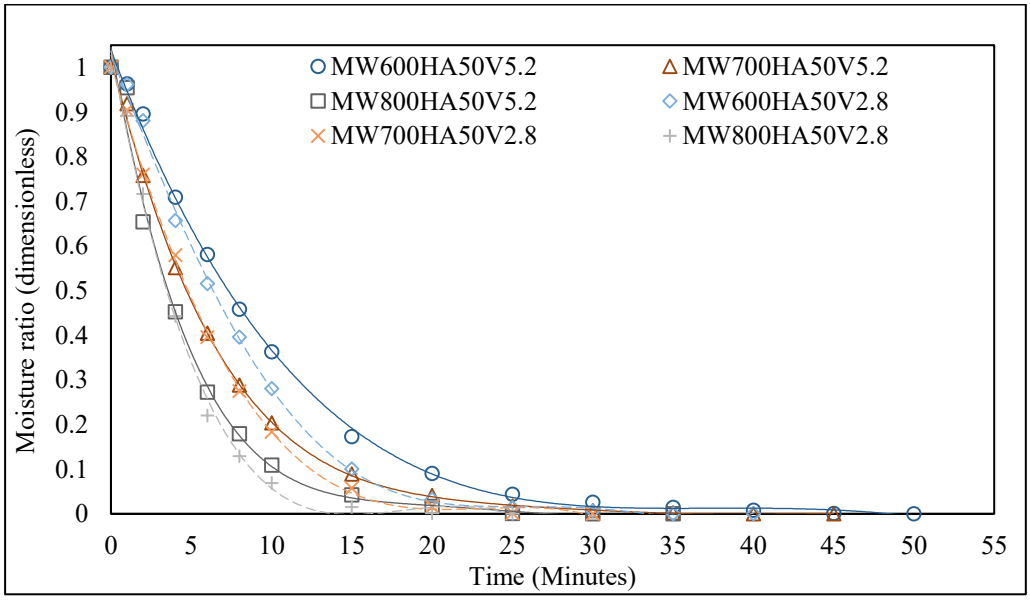


Figure 4.25: Variation of moisture ratio with time (MWHA50V5.2 vs MWHA50V2.8)

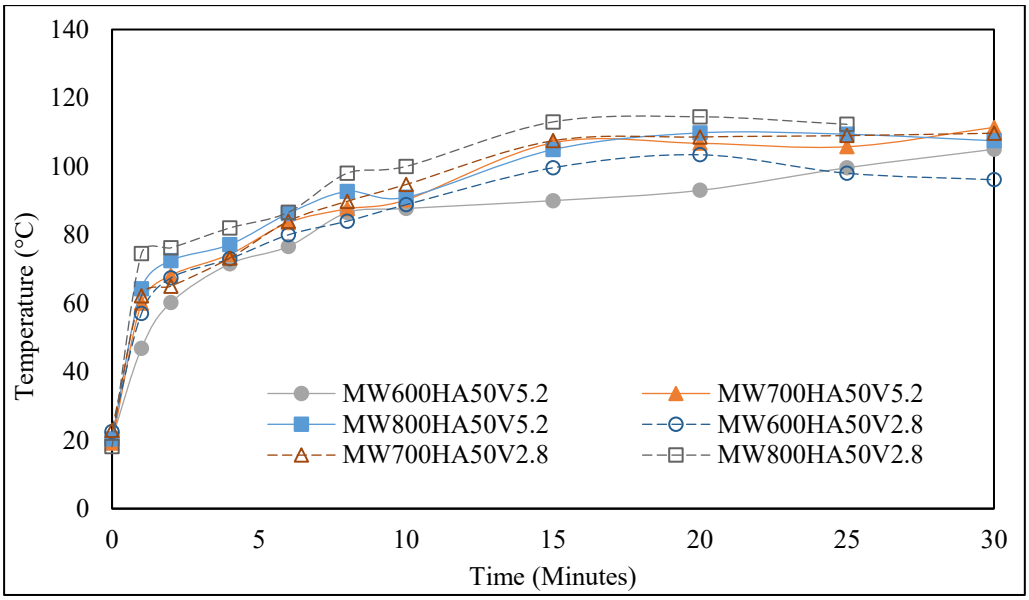


Figure 4.26: Effects of air velocity on rubber temperature (5.2m/s vs 2.8 m/s)

As shown in Figure 4.27, the drying rate value of MWHA50V2.8 was close to MWHA50V5.2. The peak drying rate obtained from MWHA50V5.2 ranging from 0.028 g H_2O/g DM. min to 0.065 g H_2O/g DM. min; whereas MWHA50V2.8 has a range of 0.033 g H_2O/g DM. min to 0.056 g H_2O/g DM. min for MW600, MW700, and MW800 respectively. It is interesting to note that some surpass of drying rate value between MW800HA50V2.8 and MW800HA50V5.2 at different moisture content which mainly caused by the increase of sample temperature when air change cycle inside the oven reduced at lower velocity.

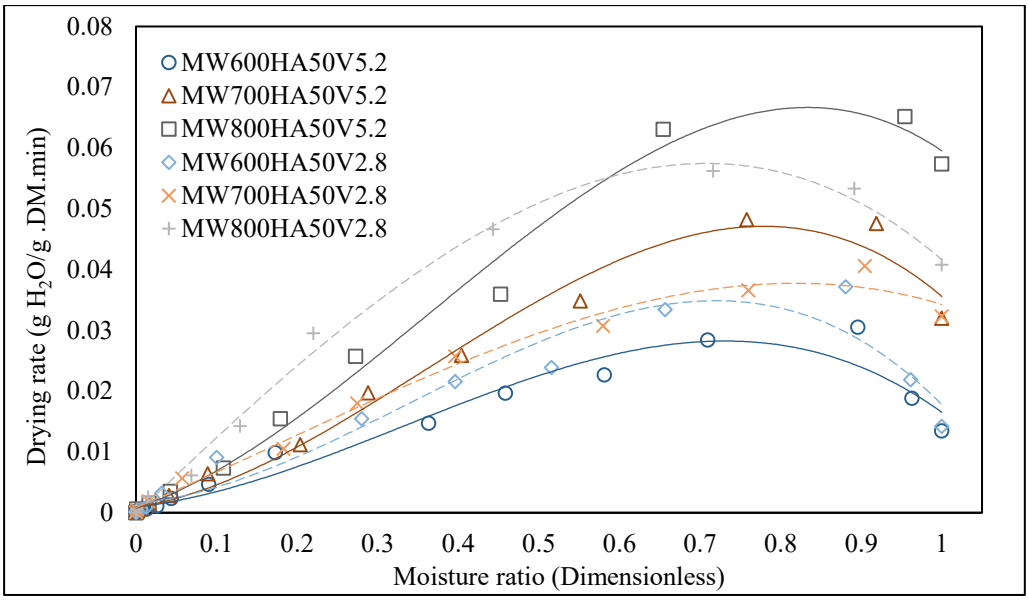


Figure 4.27: Changes in drying rate with moisture ratio (MWHA50V5.2 vs MWHA50V2.8)

Besides, it was observed that some dispersion of drying rate value occurred between MW600HA50V5.2 and MW600HA50V2.8 at the beginning of the drying. These dispersions can be explained by the variability of latex, which may not always have the same initial characteristic and or the unsteadiness of the characteristics of air especially the relative humidity (Naon, Berthomieu, Benet and Benoit, 1995). It might also due to the lack of field uniformity inside the cavity too. Similarly, the characteristic drying curve of MWHA50V2.8 ($34.44\% \leq X_d \leq 38.09$) shared similarity with MWHA50V5.2 and can be explained by 4th order polynomial model in Equation 4.9.

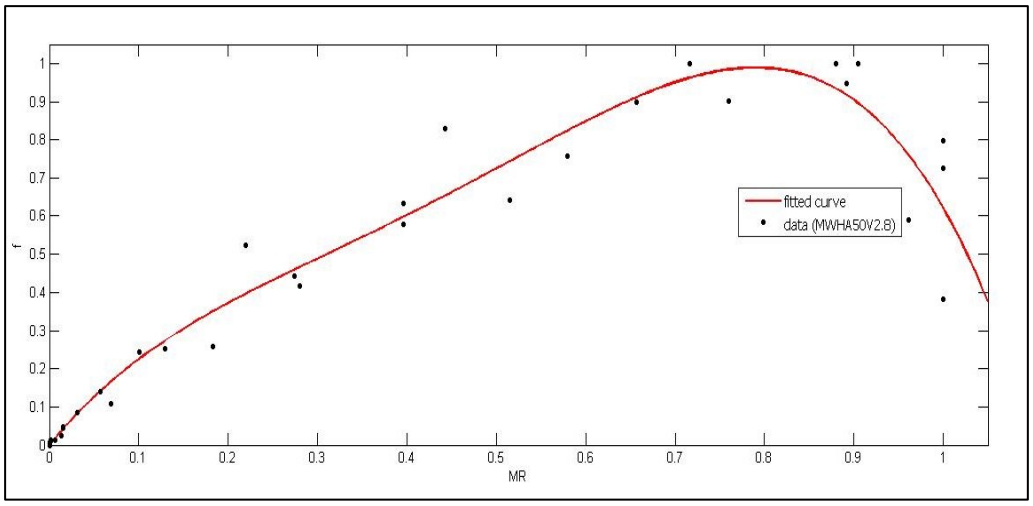


Figure 4.28: Characteristic drying curve of MWHA50V2.8

$$f_n(\emptyset) = -7.27\emptyset^4 + 12.0\emptyset^3 - 6.93\emptyset^2 + 2.824\emptyset - 0.0001539 \qquad R^2_{adj} = 0.9734 \qquad (4.9)$$

4.2.5.2 Effect of heater temperature in the drying characteristic of MWHA

Figure 4.29 depicted the variation of moisture ratio for MWHA50V2.8 and MWHA37V2.8. Heater temperature was controlled at 50 °C (high) and 37 °C (low), respectively to investigate the effect on its drying characteristic. The result shows high temperature led to a steeper gradient than low temperature because of faster free moisture removal from the rubber surface. Hence, the rise of temperature could help in enhancing the drying process. Furthermore, MW800 samples reached almost EMC in the 20 minutes whereas MW600 sample took extra five minutes to reach the same stage. These characteristic curves were closely approximated to the findings obtained from MW, MW/HA, and MWHA.

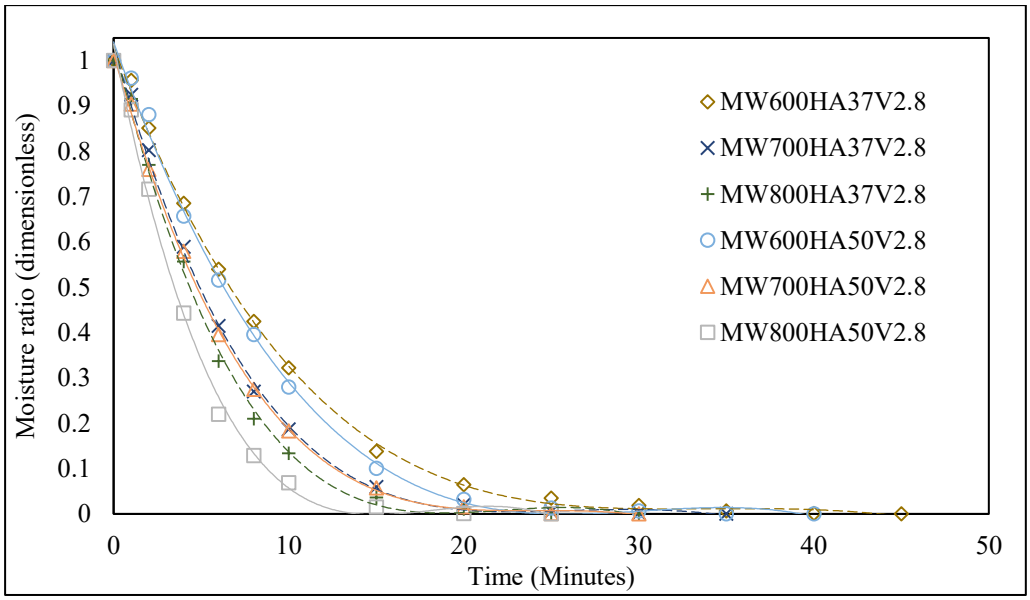


Figure 4.29: Variation of moisture ratio with time (MWHA50V2.8 vs MWHA37V2.8)

Drying rate curve in Figure 4.30 demonstrated that MWHA with high temperature has higher drying rate in comparison with MWHA at low temperature under the similar air velocity. Peak drying rate was observed at 0.044 g H_2O/g DM. min (MW800HA37V2.8) and 0.056 g H_2O/g DM. min (MW800HA50V5.2), respectively. High temperature promotes rapid water removal from the sample and thus higher drying rate is yield. In contrary, convective heat loss increased with decreasing air temperature. The results show the drying momentum sustain over the drying at moisture ratio of 0.9 down to 0.4 due to the powerful effects of MW. The difference of heater temperature did affect the drying rate of the sample; however, the effect is less significant if comparing to the variation in MW. Thus, the power level of MW still rules over the heat and mass transfer process and therefore enhance the overall drying performance.

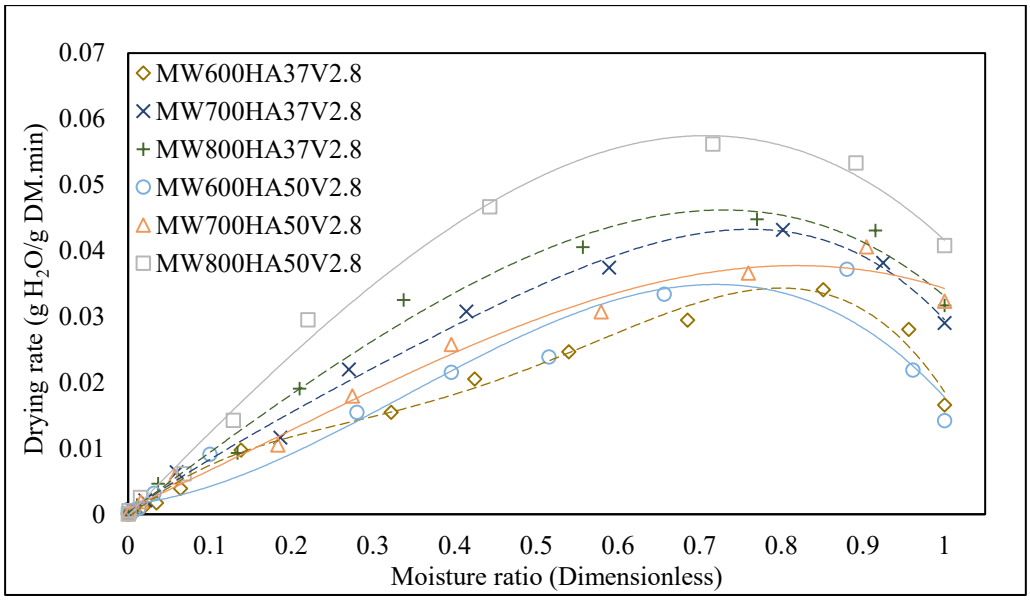


Figure 4.30: Variation of drying rate with moisture ratio (MWHA50V2.8 vs MWHA37V2.8)

Unlike the MWHA50V5.2 and MWHA50V2.8 which exhibited 4th order polynomial model, the experimental data of MWHA37V2.8 ($38.0\% \leq X_d \leq 39.28$) was found best fit with 3rd order polynomial model as shown in Equation 4.10.

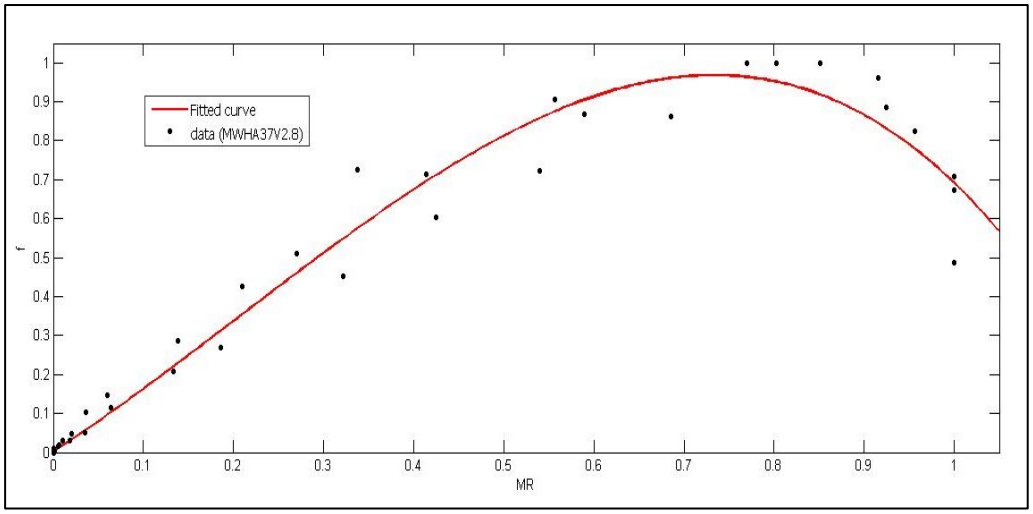


Figure 4.31: Characteristic drying curve of MWHA37V2.8

$$f_n(\emptyset) = -2.114\emptyset^3 + 1.306\emptyset^2 + 1.497\emptyset + 0.001826 \qquad R^2_{adj} = 0.983 \qquad (4.10)$$

Besides continuous hybrid drying, the MWHA also conducted in step up and down to investigate and relate the trend to individual strategies. In Figure 4.32, both step down methods e.g. MWHA876_111 and MWHA876_321 exhibited excellent moisture reduction than drying started with MW600 in either case. These findings were expected since the rate of moisture diffuse is directly proportional to the degree of MW power. Similar resulted were reported in Figure 4.8.

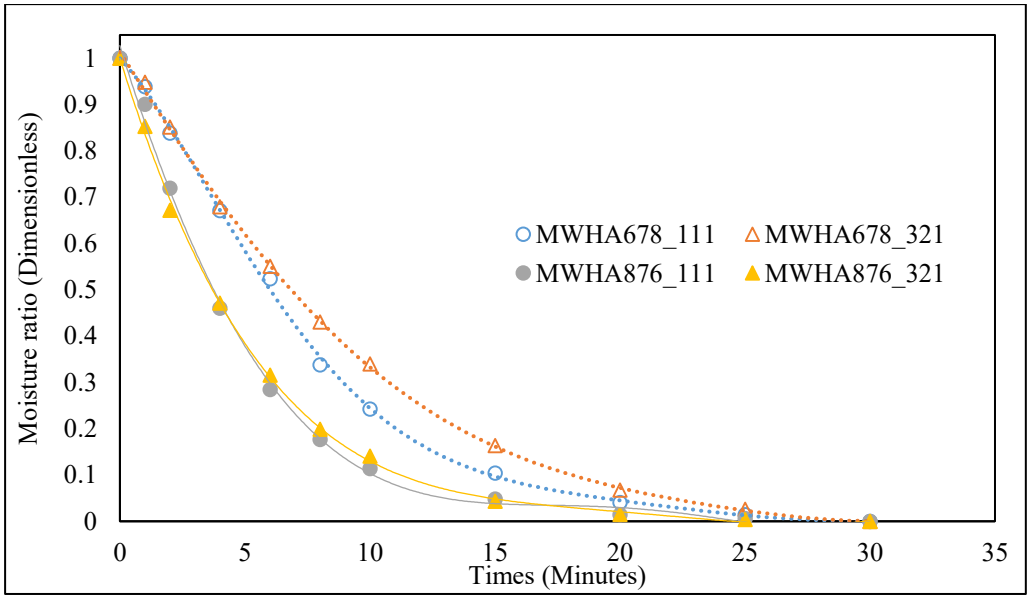


Figure 4.32: Variation of moisture ratio with time MWHA_SU/SD

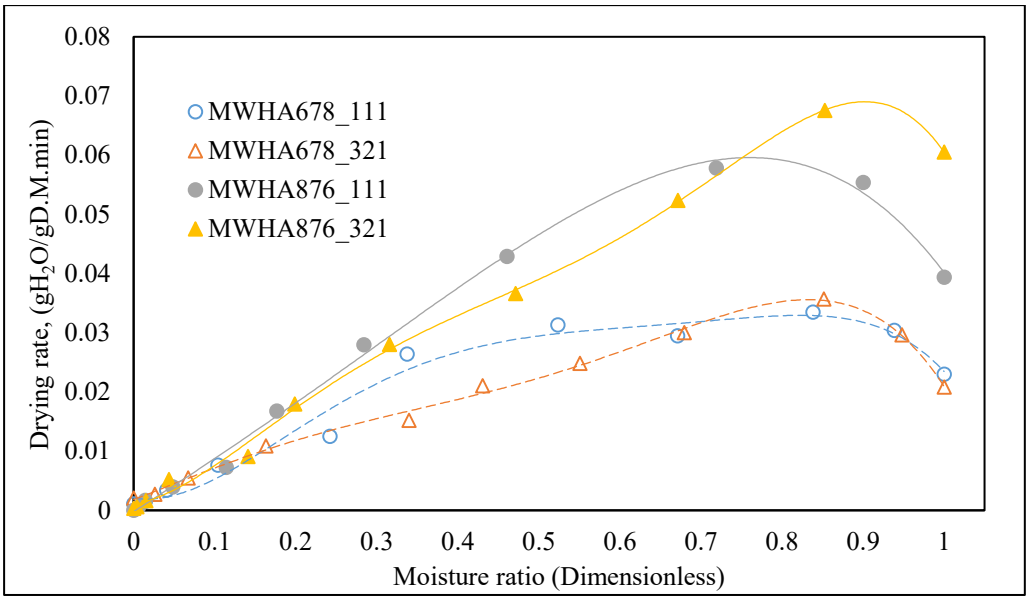


Figure 4.33: Variation of drying rate with moisture ratio MWHA_SU/SD

Maximum drying rate achieved by both step up and down of MWHA i.e. MWHA876_321 and MWHA678_321 were recorded at 0.067 g H_2O/g DM. min and 0.036 g H_2O/g DM. min, respectively. The drying rate gradually reduced thereafter after more than 50% of its moisture removed from the sample. Main cause contributes to this effect was indeed by the loss of energy absorption due to the lack of residue moisture and loss of energy dissipated to the environment. The characteristic of drying curve of MWHA with stepping up or down $(38.54\% \leq X_d \leq 42.16)$ in Figure 4.34 can be described by 3rd order polynomial model which fulfilled the Equation 4.11.

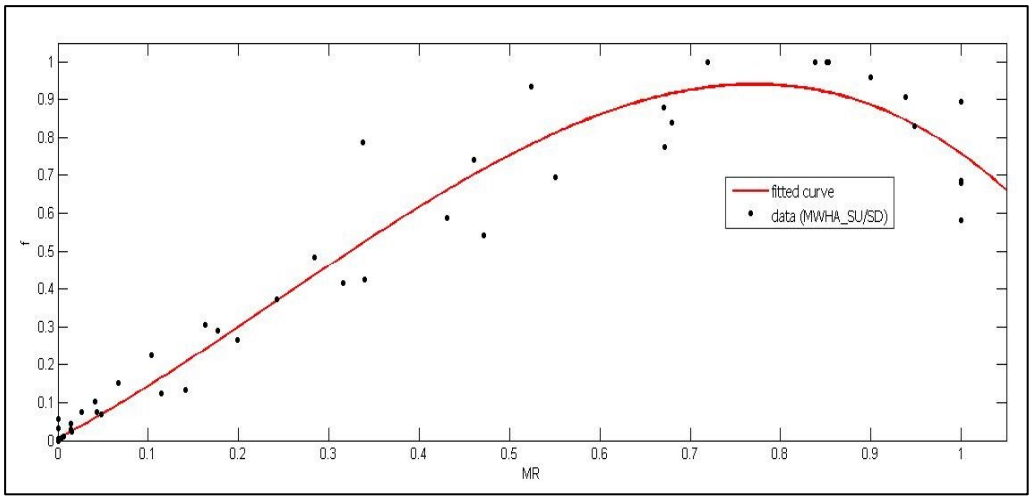


Figure 4.34: Characteristic drying curve of MWHA_SU/SD

3rd order polynomial model was found to best fit the dimensionless experimental data:

$$f_n(\emptyset) = -1.944\emptyset^3 + 1.425\emptyset^2 + 1.272\emptyset + 0.004173 \qquad R^2_{adj} = 0.9713 \quad (4.11)$$

4.3 Effective diffusivity coefficient of different drying methods

By referring to Figure 4.35, HA dried rubber has the lowest D_{eff} i.e. 0.22 x10^{-8} m^2/s among the drying strategies. Tirawanichakul and Tirawanichakul (2008), however, reported D_{eff} of crumb rubber was in the range of 1.0 x 10^{-9} to 2.15 x 10^{-5} m^2/s corresponding to HA drying temperatures between 40 and 150°C, respectively. In conventional HA drying, the heat is conducted from outer to the interior of the material and this cause the outer skin of the material get dried faster than the internal of the material. Indeed, this is unfavourable for drying process due to counter flow of the water diffuse out and heat enter to the core. Moreover, slow drying is observed due to poor water diffusivity.

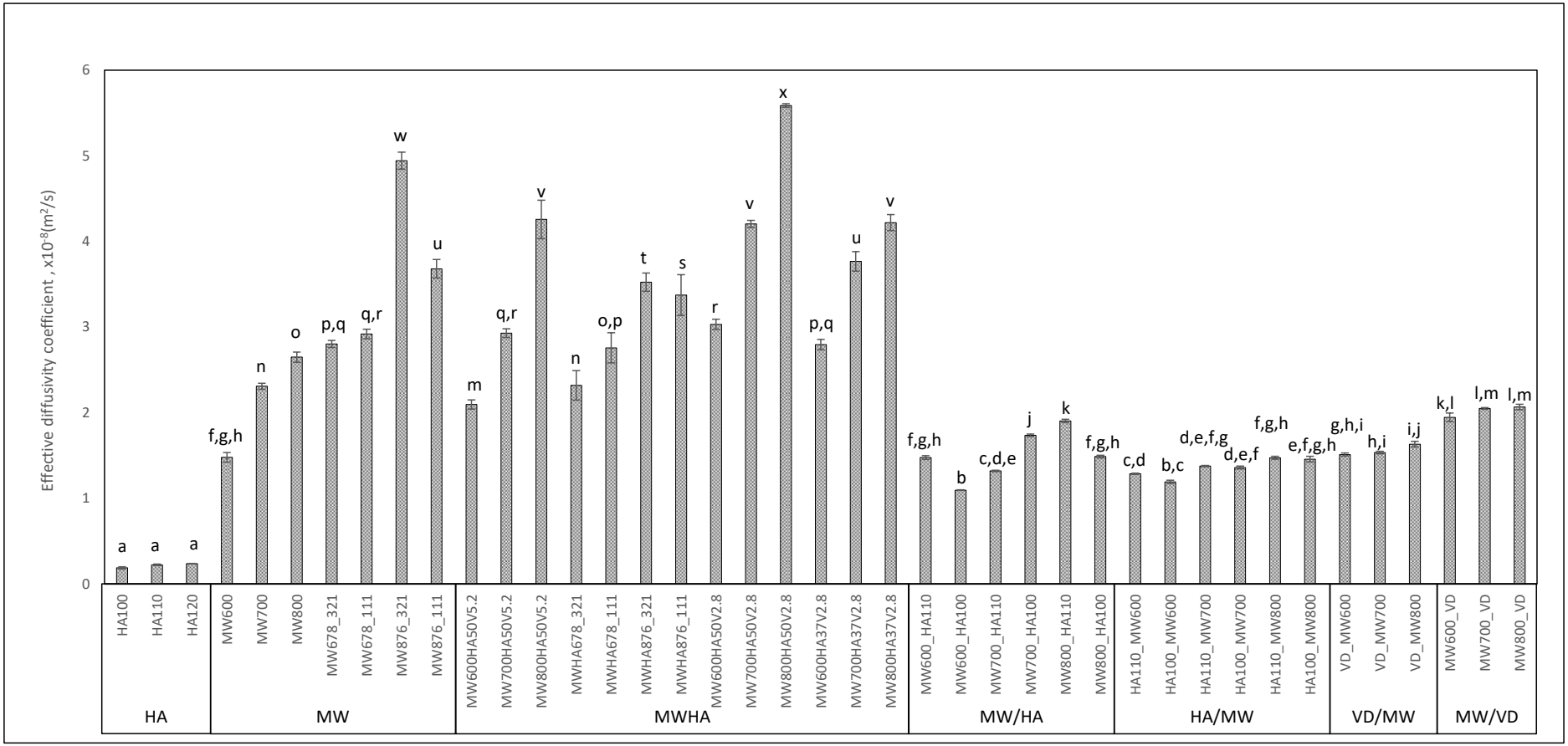


Figure 4.35: Effective diffusivity coefficient for dried rubber samples obtained for different drying strategies in various conditions

Due to low D_{eff} , long drying time is needed especially in the FRP before the drying ends. This may incur higher operation cost to the manufacturing process. Also, Pianroj *et al.* (2013) reported the D_{eff} of rubber sheets were measured at 2.50 x10^{-10} and 4.51 x10^{-10} m^2/hr for hot air drying at 40° C and 60 ° C respectively. Both MW drying and MWHA achieved exceptional D_{eff} with ranging from 1.48 x10^{-8} to 5.59 x10^{-8} m^2/s. Along with the increase of drying air temperature, it was observed that the increase of MW will causing the surge in D_{eff} too (Dadalı, Apar and Özbek, 2007; J.Varith. *et al.*, 2007). Thus, a MW power at 800W resulted higher effective moisture diffusivity than 700W and 600W. Similar trend was applied to temperature increment in leading to a higher D_{eff} too. D_{eff} of two- stage drying e.g. MW/HA, HA/MW and VD/MW were found in the range 1.09-1.90 x10^{-8} m^2/s. In this two-stage drying, D_{eff} was computed separately in each single stage and average results of both stages were used for comparison. These drying methods have slightly higher D_{eff} than HA alone due to the introduction of MW at initial or finish of drying. This is because MW energy able to supply sufficient potential energy to the water molecules to overcome the barrier and allow a rapid transfer along the capillary or surface evaporation. In contrast, both drying mechanism of VD and HA heavily depends on conduction or convection in the heat and mass transfer. On the other hand, MW/VD recorded approximate D_{eff} values among its three different conditions. It was found that vacuum assisted drying could have improve the D_{eff} of samples due to structural alteration of the sample which giving ample amount of pore for water removal (Reyes *et al.*, 2008; Rui, Min and Mujumdar, 2010).

A higher D_{eff} in the hybrid drying, where MW and HA were applied concurrently (MWHA) was supported by the rapid heat and mass transfer of water molecules that travelled in the same direction whereas forced heated air provides surface cooling on the rubber's surface allow the expedited of mass transfer. These combination effect of MW and HA in the drying systems therefore yielding higher diffusivity in average than the rubber dried in MW and the rest of drying methods.

4.4 Kinetic study of drying characteristic and mathematical modelling

The experimental data were fitted to ten different drying models derived from semi theoretical and empirical models as demonstrated in Section 2.5.2 to elaborate and forecast the moisture loss of crumb rubber during the drying process under different strategies. By simplifying Fick's second law and considering the relationship between the moisture ratio of dry material and drying time, the optimal drying mathematical models were tabulated and summarized in Table 4.1. In overview, the statistical parameters approximation showed that R^2,χ^2 and RMSE values ranged from 0.9529 to 0.9999, 0.0020 to 0.7098, and 0.0028 to 0.0730, respectively. Based on the model having the highest R^2 value, lowest χ^2 and RMSE, it can be concluded that 73 % of the samples confirmed Hii and Law model gave better results than other models. In other word, there were a very good agreement between the experimental and predicted moisture ratio values. This empirical model was adequately defined the drying characteristic found in NR dried with HA, MW, HA/MW, VD/MW and MWHA. The finding contrasted with that of crumb rubber dried by hot air fixe-bed drying (HA) where Logarithmic model was found best fitted to the

experimental data (Tirawanichakul and Tirawanichakul, 2008). It was interesting to note that all MW-assisted drying (except the MW/HA) can be optimally described via Hii and Law model. The Hii and Law model which adopted from combination of the Page and Two terms models has also been suggested to best describe the drying behaviour of cocoa (Hii, Law and Cloke, 2009), pumpkin (Onwude *et al.*, 2016), natural rubber sheet(ADS) (Dejchanchaiwong *et al.*, 2016),persimmon (Muhammad Hanif. *et al.*, 2015) and carrot pomace (Kumar, Sarkar and Sharma, 2012). Despite the dominance of Hii and Law model over the rubber drying, it was found that 10% of the samples including MW/HA, MW/VD and, some MWHA adapted to Midili *et al*., and Two-term exponential models, respectively. Nevertheless, both Modified Henderson and Pabis model and Two-term model can be applied to MW/HA samples which represents 7% of the total drying sample. In a nut shell, Hii and Law model best illustrates the drying characteristics of hybrid drying in respect of moisture content and drying time. The input from this thin layer drying model may be useful in predicting the thick layer drying behaviour which might serves as a useful guideline for the design of rubber drying process.

Table 4.1: Optimal results of statistical analysis on the modelling of moisture contents and drying time for the respective drying method of dried rubber. (Note: *a, b, c, g, h, k* and *n* are empirical constant for respective model)

Methods	Drying conditions	Optimal models	Constants												R^2	χ^2	RMSE
HA	100° C	Hii and Law	k=	0.0028	a=	0.8563	c=	0.1245	n=	1.2203	g=	0.0284			0.9984	0.0449	0.0146
	110° C		k=	0.0092	a=	0.9412	c=	0.0353	n=	1.0466	g=	0.0734			0.9981	0.0262	0.0160
	120° C		k=	0.0146	a=	0.9616	c=	0.0379	n=	0.9982	g=	1.0172			0.9988	0.0198	0.0124
MW	600W	Hii and Law	k=	0.0690	a=	0.5458	c=	0.4615	n=	1.4312	g=	0.0194			0.9991	0.3842	0.0106
	700W		k=	0.0428	a=	0.8110	c=	0.1833	n=	1.5052	g=	0.0078			0.9999	0.0040	0.0040
	800W		k=	0.0705	a=	0.8198	c=	0.1757	n=	1.4795	g=	0.0175			0.9999	0.0020	0.0037
	678_111		k=	0.0983	a=	1.0096	c=	0.0000	n=	1.1605	g=	0.6778			0.9991	0.0099	0.0106
	678_321		k=	0.0750	a=	0.9963	c=	0.0055	n=	1.2499	g=	0.6837			0.9997	0.0060	0.0066
	876_111		k=	0.1333	a=	0.9986	c=	0.0000	n=	1.2374	g=	0.6918			0.9994	0.0506	0.0091
	876_321		k=	0.0958	a=	1.0126	c=	0.0000	n=	1.3708	g=	0.6922			0.9987	0.0127	0.0134
HA/MW	100° C /600W	Hii and Law	k=	0.0079	a=	0.6548	c=	0.3187	n=	1.1811	g=	0.1095			0.9959	0.1287	0.0225
	110° C /600W		k=	0.0162	a=	0.7451	c=	0.2433	n=	1.0413	g=	0.1728			0.9969	0.1230	0.0196
	100° C /700W		k=	0.0043	a=	0.6457	c=	0.3234	n=	1.3251	g=	0.1112			0.9931	0.2553	0.0294
	110° C /700W		k=	0.0002	a=	0.6132	c=	0.3276	n=	1.9056	g=	0.0241			0.9706	0.6171	0.0606
	100° C /800W		k=	0.0002	a=	0.6284	c=	0.3145	n=	1.9044	g=	0.0246			0.9805	0.2988	0.0470
	110° C /800W		k=	0.0008	a=	0.6502	c=	0.2961	n=	1.6320	g=	0.0613			0.9750	0.6049	0.0559
MW/HA	600W /100° C	Modified Henderson & Pabis	k=	0.2010	a=	0.4166	c=	0.5563	b=	0.0633	g=	0.0181	h=	0.2010	0.9968	0.0173	0.0192
	600W /110° C		k=	0.2321	a=	0.3810	c=	0.5225	b=	0.1019	g=	0.0800	h=	0.3282	0.9997	0.2470	0.0055
	700W /100° C	Midilli	k=	0.1505	a=	1.0269	b=	0.0000	n=	1.1799					0.9979	0.0658	0.0164
	700W /110° C	Two term	k=	0.0344	a=	0.0636	c=	0.9908	g=	0.3358					0.9858	0.0644	0.0390
	800W /100° C	Midilli	k=	0.1272	a=	1.0301	b=	0.0001	n=	1.2009					0.9930	0.7098	0.0303
	800W /110° C		k=	0.1882	a=	1.0125	b=	0.0000	n=	1.1895					0.9968	0.5185	0.0192
VD/MW	100 mbar&80° C /600W	Hii and Law	k=	0.0040	a=	0.8475	c=	0.1528	n=	1.5258	g=	0.6663			0.9766	0.3427	0.0530
	100 mbar&80° C /700W		k=	0.0049	a=	0.8237	c=	0.1764	n=	1.4438	g=	0.6610			0.9529	0.4811	0.0730
	100 mbar&80° C /800W		k=	0.0016	a=	0.7641	c=	0.2360	n=	1.7134	g=	0.6554			0.9596	0.4314	0.0672
MW/VD	600W/100 mbar&80° C	Two term exponential	k=	0.3585	a=	1.9754									0.9987	0.0217	0.0115
	700W/100 mbar&80° C		k=	0.4096	a=	2.0267									0.9999	0.0112	0.0028
	800W/100 mbar&80° C		k=	0.5159	a=	2.0691									0.9992	0.3024	0.0085
MWHA	600W/HA50V5.2	Hii and Law	k=	0.0602	a=	1.0127	c=	0.0000	n=	1.2386	g=	0.2873			0.9995	0.0199	0.0083
	700W/HA50V5.2		k=	0.1375	a=	0.2791	c=	0.7253	n=	1.2931	g=	0.0599			0.9996	0.0026	0.0071
	800W/HA50V5.2		k=	0.1231	a=	0.8988	c=	0.1026	n=	1.2017	g=	0.6767			0.9996	0.0033	0.0071
	600W/HA37V2.8		k=	0.0391	a=	0.8622	c=	0.1437	n=	1.4042	g=	0.2215			0.9997	0.0241	0.0066
	700W/HA37V2.8		k=	0.0914	a=	1.0043	c=	0.0000	n=	1.2710	g=	0.5383			0.9999	0.0022	0.0041
	800W/HA37V2.8		k=	0.1000	a=	1.0027	c=	0.0000	n=	1.3139	g=	0.6540			0.9995	0.0078	0.0084
	600W/HA50V2.8		k=	0.0622	a=	1.0105	c=	0.0000	n=	1.3236	g=	0.5393			0.9991	0.0073	0.0113
	700W/HA50V2.8		k=	0.0623	a=	0.8730	c=	0.1298	n=	1.4024	g=	0.5485			0.9997	0.0027	0.0064
	800W/HA50V2.8		k=	0.1310	a=	1.0050	c=	0.0000	n=	1.3374	g=	0.4453			0.9994	0.0116	0.0088
	678_111	Two term exponential	k=	0.1919	a=	1.8819									0.9989	0.0096	0.0122
	678_321	Hii and Law	k=	0.0229	a=	0.7822	c=	0.2191	n=	1.5615	g=	0.1964			0.9999	0.0079	0.0034
	876_111	Midilli	k=	0.1465	a=	1.0127	b=	0.0003	n=	1.1974					0.9989	0.0272	0.0120
	876_321	Hii and Law	k=	0.0745	a=	0.7375	c=	0.2651	n=	1.3576	g=	0.5058			0.9996	0.0077	0.0068

4.5 Specific energy consumption of various rubber drying systems

For every drying process, the selection of an efficient drying system is essential to minimize the energy usage and to save the operating cost. SEC is one of the effective indices to gauge the energy consumption in each individual drying system. Figure 4.36 shows the SEC values obtained from a single stage, two-stage and hybrid drying of crumb rubber samples. It was found that two-stage drying of both VD/MW and MW/VD gave higher SEC in average in comparing to others drying strategies. The highest SEC was recorded at 0.75 MJ/g H_2O by VD_MW600 and followed by VD_MW700 which measured at 0.68 MJ/g H_2O. However, SEC of VD/MW reduced when the MW power rating decreases. The SEC values dropped to 0.43-0.52 MJ/g H_2O when 800W was selected for the drying in VD/MW and MW/VD. On the other hand, both MW600_VD and MW700_VD shared insignificant difference in terms of SEC values ($p > 0.05$). The average high value of SEC for both VD/MW and MW/VD was in fact due to the substantial power consumption of vacuum drying system i.e. 2.4kWh in comparison to other types of the dryer. Likewise, both MW/HA and HA/MW has slightly higher due to the involvement of two different dryers in the two-stage drying process.

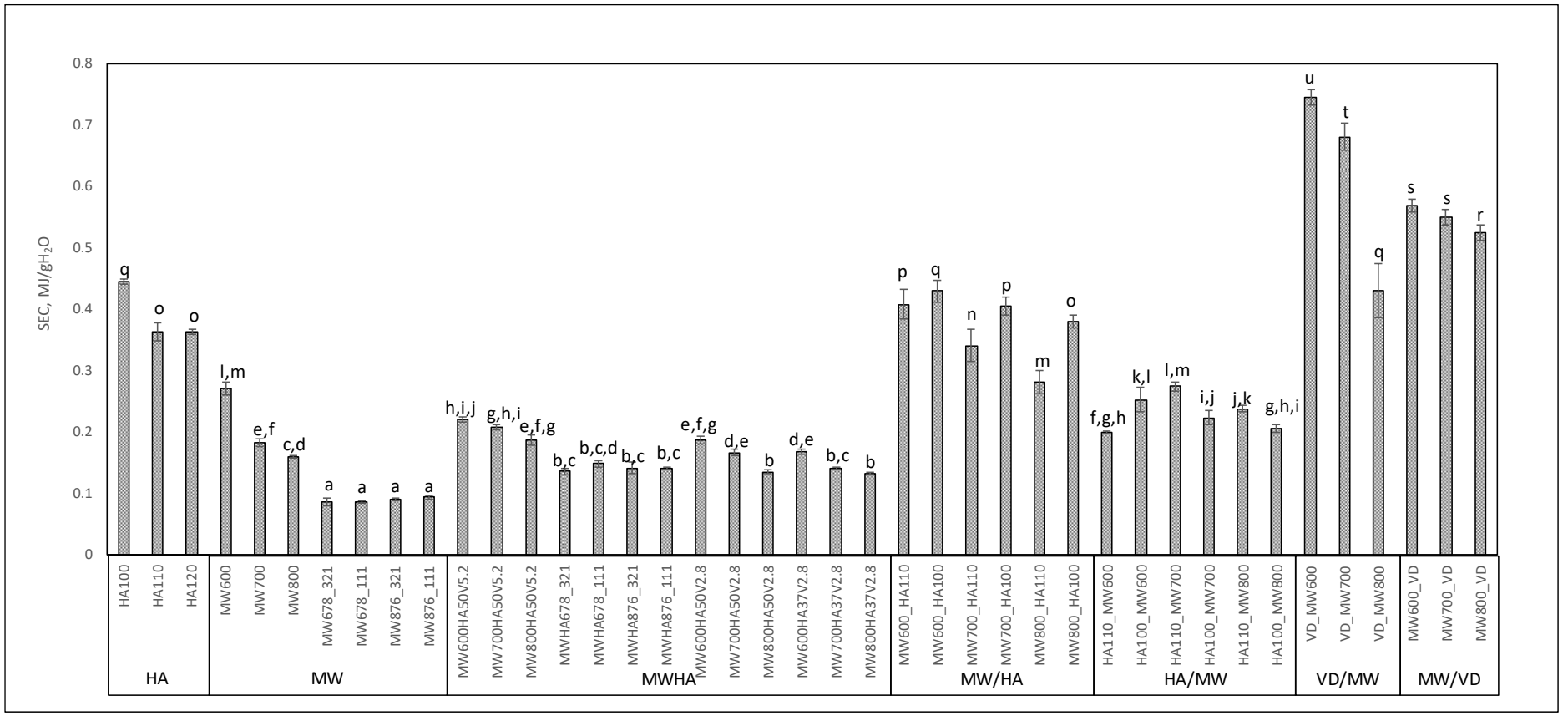


Figure 4.36: SEC for dried rubber samples obtained for different drying strategies in various conditions

It's also worth to mention that the lowest SEC was attained by the MW multi-steps drying. This strategy involving three drying cycles to allow the step up or step down of MW power in a designated period. When the drying process approaching the final stage of drying, the moisture content of the sample is reduced as well as the absorption of microwave energy (Ratanadecho, Aoki and Akahori, 2001). Hence, the changes of MW power level and drying time at these stages could have effectively reduced overall drying time and faster moisture removal which contributes to low SEC value. For single stage drying, both MW or HA consumed a significant amount of energy which yields SEC values of 0.16-0.27 MJ/g H_2O and 0.36-0.44 MJ/g H_2O, respectively. Unlike industrial dryer reported in the literature (Tirawanichakul and Tirawanichakul, 2008; Utomo, Hasanudin and Suroso, 2010), laboratory dryers drying a small quantity of sample tend to be much higher in terms of SEC values (Tarhan *et al.*, 2011). Although the MW power output has a higher rating than that of HA oven, however, SEC value of MW performed much better. Similar observations were reported by Torki-Harchegani *et al.* (2016) and Motevali *et al.* (2011). The reason lies in the rapid water removal from the sample within a short effective drying time with the application of MW energy. Besides, it is clearly shown that the SEC value is inversely proportional to MW power level and temperature but proportional to the drying time.

Second lowest SEC, after MW group was obtained by MWHA's group with an average SEC scored below 0.22 MJ/g H_2O. In this hybrid drying, both SEC values of MW800HA50V2.8 and MW800HA37V2.8 were measured at around 0.13 MJ/g H_2O. On the other hand, the SEC for MWHA50V5.2 was increased

by 40.9% when operated in full heater power and air velocity with similar MW power applied in all drying condition. A similar observation was made by Motevali, Minaei and Khoshtagaza (2011) where an increase of air velocity from 0.3 m/s to 1.0 m/s cause an increase of energy consumption in the drying of pomegranate arils. This phenomenon can be explained by surface cooling due to an increase in air velocity has reduced drying temperature and thus increase in drying time (Tulasidas, Raghavan and Mujumdar, 1995). Clearly, a reduction of SEC value can be achieved by decreasing the air velocity and the hot air temperature supplied to the dryer. Figure 4.36 shows average SEC values of MWHA are considered within a lower range in comparing to others drying methods. The result demonstrated that MWHA can be used to efficiently dry the crumb rubber. Besides, the comparison between the SEC values demonstrated the significant difference in the drying sequence applied in drying methods. For instance, the introduction of HA at the first stage before MW can significantly reduce the SEC value by an average of 37.5% in comparison to MW/HA. This is due to a substantial amount of moisture was removed earlier in the HA whereas MW power is applied later to remove residue moisture in the second stage. Meanwhile, SEC values of MW followed by VD were 11.3% lower than VD /MW. Unlike the HA/MW, the introduction of MW at the first stage could marginally reduce the SEC values as the MW can effectively remove moisture in a shorter time in comparison to VD alone.

4.6 Physicochemical change of dried rubber

4.6.1 Colour changes of dried rubber under different drying strategies

In the current research, the relations of drying strategies to dried rubber colours especially the total colour changes, lightness/darkness were assessed and is tabulated in Table 4.2. Raw NR and SMR 20 block rubber were obtained from the Kuala Pilah Rubber Factory for the colour reference and benchmarking purpose. These two samples are the benchmark for the colour prior to drying (raw crumb rubber) and colour after drying (SMR20), respectively. Preferred colours are those closest to the original colour of the fresh sample. With reference to Figure 4.37 and Table 4.2, the lightest coloured dried rubber was found in the group of MWHA with reduced air velocity ($L^* > 30$). The light colour of the MWHA samples could be caused by the cooling effect of the air flow over the sample surface while the MW heated the interior (Nadia R. Pereira., Antonio Marsaioli Jr. and Ahrne, 2007).

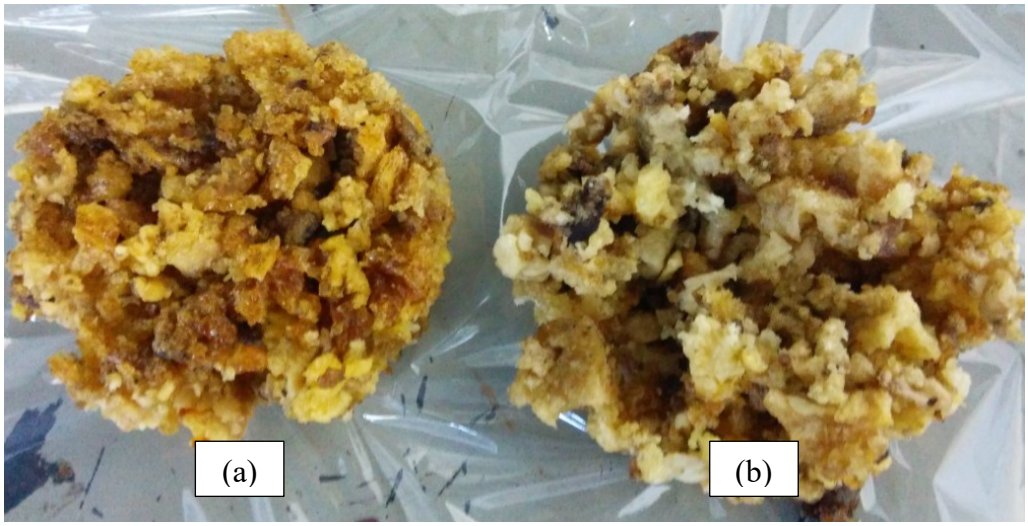


Figure 4.37: Colour of dried rubber obtained from (a) MW drying, (b) MWHA

Conversely, the presence of static hot air at elevated temperature promotes charring effect especially in some of MW dried samples such as VD/MW which possessed darker colour. The charring on the sample was represented by low redness ($a*$ value) and yellowness ($b*$ value) found in dried sample obtained from both MW and VD/MW which also scoring $L^* < 20$ represented a darker product. The fact of colour darkening as an influence of air temperature was reported by Chin Peng Sung (1969) who observed that increasing the drying temperature to over 100° C in viscosity-stabilised Heveacrumb will increase the darkening effect. Hence, adjustment of air temperature to around 100° C is recommended for better colour preservation. Besides, the change of the colour from light yellow to black also depending on the extent of degradation.

The total colour change, ΔE^* values, which consider changes in redness and yellowness, were also compared. The minimum colour change with ΔE^* less than 5.0 units by referring to SMR 20 was found in the dried rubber produced from HA110, MW800_HA100, HA100_MW700 with $(p < 0.05)$. In contrast, most of MW, MW/VD and MWHAV2.8 dried rubber samples yield larger ΔE^* by exceeding 10. Also, HA, MW/HA and HA/MW demonstrated inconsistent colour changes as the temperature varied between 100°C and 120°C $(p < 0.05)$. In other words, there was no direct relationship between the degree of temperature, microwave power density in colour changes (Maskan, 2000). Meanwhile, most of MW and MWHA dried rubbers did not differ significantly from each other in terms of ΔE^* in numerous drying conditions$(p > 0.05)$. They demonstrated colour consistency in various drying conditions.

Colour assessment also compare the colour changes of raw NR with the dried sample. It is deduced that the lesser ΔE^* in comparison to raw NR, the more approximate the dried rubber to the colour tone of raw NR (See APPENDICES Figure C1). Table 4.2 tabulated ΔE values ranges from 33.32 to 56.90 in which MWHA with reduced air velocity gave average lower total colour change in comparison to other drying strategies. Apart from the reduction in drying time, lesser colour change can be an indication of reduced oxidative reaction which indicate a better retention of antioxidant in the product (Vega-Galvez et al, 2012; Zielinska and Michalska, 2016). Hence, MWHA product will have better ageing properties due to maximum colour retention. MW, however, demonstrating sizeable colour change in comparison to its raw NR which is represented by the darker colour of the dried sample. Evidently, single stage drying often produce darker product than the product obtained from either two-stage or hybrid drying method. On the other hand, no visible white spot has been detected during the visual inspection of dried rubber as shown in Figure 4.37 and Figure C2 (See APPENDICES). This shows the great ability of microwave-assisted drying in eliminating the white spot on rubber. In a nutshell, MWHA is a preferred method than other drying strategies due to its light colour, small ΔE^* values and having consistency in colour presentation.

Table 4.2: Colour difference of different drying strategies in comparison with industrial and raw rubber samples

Drying methods	Colour parameters			Total colour change, Δ*E*	
	L*	**a***	**b***	**dE_{ind}**	**dE_{raw}**
Industrial graded rubber (SMR 20)	23.00±1.71	8.38±0.93	11.90±2.32	-	-
Fresh raw natural rubber	73.17±4.18	-0.33±0.38	12.37±2.86	-	-
(I) Single stage drying					
(A) HA					
HA100	27.75±1.06^{d}	5.90±0.45^{c}	17.00±0.57^{f}	7.40±1.21^{e}	46.08±1.05^{ab}
HA110	22.65±0.35^{bc}	6.60±0.85^{e}	11.8±1.27^{c}	2.06±0.73^{a}	51.00±0.45^{bc}
HA120	23.75±3.18^{c}	7.55±1.06^{f}	13.45±3.04^{c}	3.51±1.77^{b}	50.11±2.91^{b}
(B) MW					
MW600	16.33±1.15^{a}	2.07±0.50^{b}	3.10±0.26^{b}	12.69±0.25^{f}	56.90±0.70^{d}
MW700	17.87±0.78^{a}	1.93±0.42^{b}	1.87±0.25^{a}	12.57±0.70^{f}	56.23±0.85^{d}
MW800	19.00±0.70^{a}	1.37±0.29^{a}	2.00±0.96^{a}	13.63±0.73^{f}	55.19±0.73^{cd}
MW678_111	24.75±3.89^{c}	6.25±0.92^{d}	11.80±1.56^{c}	4.01±1.17^{bc}	48.89±3.75^{b}
MW678_321	23.30±1.98^{c}	5.45±0.49^{c}	13.15±0.92^{d}	3.51±0.91^{b}	50.21±2.01^{b}
MW876_111	19.85±1.63^{ab}	7.30±0.42^{ef}	14.85±1.06^{e}	4.65±0.53^{c}	53.93±1.50^{c}
MW876_321	29.05±0.78^{d}	6.85±0.21^{e}	18.25±0.21^{f}	8.92±0.41^{e}	45.08±0.82^{a}

(II) Two-stage drying					
(A) MW/HA					
MW6_HA100	29.30±1.13^{ij}	5.55±0.63^{j}	12.05±2.05^{l}	7.06±1.22^{l}	44.29±1.19^{f}
MW7_HA100	23.05±0.92^{g}	6.00±0.14^{k}	8.85±1.06^{jk}	3.94±0.89^{hi}	50.64±0.97^{gh}
MW8_HA100	23.85±0.78^{gh}	6.55±0.35^{k}	13.20±0.85^{l}	2.52±0.43^{gh}	49.81±0.74^{g}
MW6_HA110	23.10±0.85^{g}	6.85±1.06^{l}	9.35±0.78^{k}	3.06±1.21^{h}	50.68±0.94^{gh}
MW7_HA110	23.25±3.04^{gh}	5.30±0.57^{j}	7.90±0.57^{i}	5.48±0.87^{k}	50.44±3.02^{gh}
MW8_HA110	20.00±1.13^{f}	4.65±0.49^{i}	6.05±0.21^{h}	7.60±0.53^{l}	53.77±1.05^{i}
(B) HA/MW					
HA100_MW6	29.85±0.49^{ij}	5.55±0.35^{j}	12.80±0.57^{l}	7.47±0.66^{l}	43.72±0.53^{f}
HA100_MW7	24.70±1.27^{h}	7.25±0.21^{l}	11.95±0.07^{kl}	2.08±1.16^{g}	49.06±1.29^{g}
HA100_MW8	21.80±2.26^{fg}	6.40±0.14^{k}	9.90±0.57^{k}	3.39±1.05^{hi}	51.87±2.29^{h}
HA110_MW6	29.95±1.48^{j}	7.70±0.28^{l}	14.65±0.92^{l}	7.51±1.74^{l}	44.02±1.46^{f}
HA110_MW7	28.25±1.06^{i}	7.40±0.42^{i}	13.70±0.71^{j}	5.68±0.83^{k}	45.60±1.14^{f}
HA110_MW8	24.15±0.49^{h}	5.70±0.42^{jk}	8.70±0.14^{j}	4.35±0.23^{j}	49.52±0.45^{g}
(C) MW/VD & VD/MW					
MW6_VD	37.2±0.00^{kl}	8.90±0.14^{m}	21.25±0.07^{m}	17.01±0.03^{n}	38.18±0.02^{e}
MW7_VD	38.5±1.98^{l}	9.65±0.78^{n}	23.6±0.71^{m}	19.51±1.20^{o}	37.80±1.82^{e}
MW8_VD	36.75±6.86^{k}	3.73±0.25^{h}	7.90±1.04^{i}	16.59±6.73^{n}	38.16±6.23^{ef}

VD_MW6	21.35±1.77^{fg}	6.30±0.42^{k}	8.30±0.28^{j}	4.65±0.22^{j}	52.40±1.78^{hi}
VD_MW7	18.05±5.16^{ef}	2.80±1.41^{g}	4.55±2.76^{g}	10.73±5.00^{m}	55.79±5.41^{ij}
VD_MW8	17.25±0.64^{e}	2.70±0.00^{g}	4.15±0.07^{g}	11.20±0.28^{m}	56.60±0.62^{j}
(III)Hybrid drying					
(A)MWHA					
MW600HA50V5.2	22.80±4.67^{no}	7.50±1.27^{s}	12.75±0.21^{o}	3.62±0.52^{p}	50.99±4.41^{no}
MW700HA50V5.2	24.70±3.25^{o}	5.00±0.85^{o}	12.95±2.33^{o}	4.82±1.06^{q}	48.80±3.11^{m}
MW800HA50V5.2	24.63±3.65^{o}	4.80±0.71^{o}	13.00±2.69^{o}	4.84±2.27^{q}	48.78±3.27^{m}
MW800HA37V2.8	35.30±3.11^{pq}	5.30±0.99^{p}	18.45±0.07^{q}	14.29±2.89^{u}	38.76±3.18^{kl}
MW700HA37V2.8	39.35±7.71^{r}	5.85±0.49^{qr}	20.95±0.64^{r}	19.03±6.86^{v}	35.50±7.10^{kl}
MW600HA37V2.8	33.05±6.43^{p}	5.00±0.31^{o}	18.1±1.56^{q}	12.45±5.97^{t}	40.91±6.09^{l}
MW800HA50V2.8	36.60±4.24^{q}	6.35±0.78^{r}	18.35±2.62^{pqr}	15.23±4.79^{u}	37.74±3.56^{kl}
MW700HA50V2.8	31.65±3.89^{p}	6.10±0.71^{qr}	15.85±3.04^{pq}	9.91±4.45^{s}	42.23±3.46^{l}
MW600HA50V2.8	41.15±0.49^{s}	5.35±0.92^{p}	19.6±0.28^{r}	19.96±0.20^{v}	33.32±0.38^{k}
MWHA678_321	16.70±0.85^{m}	4.60±0.57^{o}	10.95±0.92^{n}	7.43±1.12^{r}	56.70±0.82^{p}
MWHA678_111	25.95±0.63^{o}	4.85±0.35^{o}	16.55±0.35^{p}	6.54±0.73^{r}	47.69±0.64^{m}
MWHA876_111	22.00±0.14^{n}	5.90±0.25^{q}	10.40±1.27^{n}	3.16±0.65^{p}	51.59±0.19^{o}
MWHA876_321	22.85±1.06^{n}	6.50±0.42^{r}	16.30±2.40^{p}	4.88±2.30^{q}	50.96±0.92^{n}

Note: Mean values ± standard deviation$(n = 2)$ within the same column and drying strategies with the same letter (in superscript) are not significantly different $(p > 0.05)$

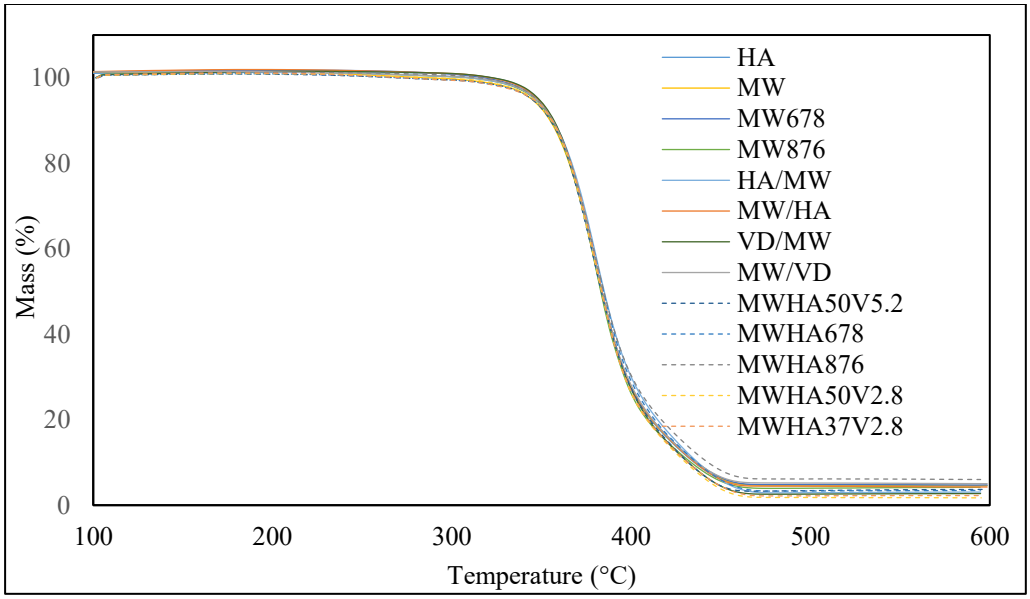
dE_{ind}: Total colour difference with reference to industrial graded dried rubber (SMR 20)

dE_{raw}: Total colour difference with referring to raw crumb rubber (field coagula)

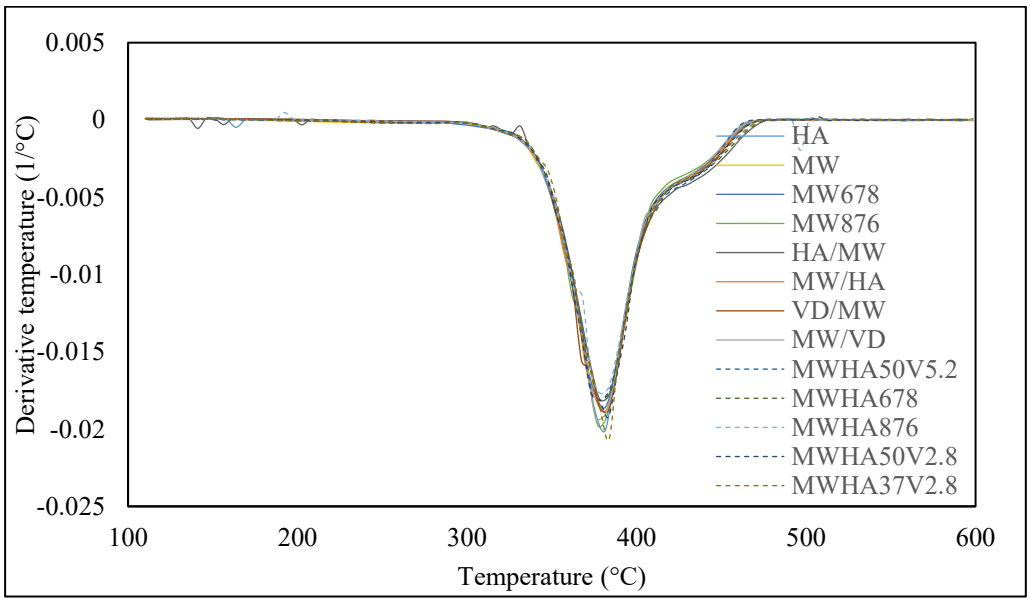
4.6.2 Studies on thermal oxidative degradation behaviours of dried rubber by thermogravimetry analysis

Thermal decomposition temperatures of the dried rubbers were evaluated by TGA and derivative thermogravimetric (DTG) to study decomposition behaviour in an inert atmosphere. The decomposition stages were identified using a derivative of the TG curves and thus, Table 4.3 shows the maximum temperature of each stage of degradation. The maximum temperature comprises of onset temperatures of decomposition, T_o which can be deduced from TGA curves by extrapolating the curve at the initial mass loss of the natural rubber compounds. From the DTG curves, the maximum decomposition data comprised of temperature of the maximum mass loss rate, T_r and end temperature of degradation, T_e can be used to determine the temperature of degradation. A compilation of TGA curves from different drying methods as shown in Figure 4.38 has comparable thermal degradation curve which was reported in other literatures (Li *et al.*, 2010; Khaled H. Mahmoud. and Makled, 2012). From the results in Figure 4.38, it can be seen that some volatile compounds vaporized at low temperature before pyrolysis begins i.e. in the regions of 2% from the initial mass loss. The second decomposition occurred in the ranged from 336 °C to 470 °C with a mass loss of at about 97%, indicating the breakdown of NR into monomer, dimer, trimers when temperature started to rise. A small shoulder can be observed at about 425 °C attributed to the cross-linked and cyclized networks (Martins *et al.*, 2008) which also corresponding to approximately 12% of the main pyrolysis step. The curve ended with residue which was found as ash content at the end of the process with constituent of around 6%. By referring to Figure 4.38, TGA thermogram of different drying methods exhibited

approximate trend of mass losses and mass decomposition rate. However, there are some dissimilarity in terms of the degradation temperature for each drying method which are tabulated in Table 4.3



(a) Mass losses in percentage trend



(b) Mass decomposition rate (DTGA)

Figure 4.38: TGA thermogram for different drying strategies

Table 4.3: Summary of degradation temperature of dried rubber samples obtained from different drying methods

Drying strategies	T_o	T_r	T_e
HA	337.84	381.84	463.47
MW	340.61	381.51	466.70
MW678	338.25	379.36	463.28
MW876	335.72	380.05	464.49
HA/MW	339.96	379.81	467.67
MW/HA	341.64	378.91	465.31
VD/MW	335.89	380.14	466.36
MW/VD	343.87	375.37	464.87
MWHA50V5.2	344.07	377.74	467.05
MWHA678	340.57	381.26	470.40
MWHA876	343.54	379.55	467.51
MWHA50V2.8	338.38	383.70	466.25
MWHA37V2.8	340.59	385.30	469.00

Table 4.3 shows the temperatures of the initial decomposition or onset and the maximum decomposition of the dried rubber derived from the DTG curves. The initial decomposition temperature was used to predict the thermal stability of the rubber. On average, the onset temperature of decomposition for MW dried rubber sample was higher than those of HA- dried sample except in VD/MW and MW876. MW drying has shown 1°C to 7°C higher than the HA- dried sample by comparing the end temperature of degradation. This means that the

MW dried product has higher thermal resistance than HA dried product. Meanwhile, small ranges of end temperature of degradation were detected in another drying method. Results proved that the sample with MW treated had better thermal oxidation resistance in comparison with the sample without MW treated. The similar outcome was reported by Chen et al. in the thermal behaviour study of NR (Chen, Wang, *et al.*, 2012).

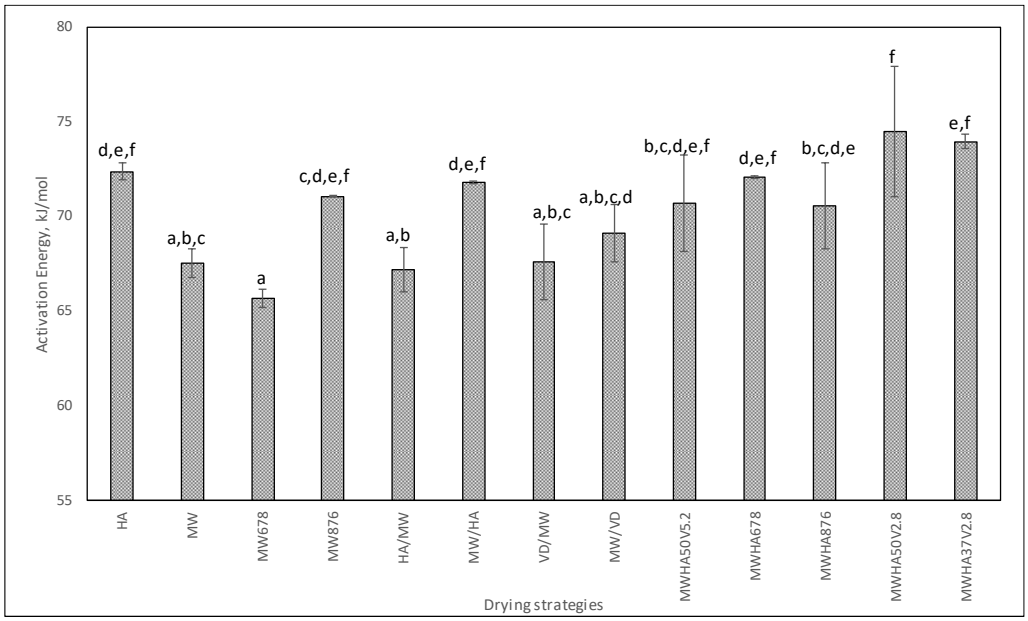


Figure 4.39: Activation energy E_a of thermal decomposition for dried rubber samples obtained for different drying strategies. Vertical bar and line graph indicated with the same letter are not significantly different ($n = 2; p > 0.05$)

On the other hand, the kinetic patterns of the thermal degradation of dried rubber, in the form of activation energy, E_a were calculated by using the Coats-Redfern methods. Figure 4.39 shows the E_a values of the rubber samples obtained from different drying methods. The linear plot as a function of temperature with concurrency value R^2 around 0.97 was chosen for all methods. The E_a values varied between 65.66 and 74.46 kJ/mol, which were in agreement with the

literature (Galiani *et al.*, 2013). From all methods, MW678 suffered the lowest E_a ($p < 0.05$), followed by VD/MW and MW which have an approximate value of E_a ($p > 0.05$). The reduction in the energy of activation of a process is resulted from an increase in the average energy of the molecules. It was noticed that the E_a of MWHA dried rubbers were averagely higher which representing a more stable material in terms of resistance to thermal degradation (Galiani *et al.*, 2013). Apparently, these variations in E_a were predicted because when more than one degradation mechanism such as oxygen uptake, chain scission, peroxide formation and cross linking are occurring, the E_a will not always the same (Medeiros *et al.*, 2010). Further to this, another investigation of thermal degradative behaviour has been done via PRI measurement (in Section 4.6.3) to understand the relationship between E_a and PRI values in different rubber samples.

4.6.3 Rheological investigation of dried rubber (P_0, PRI and VM) obtained in various drying strategies

A rheological analysis consisted of P_o and PRI may reveal the molecular degradation level of raw NR when undergoes temperature oxidative ageing. In this regard, the PRI is a direct measurement to denote the susceptibilities of NR to thermal oxidative breakdown. According to the study of PRI by Sambhi (1989), the reduction in plasticity can be caused by thermal oxidative degradation whereas the increase in plasticity is due to the hardening of the rubber arising from crosslinking reaction of functional groups present in the NR system or crosslinking of radical intermediates produced during the oxidative process. Rheological properties of dried rubber samples were tested (See

APPENDICES Figure A1, A2 and A3) and presented in Figure 4.40. With reference to SMR scheme, all drying methods have surpassed the minimum PRI value of 40% and perfectly maintained at below the maximum allowable value of VM($\mathrm{VM} \leq 0.8\, wt\%$). However, MW treated rubber samples have suffered from low P_o; meanwhile HA and MWHA (except MWHA678) rubber samples have passed the SMR20 requirement by exceeding the minimum values of 30%. In fact, a higher P_o for a given rubber sample is indicating a low plasticity and a high viscosity (Subramaniam, 1975; Sambhi, 1989).

Besides, PRI indices are often related to crosslink density, molecular size, vulcanizate ageing, and presence of non-rubber content (Bateman and Sekhar, 1966). A higher PRI value indicates better quality rubber whereas lower PRI has both inferior quality and greater variability in their property. It can be observed that rheological properties of MWHA's rubber samples were on par with industrial products with 28.6% even lower of volatile matter value to that of SMR product. The finding is in agreement with Zhang *et al.* (2011) who stated that MW drying has not influenced the viscoelastic properties of NR greatly, as well as the profiles of its rheological behaviour. Nonetheless, both MW/VD and MW/HA had distinguished value of P_o but were inferior in terms of volatile content when compared to other. It is interesting to highlight that there are some correlations between E_a and P_o, PRI of rubber sample. It can be seen from both Figure 4.39 and Figure 4.40, the HA/MW and VD/MW shared lower quality indices in terms of P_o and PRI whereby similar pattern was also found in E_a values despite their slight variation. Obviously, MWHA dried rubber samples have demonstrated relatively stable thermal resistance with reference to both PRI

and E_a .In other word, a higher retention of technological properties can be achieved by using MWHA method.

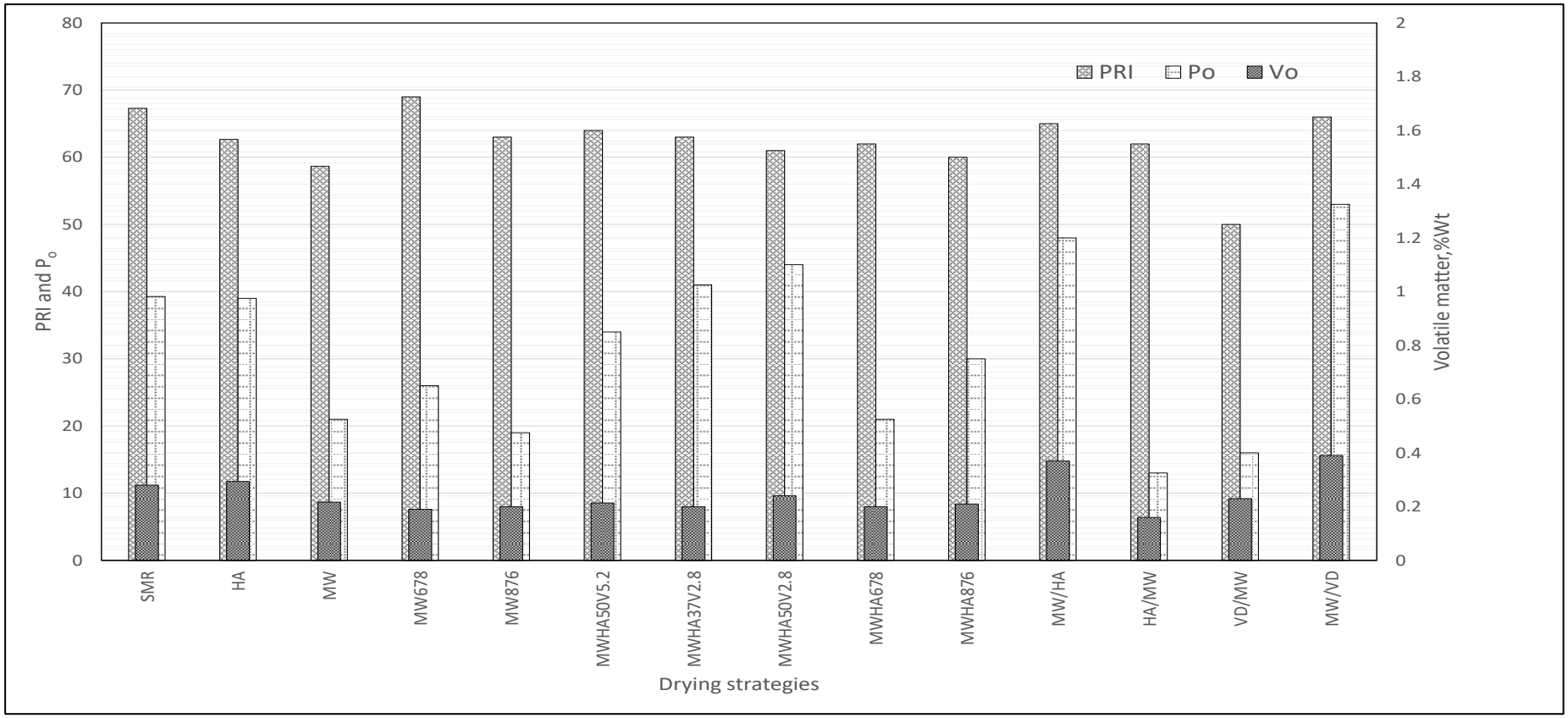


Figure 4.40: Comparison of mechanical and chemical properties of dried rubber samples obtained from different drying strategies

Legends:

Data are the mean values; Minimum requirements to meet SMR20 in accordance to Standard Malaysian Rubber scheme SMR: Volatile matter, VM ≤0.8 wt%; Plasticity retention index, PRI ≥40%; Wallace plasticity, P_o≥30%

4.6.4 Textural properties of dried rubber

The key analysis in this scope is to investigate the springiness of dried rubber by using mechanical evaluation technique. The evaluation measuring the ability of the sample to recover its original form after deforming force was removed. As shown in Figure 4.41, the springiness indices ranging from 50% to 60% comprised of 24.4% of total dried rubber samples. Meanwhile, springiness index below 50 was obtained from VD/MW samples (7.31% of total samples) at 26% to 35% followed by HA110 at 45.69%. MW600HA50V2.8 dried rubber has the highest springiness index at 72.23% after MW800HA50V5.2 and MW600HA37V2.8 recorded at 67.1% and 65.9% respectively. This has indicated that MWHA dried samples have marked ability in returning to its original shape after it was compressed. Overall, the springiness indices measured in most of the MW/HA samples were close to 60% to its original springiness. Other drying strategies were reported at HA (45.69-56.35%), MW (49.71 to 65.16%), MWHA (49.14 to 72.23%), HA/MW (50.4 to 56.98%), VD/MW (26.04 to 35.28%) and MW/VD (57.34-58.91%) respectively. This information is useful to measure the impact of the design of the experiment e.g. single stage, two-stage and hybrid drying in affecting the mechanical properties of dried rubber. In fact, this finding shows that springiness is significantly affected by the combined drying process and drying conditions. Hybrid drying shows the maximum retention of mechanical properties by having a high range of springiness value. Two-stage drying, however, has mixed springiness results and it is depending on the drying method. For example, the springiness value of VD/MW was not as high as the MW/VD despite similar drying instruments were employed but in different sequences. This could be because of the alteration of

cell walls whereby a matrix upholding the structure during the action of external force when longer microwave drying was applied to the samples (Markowski, Bondaruk and Błaszczak, 2009; Witrowa-Rajchert and Rząca, 2009). The springiness measured in various drying conditions was different because of the different cell wall structure or thermal treatment. It is noteworthy to mention that 46.15% of MWHA's samples successfully yield springiness index that is more than 60% which was higher than MW/HA (33.33%) and MW (14.3%), respectively. This shows good retention of mechanical properties of rubber in retaining its original elasticity potential. Conversely, VD/MW is likely to create rubber product with low springiness as shown in Figure 4.41. This phenomenon can be explained by the formation of hydrogen with water in phenolic derivate, which can influent the sample's water holding capacity (Maga, 1988) and springiness decreased with increasing water content (Li, Carpenter and Cheney, 1998). This decrease could be related to the gaps and channels formed during heterogeneous and fast moisture diffusion. By comparing the trend between Figure 4.40 and Figure 4.41, some correlation can be found particularly in the VD/MW and MW/HA, HA/MW. For instance, VD/MW has the lowest springiness and this is corresponding to low PRI as reported in Section 4.6.3. Similarly, the MW/HA has slightly higher PRI and P_o than HA/MW and this is also reflected in the average lower springiness obtained from HA/MW in comparison with MW/HA.

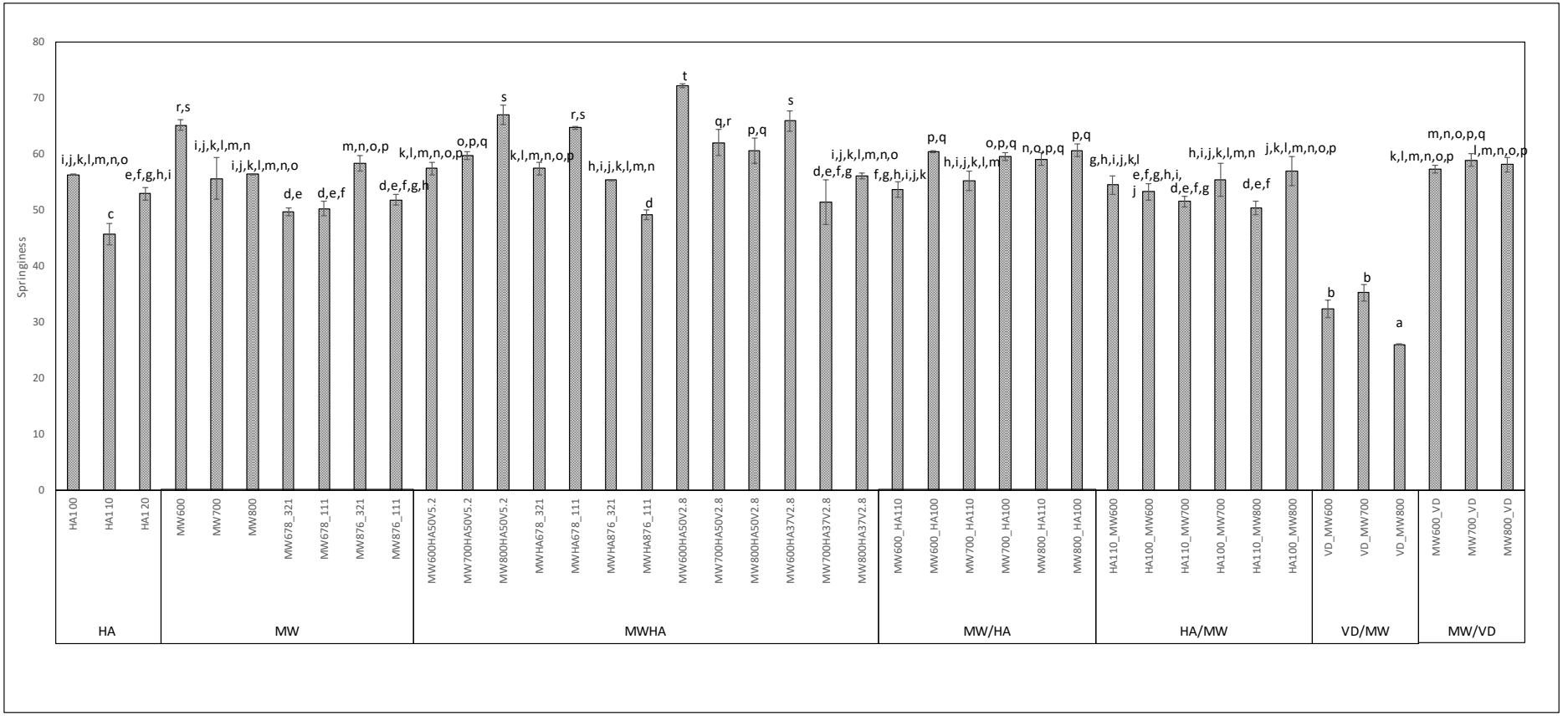


Figure 4.41: Effect of drying strategies on springiness index of dried rubber. *Results are means of 3 observations ± standard error; Vertical bar and line graph indicated with the same letter are not significantly different (p>0.05)*

4.6.5 Study of dried rubber surface topography via scanning electron microscope (SEM)

Rubber specimens obtained from various drying methods were analysed through SEM imaging by secondary electrons and backscattered electrons. SEM was employed to illustrate the morphology of the rubber surface. According to Vanderhoff, Bradford and Carrington (1973), the drying process apparently leads to the globules gradually coming together until a compact state is reached, corresponding to the dehydrated structure observed. The SEM micrographs of rubber dried with HA and MWHA are depicted in Figure 4.42.

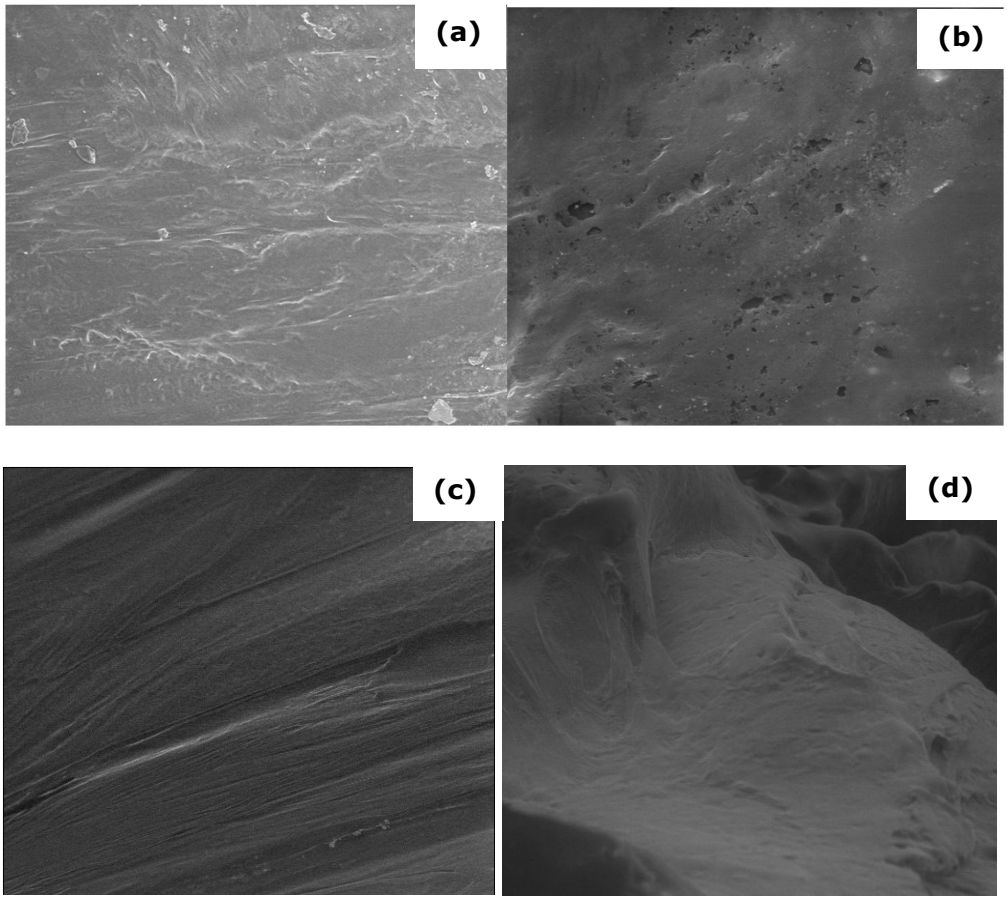


Figure 4.42: SEM micrographs (top view and cross-sectional view) of the fracture of dried rubber: (a,c) HA; (b,d) MW assisted drying (Bar=50 and 200 µm)(magnification level: 1600-2000x)

Micropores formed on dried rubber surface as can be seen in Figure 4.42 (b) & (d) provides evidence of migration of water vapour. For better visibility of the occurrence of micropore at the dried rubber surface, an open source image processing package named Fiji (Schindelin, Arganda-Carreras and Frise, 2012) was applied to produce the binary image between MW assisted drying rubber sample as shown in Figure 4.43.

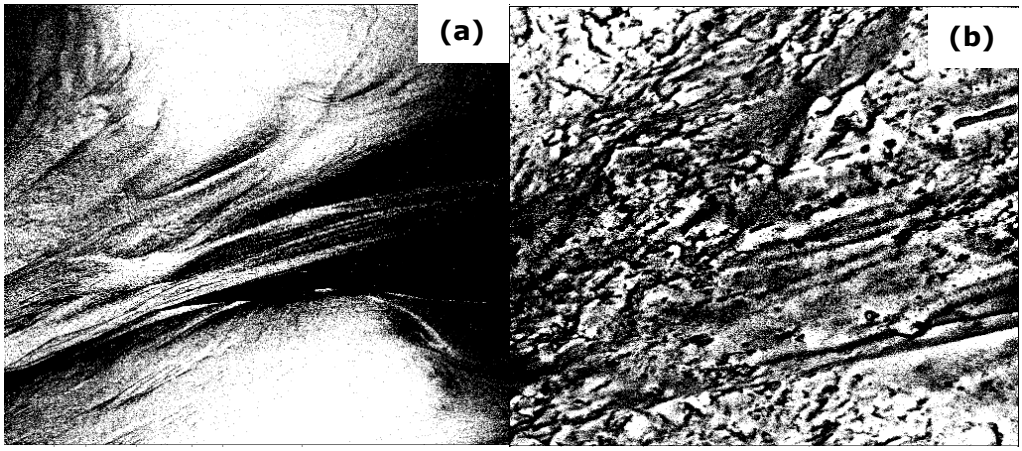
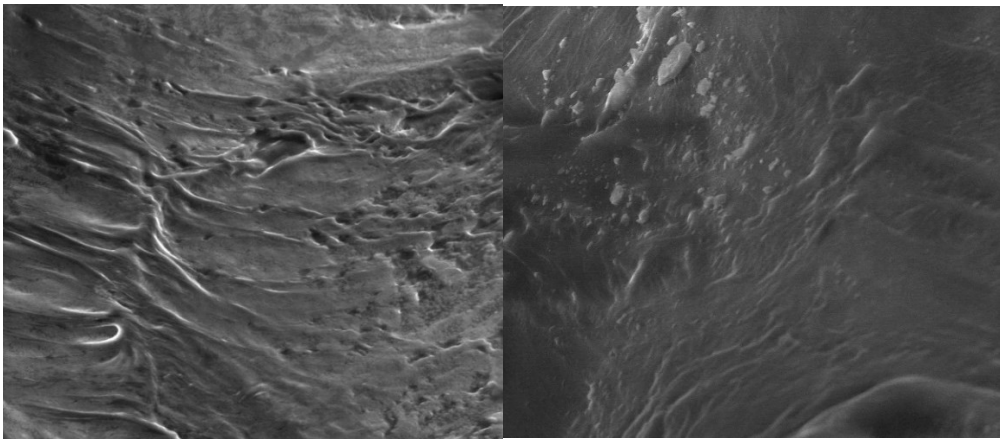


Figure 4.43: Binary image of the fracture of dried rubber (a) HA; (b) MWHA (Bar=50 μm; magnification level: 1600-2000x)

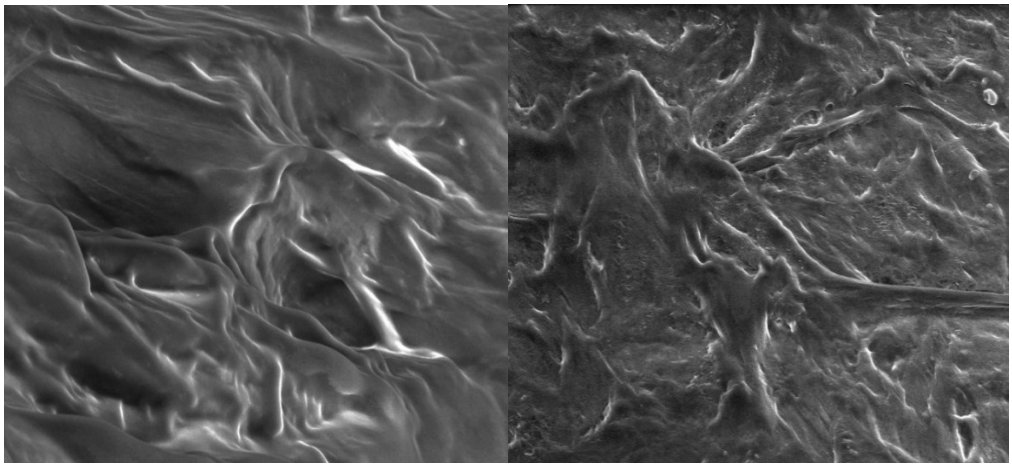
Figure 4.43 (a) shows smooth surface as the effect of melting process after applied with the HA. In contrast, Figure 4.43 (b) shows an irregular wavy pattern with lots of white dotted spot that reflect the existence of the micropores left by water diffusion when rubber undergoes the microwave treatment. In contrast, A similar observation was reported by Auria *et al.* (1991) where no significant porosity can be seen in the hot air dried natural rubber sheet. Likewise, identical topographical observation on dried rubber surface was also reported by Chen *et al.* (2008). Unlike HA drying, MW energy heated polar molecules inside the product forces the moisture to travel out of the sample due to the pressure

gradient of water when the water's temperature is increased instantaneously. In another word, the presence of micropores in MW dried rubber give a positive effect to a higher moisture diffusivity value whereas HA dried rubber exhibited smooth surface due to melting or case hardening at high temperature has lower moisture diffusivity value. It is also confirmed in the study conducted by Rattanadecho *et al.* (2007) that MW dried products had a better microstructure arrangement because of uniform energy absorption and a minimum of shrinkage. The finding is further supported by Rahman (1985) who has suggested that the overall drying time is considerably dominated by the diffusion process of water in the drying of rubber under HA environment. A higher moisture diffusivity values are favourable in drying process because of quick water removal from the sample and hence shorter drying period is needed. The rise of temperature of the sample permits an increase of the water vapour pressure inside the pores and results in the pressure-induced opening of pores.

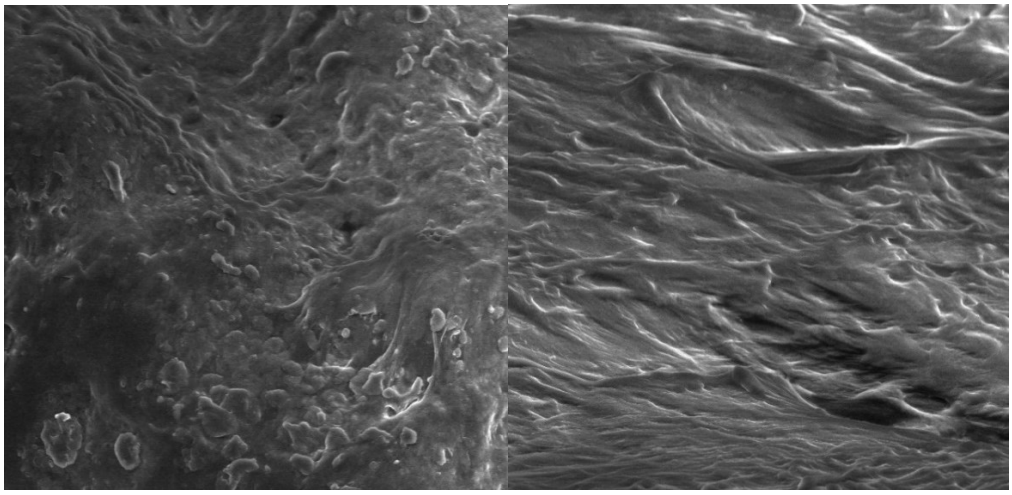
Figure 4.44 shows SEM micrographs obtained from MWHA dried samples. The surfaces of dried rubber samples are corrugated and micropores can be spotted in some area. This shows that the MW facilitated the rapid transport of moisture and caused morphological changes in rubber.



(a)HA/MW (b)MW/HA



(c)VD/MW (d)MW/VD



(e)MWHA37V2.8 (f)MWHA50V2.8

Figure 4.44: SEM micrographs of dried rubber surface obtained from different drying strategies. (Bar=50 μm and 200 μm) (Magnification level: 1600-2000x)

4.7 Multiple regression analysis of microwave assisted-drying strategies in relation to its drying parameters

In this research, multiple types of strategies namely two-stage drying and hybrid drying have been tested for the drying of rubber. These dryings gave different impact on drying performance from SEC to D_{eff} which also represent the effectiveness of the method in treating the NR. This complex relationship in

between independent variable viz. drying method and dependent variable i.e. drying performance in random design of experiment was analysed. Multiple regression analysis was applied to compare two or more different independent variables in affecting the single dependent variable. First three models were considered to establish the relationship between D_{eff} and drying conditions applied in MW-assisted drying. Model 1 as shown in Equation 4.12 can be used to describe the relationship between the D_{eff} and two independent variables in two- stage drying (HA/MW).

$$\text{Model 1}_{D_{eff_1}} = 0.137HA + 0.971MW \quad (4.12)$$

Table 4.4 shows that the R^2 values is 0.962 which indicate 96.2% of the change in D_{eff} is due to the change in hot air temperature and power level of MW.

Table 4.4: R^2 values for Model 1

Model	R	R Square	Adjusted R Square	Std. Error of the Estimate
1	.981[a]	.962	.936	.0282869

a. Predictors: (Constant), HA, MW

On the other hand, from Table 4.5, t-test value reveals the effect of MW power level has significant effect toward D_{eff} in comparison to hot air temperature ($p < 0.05$ level of significance). This is supported by the result presented in Section 4.2.3.

Table 4.5: Coefficients for Model 1

Model		Unstandardized Coefficients		Standardized Coefficients		
		B	Std. Error	Beta	t	Sig.
1	(Constant)	.212	.262		.809	.478
	HA	.003	.002	.137	1.219	.310
	MW	.001	.000	.971	8.612	.003

The second model is given in Equation 4.13 can be used to describe the relationship between the D_{eff} and two independent variables in two- stage drying (MW/HA).

$$\text{Model 2}_{D_{eff_2}} = 0.609MW + 0.262HA \quad (4.13)$$

Unlike the first model, R^2 for the second model in Table 4.6 shows 44% of the change in D_{eff} was due to the change in HA temperature and MW power level. Indeed, no significant relationship was found between these variables.

Table 4.6: R^2 values for Model 2

Model	R	R Square	Adjusted R Square	Std. Error of the Estimate
2	.663[a]	.440	.066	.2812983

a. Predictors: (Constant), MW, HA

Similarly, the t-test values in Table 4.7 show that both microwave power level (MW) and hot air temperature (HA) had no effect toward the varies of D_{eff} ($p > 0.05$ level of significance) in MW/HA. In other word, the changes of D_{eff} is minimal in MW/HA. This agrees with the data presented in Section 4.2.3 in

which the MW drying in the initial stage has almost dried up the sample whereby the introduction of HA drying in the second stage become less significant in terms of water removal rate.

Table 4.7: Coefficients for Model 2

Model		Unstandardized Coefficients		Standardized Coefficients	t	Sig.
		B	Std. Error	Beta		
2	(Constant)	-1.339	2.607		-.514	.643
	MW	.002	.001	.609	1.409	.254
	HA	.014	.023	.262	.605	.588

Furthermore, the statistical analysis of MWHA and D_{eff} in the third model as given by Equation 4.14 revealed that 74.1% of the change in the D_{eff} is due to the change of the MW power level.

$$\text{Model 3}_{D_{eff_3}} = 0.861MW - 0.534V + 0.323HA \quad (4.14)$$

Table 4.8 shows the combination of air velocity, V and MW power contributes (88.0-74.1%) 13.9% additional changes in D_{eff}. HA temperature only contribute (95.8-88.0%) 7.8% of the variance in D_{eff}. The three-predictor combined contributed 95.8% of the variance in the D_{eff} criterion variable.

Table 4.8: R^2 values of Model 3

Model	R	R Square	Adjusted R Square	Std. Error of the Estimate
i	.861[a]	.741	.704	.58636
ii	.938[b]	.880	.840	.43186
iii	.979[c]	.958	.933	.27906

a. Predictors: (Constant), MW
b. Predictors: (Constant), MW, V
c. Predictors: (Constant), MW, V, HA

This finding was also supported by Nadia R. Pereira., Antonio Marsaioli Jr. and Ahrne (2007) who stated that MW power on the drying rate was much stronger than that of the air temperature and velocity. By comparing three different models, it is obvious that MW power level influences the D_{eff} in larger extent while the variation of HA temperature has lesser impact in affecting the D_{eff} in both case studies. This shows that MW is the main drying variable in governing the D_{eff} and this is followed by air velocity and lastly by HA temperature.

Table 4.9: Coefficients for Model 3

Model		Unstandardized Coefficients		Standardized Coefficients	t	Sig.
		B	Std. Error	Beta		
i	(Constant)	-3.841	1.687		-2.277	.057
	MW	.011	.002	.861	4.478	.003
ii	(Constant)	-2.637	1.324		-1.991	.094
	MW	.011	.002	.861	6.081	.001
	V	-.334	.127	-.372	-2.628	.039
iii	(Constant)	-4.564	1.062		-4.296	.008
	MW	.011	.001	.861	9.410	.000
	V	-.480	.095	-.534	-5.052	.004
	HA	.054	.018	.323	3.061	.028

a. Dependent Variable: D_{eff_3}

On the other hand, SEC is placed as a performance indicator to evaluate the efficiency of energy consumption. Hence, variables that contribute most significantly to the SEC values are presented in Equation 4.15 (Model 4) and Equation 4.16 (Model 5), respectively. With reference to Table 4.10, two-stage drying in Model 4 shows 89.9% of changes in SEC value is due to the change of HA temperature and MW power level.

$$\text{Model } 4_{SEC1} = -0.715MW - 0.623HA \tag{4.15}$$

Table 4.10: R^2 values for Model 4

Model	R	R Square	Adjusted R Square	Std. Error of the Estimate
4	.948[a]	.899	.832	.0226039

a. Predictors: (Constant), HA, MW

Moreover, T-test analysis in Table 4.11 shows both variety of MW power level and HA temperature has a considerable effect on SEC value ($p < 0.05$ level of significance). Hence, the energy consumption in two stage drying are being affected by the temperature level and the MW power level.

Table 4.11: Coefficients for Model 4

Model		Unstandardized Coefficients		Standardized Coefficients	t	Sig.
		B	Std. Error	Beta		
4	(Constant)	1.340	.210		6.397	.008
	MW	.000	.000	-.715	-3.895	.030
	HA	-.006	.002	-.623	-3.397	.043

a. Dependent Variable: SEC_1

Model 5 is expressed in Equation 4.16 which describe the relationship between SEC and three different variables such as velocity, MW power and HA temperature.

$$\text{Model } 5_{SEC2} = 0.669V - 0.552MW + 0.246HA \quad (4.16)$$

With reference to Table 4.12, it was discovered that 62.7% of the change in the SEC value is caused by the variation of air velocity, V. Association of both V and MW power level contributes (93.2-62.7%) 30.5% additional changes in SEC. However, HA temperature only has slight contribution i.e. (97.7-93.2%) 4.5% of the variance in SEC value.

Table 4.12: R^2 values for Model 5

Model	R	R Square	Adjusted R Square	Std. Error of the Estimate
i	.792[a]	.627	.574	.02060
ii	.965[b]	.932	.909	.00952
iii	.989[c]	.977	.964	.00601

a. Predictors: (Constant), V
b. Predictors: (Constant), V, MW
c. Predictors: (Constant), V, MW, HA

It was deduced that slight decrease or increase in air velocity could have a prevalent effect on the SEC values. This finding is corresponding to the data reported in Section 4.5 where the slight reduction of SEC value was observed when air velocity reduced from 5.0 m/s to 2.8 m/s in all drying conditions.

Table 4.13: Coefficients for Model 5

Model		Unstandardized Coefficients		Standardized Coefficients	t	Sig.
		B	Std. Error	Beta		
i	(Constant)	.096	.023		4.197	.004
	V	.021	.006	.792	3.433	.011
ii	(Constant)	.237	.029		8.117	.000
	V	.021	.003	.792	7.430	.000
	MW	.000	.000	-.552	-5.176	.002
iii	(Constant)	.194	.023		8.478	.000
	V	.018	.002	.669	8.606	.000
	MW	.000	.000	-.552	-8.199	.000
	HA	.001	.000	.246	3.171	.025

a. Dependent Variable: SEC_2

Furthermore, T-test analysis in Table 4.13 depicts a variety of MW power level, air velocity and HA temperature has a considerable effect on SEC value ($p < 0.05$ level of significance). Through multiple regression analysis, the interrelationship between the independent variable and dependent variable in microwave assisted drying is simplified and formulated in the forms of the model. For instance, due to the larger influential of MW in the beginning of drying process (MW/HA), the variance of D_{eff} is much lower in comparison to HA/MW. Whereas the velocity and MW energy are both significant in influencing the SEC value in MWHA. Thus, this statistical technique might be useful in the selection of optimal drying method and drying process can be designed based on the drying parameters that produce a significant overall effect to the required key performance.

CHAPTER 5

CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

Drying of crumb rubber is an energy extensive process that is generally completed in a trolley dryer with hot air as a medium to dehydrate the wet rubber. The finished product which is in the form of block rubber must possesses good elasticity, high ageing and thermal resistance which can provide good processability, particularly in the tyre industry. To find an alternative way for block rubber processing other than HA, the application of electromagnet wave viz. MW as a prime heating source was attempted and investigated in this study. This helps in developing an efficient drying technique that reduces the drying time, energy consumption and to produce the desirable dried rubber. Various drying techniques ranging from single stage drying, two-stage drying and hybrid drying have been applied in this research. The research quantifies the speed of drying for the different combination strategies, determine the effect of MW on the drying patterns, physicochemical properties and delineate the nature of individual drying mode and their combinations. Some significant findings from the research are summarized as below:

I. Rapid drying of crumb rubber could be achieved by microwave assisted drying

The drying experiments using microwave-assisted drying have demonstrated significant improvement in time-saving and drying rate. The drying time of MWHA was found to be 89% and 50% shorter than single stage HA and MW, respectively. Saving of drying time was indeed largely contributed by the introduction of MW in the drying process. Evidently, MW assisted drying appreciably reduce drying time by a factor of five or more in comparison to the conventional HA method. The result is consistent with the drying rate in which the maximum drying rate of MWHA was found to be 455% and 62.5% higher than HA and MW drying, respectively. Meanwhile, effective diffusivity, D_{eff} values of both MW and MWHA were ranging from 1.48 x 10^{-8} m^2/s to 5.59 x 10^{-8} m^2/s that is more than six times higher than HA which has the lowest D_{eff} recorded at 0.22 x 10^{-8} m^2/s. Furthermore, 73% of drying methods were found best fitted to the Hii and Law model which can be used to describe the thin layer drying curve of crumb rubber.

II. Significant energy saving can be attained by application of MW to assist the drying process and by varying the effective drying time

Two-stage drying especially vacuum assisted drying gave high SEC values than other methods which scored at 0.75 MJ/ gH_2O. Likewise, single stage continuous MW drying and HA drying yields SEC values of 0.16-0.27 MJ/gH_2O and 0.36-0.44 MJ/gH_2O, respectively. Conversely, average SEC values of MWHA's group were scored below 0.22 MJ/

gH_2O. The influences of both air velocity and MW to the SEC was significant ($p < 0.05$ level of significance) and confirmed by using Multiple Regression Analysis. Besides, it is noteworthy to mention that by varying the power level of MW and drying time at different stages, a significant reduction of SEC values can be obtained. This was confirmed by the SEC value of both MW and MWHA multi-steps drying were calculated at 0.08-0.09 MJ/gH_2O and 0.14-0.15 MJ/gH_2O, respectively.

III. Good retention of physicochemical and mechanical properties could be obtained by using MWHA drying

By comparing to other drying methods, dried rubber by using MWHA exhibited lightest colours, minimum ΔE^* and consistent colour appearance. The colour retention of MWHA dried rubber was largely contributed by the hot air introduced into the MW cavity which generates a cooling effect on the sample surface to prevent concentrated heating that leads to charring. Indeed, lesser colour change can be an indication of reduced oxidative reaction and hence better ageing properties of dried rubber. This corresponds to the high value of E_a obtained by MWHA in the thermal degradation study by using the Coats-Redfern methods. Similarly, the rheological properties of MWHA dried rubber including PRI and P_o were found in par with SMR 20 block rubber (industry standard) and the volatile matter was 28.6% lower than the SMR 20 block rubber (industry standard). In terms of springiness, MWHA dried rubbers recorded the highest value at 72.23% which indicates marked performance in mechanical properties.

5.2 Recommendations

Drying of crumb rubber is often a complex process which is relatively challenging to control and monitor. The drying process can be influenced by different components such as the mechanism of heat and mass transfer, a different clone of raw rubber, equipment used and the physicochemical properties prior to the drying process. This research demonstrated the potential of MWHA as an alternative method for drying crumb rubber. It has fortified our understanding on microwave-assisted drying of natural rubber and properties of dried rubber by using the method.

There is a possibility for future works from which this research can be built upon.

- ✓ **Validation of microwave-assisted drying process based on multi-physics modelling simulated through Finite Element Analysis (FEA)**

  Establishing a robust multi-physic model in describing the drying mechanism is of great interest. A physic based computer modelling can be used to predict the drying mechanism and parameter changes by modelling the drying equipment and sample in a virtual environment. Therefore, simulated results can be used to validate and improve the existing drying parameters to maximize the drying performance while maintaining the quality of the dried rubber.

- ✓ **Economic analyses of MWHA drying process for pilot scale studies**

  Economic viability analysis can be performed to measure the overall desirability of the drying process in financial terms and specifies the

advantage of a single approach over others to provide a decision-making tool for a pilot project as well as for demonstration purposes.

- ✓ **Incorporation of Analytical Hierarchy Process (AHP) for multi-criteria evaluation**

  Use of statistical analysis tool is an easier way to identify the desirable of a drying process upon some certain conditions, however, the solution might have been oversimplified or misinterpreted. On the other hand, AHP is one of the useful decision-making tools for integrating both quantitative and qualitative data. It decomposes complex decision criteria into a hierarchy where the user can individually investigate each of the criteria. Some examples of decision criteria can be based on cost, energy impacts, technological capability and quality control. This allows comprehensive and systematic interpretation of the user based on the correlation between quantitative and qualitative factors in selecting optimal drying solution.

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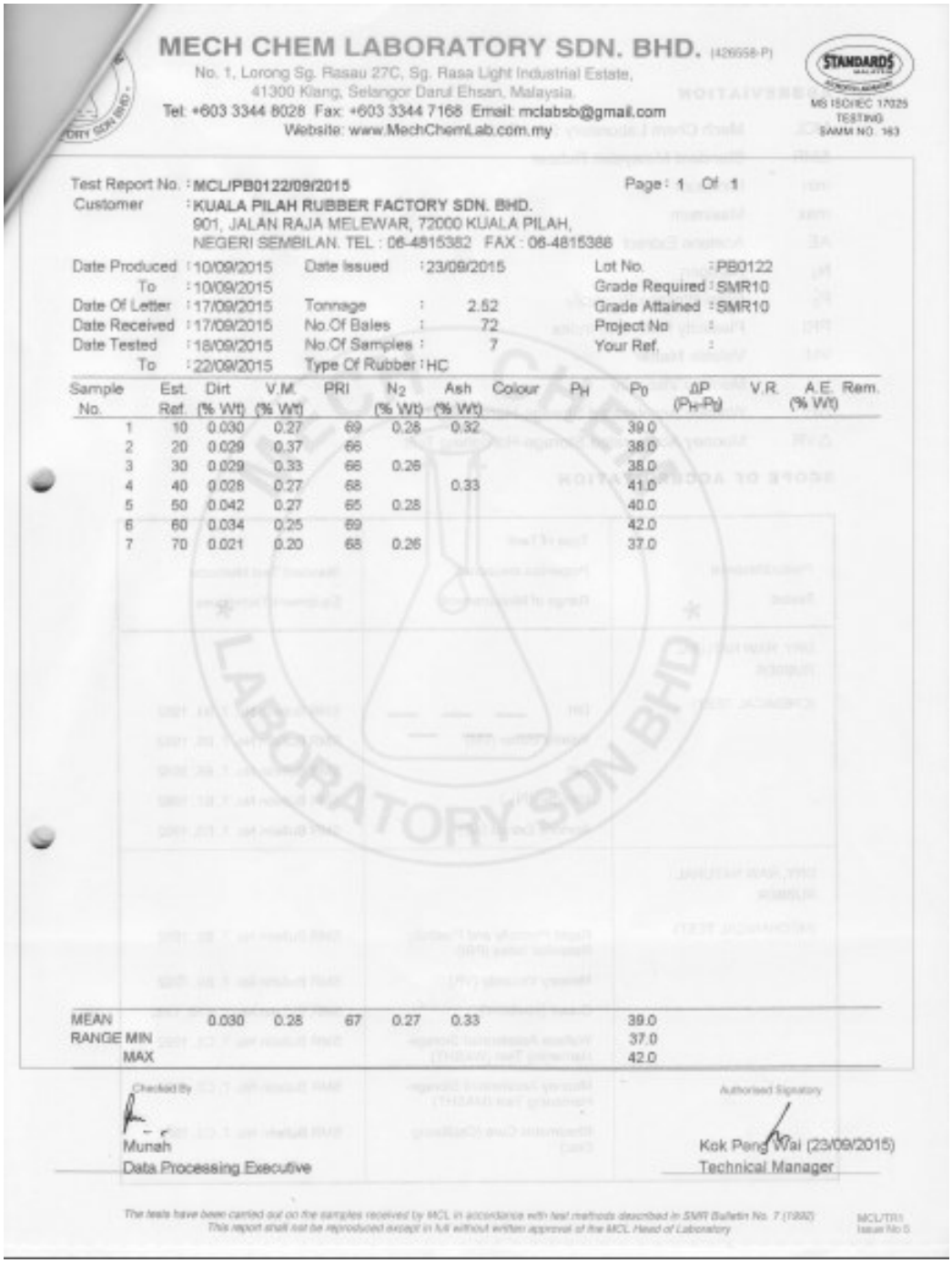
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APPENDICES



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Website: www.MechChemLab.com.my

STANDARDS MALAYSIA — MS ISO/IEC 17025 TESTING SAMM NO. 163

Test Report No. : MCL/PB0122/09/2015 Page : 1 Of 1

Customer : KUALA PILAH RUBBER FACTORY SDN. BHD.
901, JALAN RAJA MELEWAR, 72000 KUALA PILAH,
NEGERI SEMBILAN. TEL : 06-4815382 FAX : 06-4815388

Date Produced	: 10/09/2015	Date Issued	: 23/09/2015	Lot No.	: PB0122
To	: 10/09/2015			Grade Required	: SMR10
Date Of Letter	: 17/09/2015	Tonnage	: 2.52	Grade Attained	: SMR10
Date Received	: 17/09/2015	No.Of Bales	: 72	Project No	:
Date Tested	: 18/09/2015	No.Of Samples	: 7	Your Ref.	:
To	: 22/09/2015	Type Of Rubber	: HC		

Sample No.	Est. Ref.	Dirt (% Wt)	V.M. (% Wt)	PRI	N_2 (% Wt)	Ash (% Wt)	Colour	P_H	P_0	ΔP (P_H-P_0)	V.R.	A.E. (% Wt)	Rem.
1	10	0.030	0.27	69	0.28	0.32			39.0				
2	20	0.029	0.37	66					38.0				
3	30	0.029	0.33	66	0.26				38.0				
4	40	0.028	0.27	68		0.33			41.0				
5	50	0.042	0.27	65	0.28				40.0				
6	60	0.034	0.25	69					42.0				
7	70	0.021	0.20	68	0.26				37.0				
MEAN		0.030	0.28	67	0.27	0.33			39.0				
RANGE MIN									37.0				
MAX									42.0				

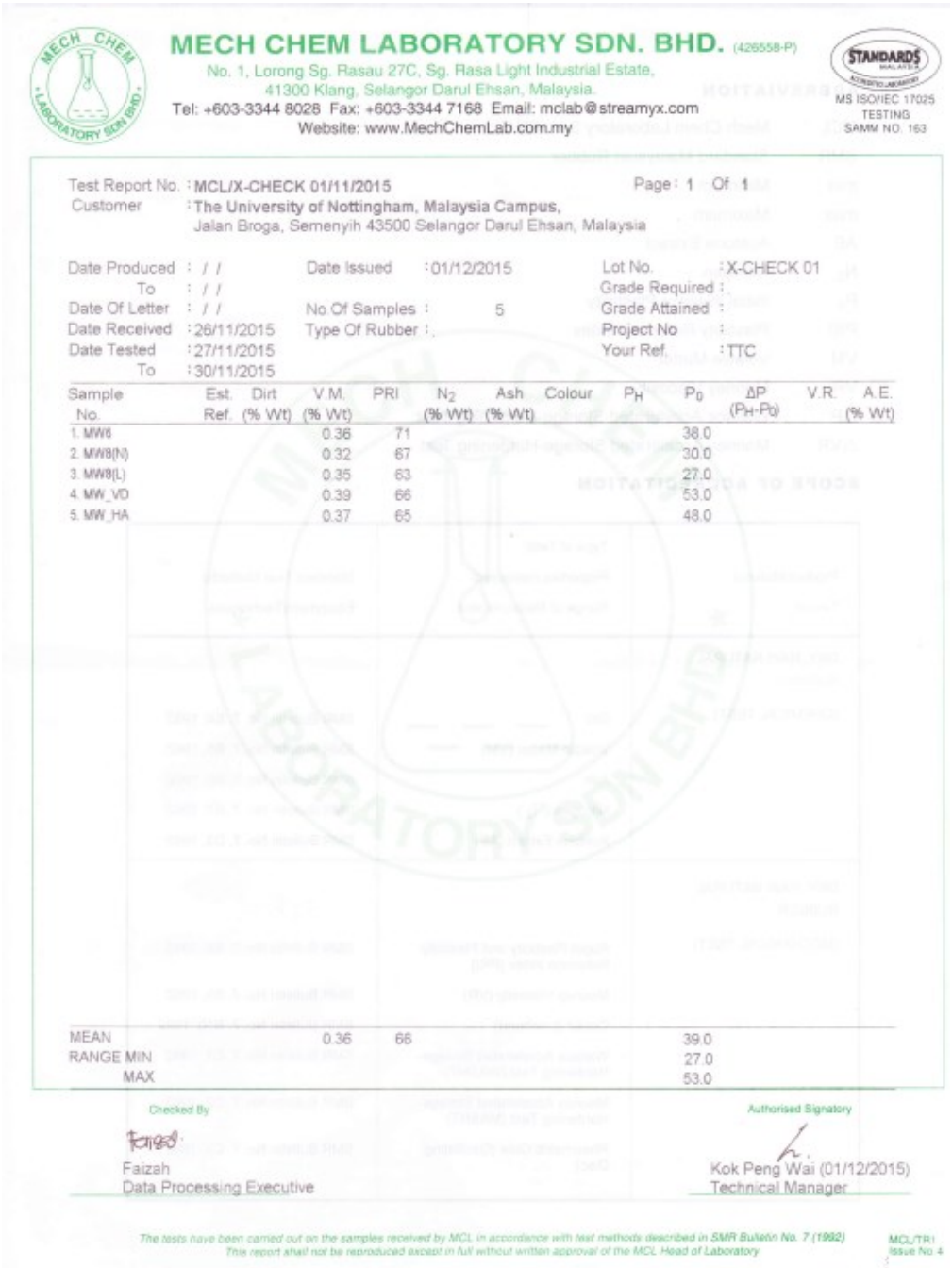
Checked By
Munah
Data Processing Executive

Authorised Signatory
Kok Peng Wai (23/09/2015)
Technical Manager

The tests have been carried out on the samples received by MCL in accordance with test methods described in SMR Bulletin No. 7 (1988)
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MCL/TR1 Issue No 0

Figure A1: Technological properties of SMR graded rubber (Courtesy of Kuala Pilah Rubber Factory Sdn. Bhd.)



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Website: www.MechChemLab.com.my

MS ISO/IEC 17025
TESTING
SAMM NO. 163

Test Report No. : MCL/X-CHECK 01/11/2015 Page : 1 Of 1
Customer : The University of Nottingham, Malaysia Campus,
Jalan Broga, Semenyih 43500 Selangor Darul Ehsan, Malaysia

Date Produced : / /
To : / /
Date Of Letter : / /
Date Received : 26/11/2015
Date Tested : 27/11/2015
To : 30/11/2015

Date Issued : 01/12/2015
No Of Samples : 5
Type Of Rubber :

Lot No. : X-CHECK 01
Grade Required :
Grade Attained :
Project No :
Your Ref. : TTC

Sample No.	Est. Ref.	Dirt (% Wt)	V.M. (% Wt)	PRI	N_2 (% Wt)	Ash (% Wt)	Colour	P_H	P_0	ΔP (P_H-P_0)	V.R.	A.E. (% Wt)
1. MW6			0.36	71					38.0			
2. MW8(N)			0.32	67					30.0			
3. MW8(L)			0.35	63					27.0			
4. MW_VD			0.39	66					53.0			
5. MW_HA			0.37	65					48.0			
MEAN			0.36	66					39.0			
RANGE MIN									27.0			
MAX									53.0			

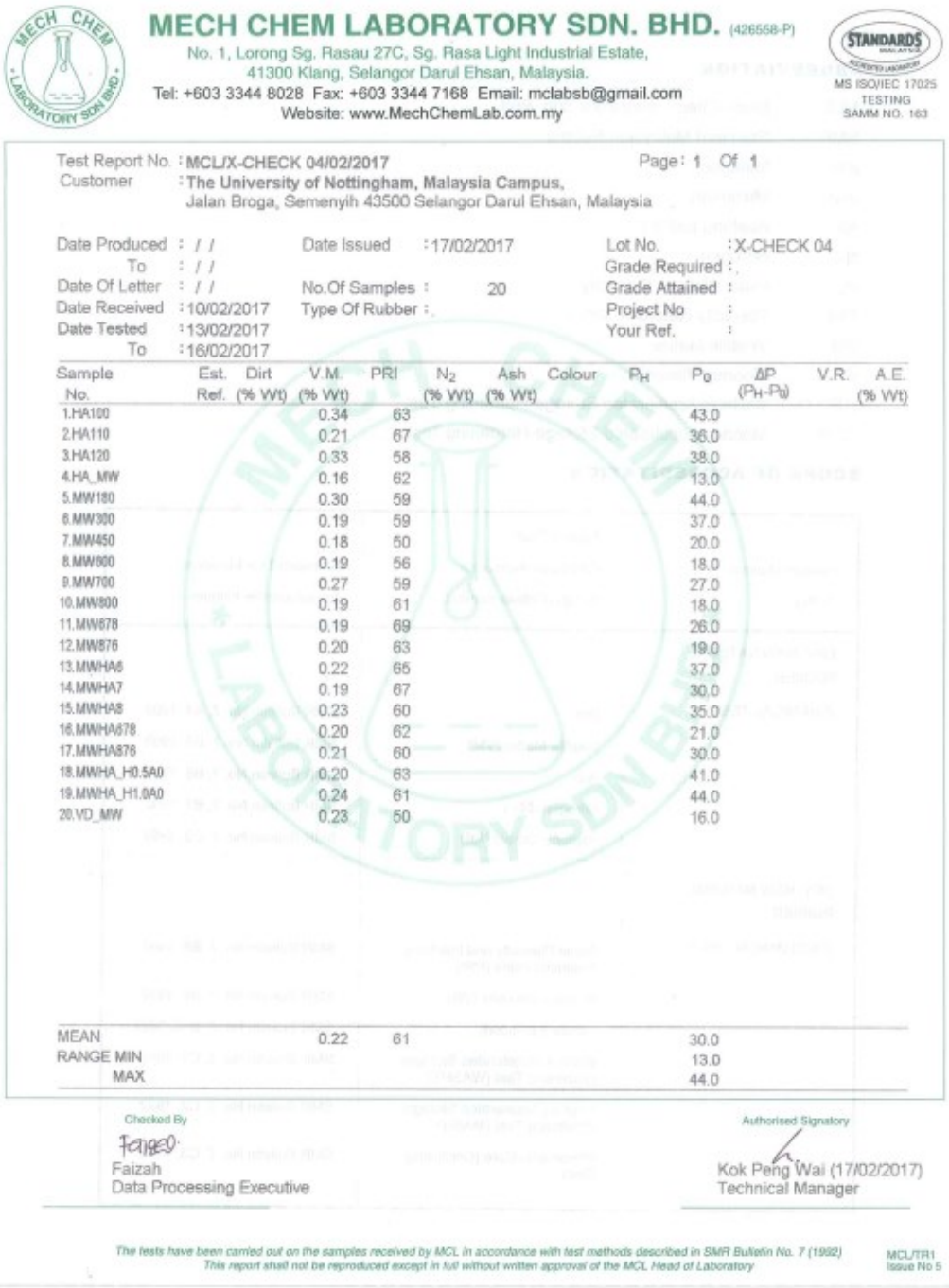
Checked By
Faizah
Data Processing Executive

Authorised Signatory
Kok Peng Wai (01/12/2015)
Technical Manager

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MCL/TR1
Issue No 4

Figure A2: Technological properties of experimental dried rubber (Set 1)



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Website: www.MechChemLab.com.my

STANDARDS
MS ISO/IEC 17025
TESTING
SAMM NO. 163

Test Report No. : MCL/X-CHECK 04/02/2017 Page : 1 Of 1
Customer : The University of Nottingham, Malaysia Campus,
Jalan Broga, Semenyih 43500 Selangor Darul Ehsan, Malaysia

Date Produced	: / /	Date Issued	: 17/02/2017	Lot No.	: X-CHECK 04
To	: / /			Grade Required	:
Date Of Letter	: / /	No.Of Samples	: 20	Grade Attained	:
Date Received	: 10/02/2017	Type Of Rubber	:	Project No	:
Date Tested	: 13/02/2017			Your Ref.	:
To	: 16/02/2017				

Sample No.	Est. Dirt Ref. (% Wt)	V.M. (% Wt)	PRI	N_2 (% Wt)	Ash (% Wt)	Colour	P_H	P_0	ΔP (P_H-P_0)	V.R.	A.E. (% Wt)
1.HA100		0.34	63					43.0			
2.HA110		0.21	67					36.0			
3.HA120		0.33	58					38.0			
4.HA_MW		0.16	62					13.0			
5.MW180		0.30	59					44.0			
6.MW300		0.19	59					37.0			
7.MW450		0.18	50					20.0			
8.MW600		0.19	56					18.0			
9.MW700		0.27	59					27.0			
10.MW800		0.19	61					18.0			
11.MW878		0.19	69					26.0			
12.MW876		0.20	63					19.0			
13.MWHA6		0.22	65					37.0			
14.MWHA7		0.19	67					30.0			
15.MWHA8		0.23	60					35.0			
16.MWHA678		0.20	62					21.0			
17.MWHA876		0.21	60					30.0			
18.MWHA_H0.5A0		0.20	63					41.0			
19.MWHA_H1.0A0		0.24	61					44.0			
20.VD_MW		0.23	50					16.0			
MEAN		0.22	61					30.0			
RANGE MIN								13.0			
MAX								44.0			

Checked By
Faizah
Data Processing Executive

Authorised Signatory
Kok Peng Wai (17/02/2017)
Technical Manager

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Issue No 5

Figure A3: Technological properties of experimental dried rubber (Set 2)

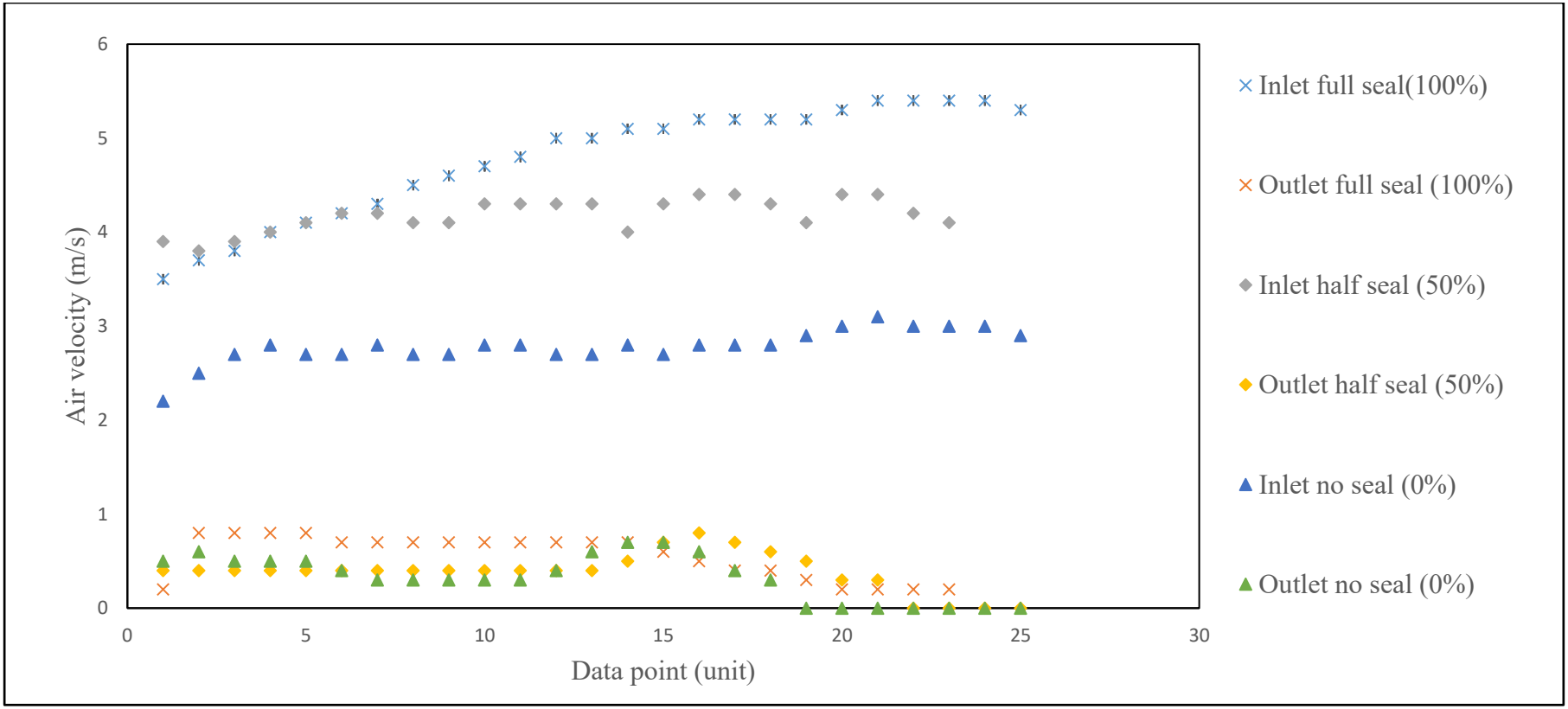


Figure B1: Air velocity measured inside the laboratory scale table top microwave-convective dryer

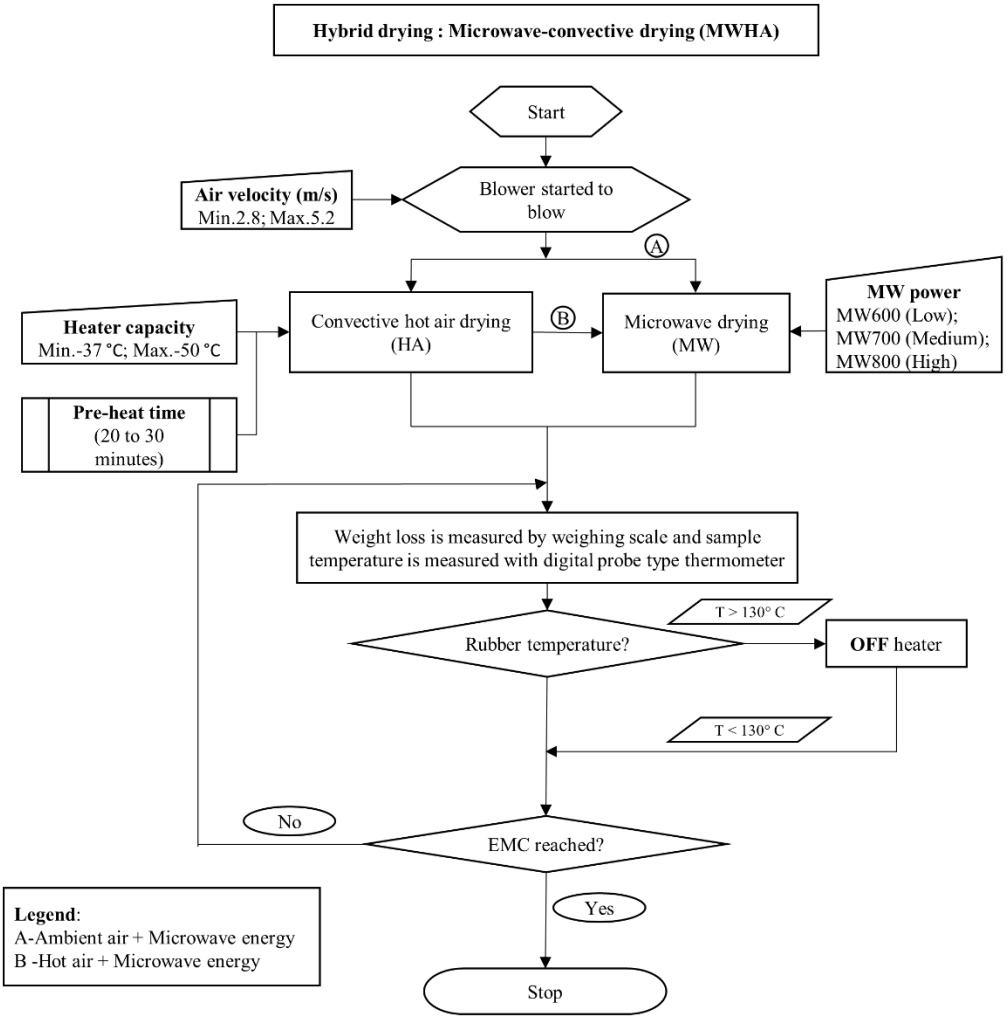


Figure B2: Process flowchart of MWHA



Figure C1: Fresh crumb natural rubber



Figure C2: Dried natural rubber