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A Review of the Parameters Related to the Rubber Sheets Production: Major Constituents of Rubber Latex, Rubber Particles, Particle Interaction of Rubber Latex, Rubber Porous Structure, Rubber Drying Kinetics and Energy Consumption

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ABSTRACT

Natural rubber (NR) is obtained principally from Para-rubber trees of the species Hevea brasiliensis which grow in tropical regions. Using ultra-centrifugation method, fresh latex (FL) from Para-rubber trees can be mainly divided into four fractions; an upper white layer consists of rubber particles, Frey-Wyssling particles in a yellow or an orange layer, C-serum phase and lutiods particles in the bottom fraction [1-3]. The averages of rubber particles have diameters of 0.02 to 3.0 micron, and they are protected by a complex film containing lipids and proteins [4-8]. The main forces of attraction between neighboring rubber particles in latex system can be divided into five force types; Structural Forces (SF), Van der Waals Interaction (VWI), Electrostatic Force (EF), Exclusion Interaction (EI) and Polymer-Polymer Interaction (PPI) [9]. The porous model of rubber structure used to describe moisture transfer was based on the existence of two different regions referred to as non-hygroscopic region and hygroscopic region. The rubber products drying always produced a considerable shrinkage effect which considered in the physical of the product, such as the diffusion coefficient, mass and heat transfer. An initial moisture content of raw material, the experimental temperature and the drying equipment had affect to the EMC isotherms and the drying kinetics. Finally, discussions on the implications of the results for strategies to reduce the energy consumption in RSS and STR20 block rubber are also presented.

1. Introduction

Polymer of isoprene $(C_5H_8)_n$ is the primary chemical constituent of NR (cis-1,4-polyisoprene). NR is an essential raw material for polymer industry and it produced by over 2,500 different latex-producing crop species [4, 10], such as Euphorbia lactfiua [6], Ficus elastica or the Indian rubber trees

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[11-13], Hevea brasiliensis or the Brazilian rubber trees [14-16], Parthenium argentatum gray or the little desert shrub Guayule [17-19], Ficus carica [20-21], Ficus racemose [22], and Ficus benghalensis [23]. The history of NR production shows the attempts to identify alternate sources of rubber both tropical and temperate [24-26], but a few crop species known to produce NR is capable of producing large amounts of high molecular weight rubber [6, 15].

NR from Para-rubber trees (the Brazilian rubber trees or Hevea brasiliensis), one of the NR species, is an important source of NR due to its excellent physical properties. Fresh NR latex consists of approximately 94% rubber hydrocarbon and 6% non-rubber components, for example, fatty acids, glycolipids, carbohydrates, phospholipids and proteins [27-29]. The raw NR is largely used in many products such as medical gloves, rubber bands, condoms, flexible tubing, toys and vehicle tires [30-34]. It is presented in form of white latex liquid (FL) when it is tapped and collected from Para-rubber trees. The life of fresh NR latex is as short as one day if it is not properly preserved [14, 31, 33-35]. Several forms of preserved fresh NR latex have been created to extend its shelf life before it is consumed by the production of the products noted above. These forms include the latex production such as skim latex (SL) and concentrated latex (CL), the rubber sheets production; ribbed smoked sheet (RSS) and air dried sheet (ADS); and the block rubber production such as Standard Thai Rubber (STR) 20. In this review, we are focus on the rubber sheets production (ADS and RSS) and STR20 block rubber production because they are the highest forms of NR production in Thailand [36].

There are several parameters such as the major constituents and particle interaction of rubber latex, rubber particles, the rubber porous structure, the rubber drying kinetics, etc. would control the drying energy consumption and the quality of NR product. The reports of several researchers [37-41] presented that the porosity, the product structure (major constituents, product particles and particle interaction) and shrinkage have affected to the drying characteristics such as critical water content, drying rate, air velocities, drying temperatures and an effective diffusion coefficient (EDC). Besides the drying characteristics, the energy consumption and quality of the product has affected to production costs [42-43]. The knowledge of the drying process and drying kinetic for maintaining high quality products is essential for the optimization of the drying process. Therefore, understanding the rubber drying kinetics is necessary to decrease energy consumption and to enhance the quality of the products [44-45].

Generally, moisture transfer in porous media of the natural products occurs mostly in their mechanisms and falling drying rate period are described by liquid diffusion. Mechanisms of this diffusion were reported by several researchers [46-51]. Addition, Philpott and Walker [52] suggested that non-rubber components would prevent moisture transfer on the surface of the rubber sheets as same as the report of Ansari *et al.,* [53] and Wichaita *et al.,* [54]. Therefore, the major constituents of rubber latex, rubber particles, particle interaction of rubber latex, the rubber porous structure, the rubber drying kinetics and the energy consumption of rubber sheets and STR 20 block rubber will be reviewed.

2. Major Constituents of Rubber Latex

Latex that flows out of the Para-rubber tree is a complex form of cytoplasm. It is containing a suspension of rubber and non-rubber particles in an aqueous serum [3, 5-6, 55-56]. Using ultra-centrifugation method, Cook and Sekhar [10] report showed that NR latex could be divided into four fractions (three particulate fractions and a serum phase); an upper white layer consists of rubber particles, Frey-Wyssling particles in a yellow or an orange layer, C-serum phase and lutiods particles in the bottom fraction [1-3, 55]. In this section, the study of nature and composition of rubber latex is reviewed. Many reports of researchers are discussed as follows.

In 1959, the study of Moir [57] presented that NR latex could be divided into eleven zones when using ultra-centrifugation method (Figure 1). In this figure, the whitish fraction appeared in the top (first) zone. It was consisted mainly of hydrocarbon particles. This result agreed with of Cook and Sekhar report [1]. Second zone was situated under the lowest portion of the top zone. It was a translucent layer and much small. Third zone was a suspension of rubber particles in the serum. Sometimes the first to third zone was called a white upper layer of rubber cream [58]. Forth zone was Frey-Wyssling particles in a yellow or an orange layer. The fifth zone was a serum phase named C-serum and the sixth to the eleventh zones together was called the bottom fraction. It mainly consisted of lutiods particles in grayish yellow gelatinous sediments [1-2, 58].

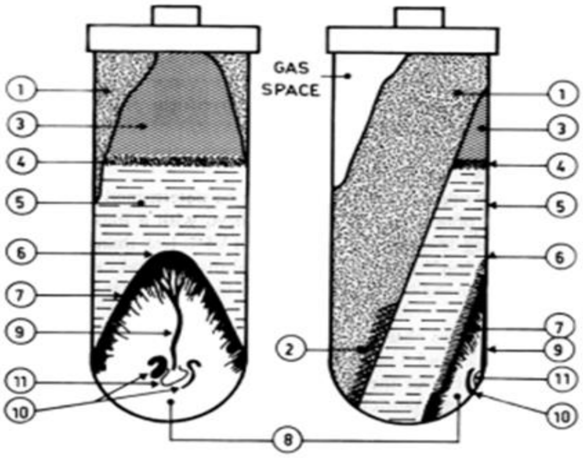

Fig. 1. Schematic shows the latex separation by ultra-centrifugation [57]

Investigated by Dai *et al.,* [59], Para-rubber latex could be divided into thirteen layers. In this report, the lutiods particles in grayish yellow gelatinous sediments were found to be damaged when using isopycnic centrifugation method. The studies of Low and Wiemken [60] presented that Para-rubber latex could be divided into thirteen fractions as same as the results from Dai *et al.,* [59]. In their report, the lutiods particles in the bottom fraction were successfully separated when using ficoll separation method. Investigated by Rippel and Galembeck [61], Para-rubber latex from clones of Hevea brasiliensis in Brazil could be divided into four fractions as same as the results from Cook and Sekhar [1]. For the structure, NR was a complex nanostructured material formed by polymer of isoprene, phospholipids, protein and a host of mineral minor components.

In 2017, the study of Liengprayoon *et al.,* [62] showed that Para-rubber latex from clones of Hevea brasiliensis in Thailand (RRIM600 and PB235) were divided into four fractions when through successive centrifugation steps and their result agreed with of Cook and Sekhar [1] and Rippel and Galembeck [61]. For non-rubber fractions, the lutoid particles were the highest in protein, lipids and minerals (mainly Magnesium and Potassium).

3. Rubber Particles

The major component of Para-rubber (Hevea brasiliensis) latex is rubber hydrocarbon particles. Figure 2 shows that the rubber particles are usually spherical [6]. The averages of these particles have diameters of 0.02 to 3.0 micron, and they are protected by a complex film containing lipids and proteins [2, 4-6, 8]. This complex film imparts a net negative charge to the rubber particles contributing colloidal stability to the liquid [63-64]. Para-rubber latex has variable composition depending on several factors, for example, age of the tree [65], season [66-67], soil conditions, tapping and frequency [68]. In this section, the study of rubber hydrocarbon particles in fresh latex of Para-rubber (Hevea brasiliensis) is reviewed. Many reports of researchers are discussed as follows.

In 1961, the study of Southorn showed that the shape of rubber particles was spherical bodies as same as the results from Pakianathan *et al.,* [69] and Cornish *et al.,* [6]. Pendle and Swinyard [70] presented that the fresh latex was consisted of rubber hydrocarbon particles which had sizes ranging from 0.01 to 5 micron. These particles had mainly size in range of 0.1 to 2 micron.

After centrifuging, Sakdapipanich *et al.,* [71-72] showed that the fresh latex was composed of small rubber particles in range of 0.04 to 0.4 micron with the mean particle diameter of about 0.1 micron. Those of large rubber particles in concentrated latex were from 0.1 to 5 micron with the mean particle diameter of about 1 micron. For Another phase after centrifuging, the serum phase, Yeang *et al.,* [73] presented that the serum contained most of the soluble substances including organic acids, amino acids, carbohydrates, proteins, inorganic salts and nucleotides-materials.

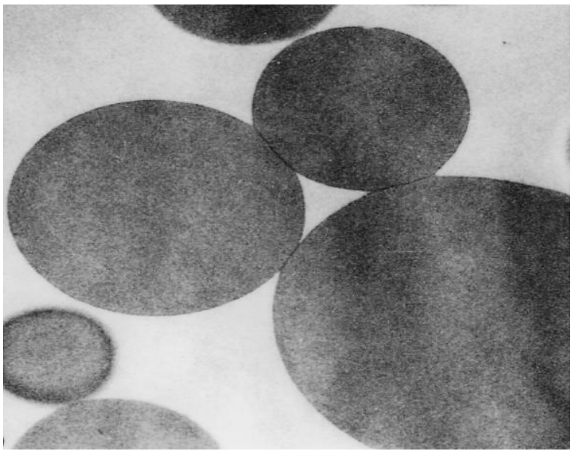

Fig. 2. The rubber particles [6]

4. Particle Interaction of Rubber Latex

Fresh NR latex from the Para-rubber tree is a thixotropic neutral liquid of density around 0.98 g/cm^3. It has a pH of 6.5 to 7.0 and molecular weight distribution 10^5 to 10^7 g/mol [74-76]. Besides rubber polymers $(C_5H_8)_n$, NR latex contains carbohydrates, fatty acids, glycolipids, phospholipids, proteins, lutoids, inorganic salts and other components [6, 77-79]. Latex can be maintained by the main forces of attraction between neighboring rubber particles that are encased with lipids and proteins [80]. In this section, the main forces of attraction between neighboring rubber particles (main inter-particle forces commonly appearing in rubber latex) are reviewed.

In 2002, the study of Quemada and Berli showed that the main forces of attraction between neighboring rubber particles in latex system could be divided into five force types; SF, VWI, EF, EI and PPI. For the SF or hydrophilic interaction, it was pertinent for majority systems that had an aqueous phase. It happened from the greatly hydrophilic surfaces which caused molecular order in the contiguous and neighboring water molecules. The superficial hydration leaded to a repulsive force between surfaces, which decayed exponentially with a characteristic length [81-84]. For VWI, it related to an attractive force between any two bodies of finite mass. VWI were included between London forces and the Keesom orientation forces [85-88]. The report of Leite *et al.,* [87] showed that VWI was reduced rapidly and it was not relatively important at large distance between center to center of particles. For EF, the report of Shubin and Hunter [89] presented that the EF happened from the presence of electric charges (either positive or negative) bound at the surface of the particles. In aqueous media, electrically charged particles were surrounded by electrolyte-ions and counter-ions. As two particles approach each other, the report of Israelachvili [90] described that the overlapping of double layers leaded to long-range repulsive forces due to entropic effect. For interaction in colloid system was depletion or EI, the report of Schramm and Shi [86] presented when polymer molecules were added to adjacent surfaces, an attractive force was created between the surfaces. This mechanism was involved either depletion or bridging, depending on the net interaction between the solvent, macromolecules and the particles. If particles were relatively large when compared with the polymer, attractive particle-particle forces were arisen by the mechanism of depletion [90-93]. For PPI, the reports of Israelachvili [90] and Quemada and Berli [9] showed that this interaction was normal to the several colloidal systems, for example, those containing polymer-covered particles (absorbed or grafted), star polymers and micro-gels. This interaction related to the thickness of the layer formed by the surface to surface distance between cores and the polymer chains got stuck to the core.

5. The Porous Structure Modeling

The arrangement of pores within network is also important to transport of moisture. The product with different porosity can be affected to moisture adsorption rate and its textural properties [94-95]. An understanding relation of moisture transfer and its micro-structure prevent the quality change in the products [96-97]. In the present, the porous model technique is utilized to describe moisture transfer [98-101]. This technique is based on the existence of two different regions (non-hygroscopic and hygroscopic). For the hygroscopic region, the report of Sanavia *et al.,* [102]; Harun *et al.,* [103] and Goyeneche *et al.,* [104] presented that this region was characterized by the exclusive presence of bound water. For the non-hygroscopic region, the report of the report of Harun *et al.,* [103]; Venil *et al.,* [105] and Chen *et al.,* [106] presented that this region had free liquid water occupied the major portion of the pores in the medium and this water was retained by capillary forces. Note: the bound waters were adsorbed on the walls of the solid structure by EF and VWI under the form of multi-molecular layers for both regions (non-hygroscopic and hygroscopic).

For the rubber porous structure, Figure 3 shows the large void spaces between pore junction and pore-throat systems. In this figure, pore body systems are consisted of the large void spaces. Therefore, pore-throat systems are consisted of elongated void spaces connecting the large void spaces [109-111]. Two reports of researchers are discussed as follows.

Studied by Cousin *et al.,* [111], the author presented for the drying condition of crumb rubber. The mechanisms and models of crumb rubber producing STR20 were investigated by determining the capillary forces, relative humidity and temperature of both the rubber structure and the ambient air. This result showed that the drying rates of these samples varied with the rubber structure, relative humidity, drying time and temperature. The report of Naon *et al.,* [112] showed that the drying of NR and developed an empirical model to predict the conditions for drying. This result showed that the drying rates of NR varied with relative humidity, drying time and temperature and it was similar to the results from Cousin *et al.,* [111].

During drying, the researchers formulated the multi-porous media models to describe the mass and heat transfer process. These models considered for all phases of porous: gas, liquid water, and solid [113-115]. Air and water vapor was the components of gas phase. Figure 4 shows the schematic of the porous media and the associated modes of transport. The volume fraction of pores in a representative elementary volume (REV) of the material at any time is denoted by porosity, Φ as follow

$$\Phi = \frac{\Delta V_w + \Delta V_g}{\Delta V} \tag{1}$$

where ΔV_w is the volume occupied by liquid water in the REV and ΔV_g is the volume occupied by gas phases.

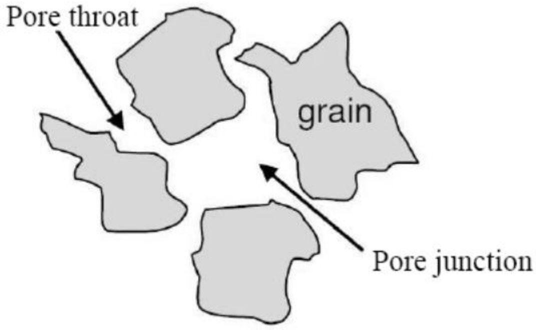

Fig. 3. Schematic shows large void spaces between pore junction and pore-throat systems [107]

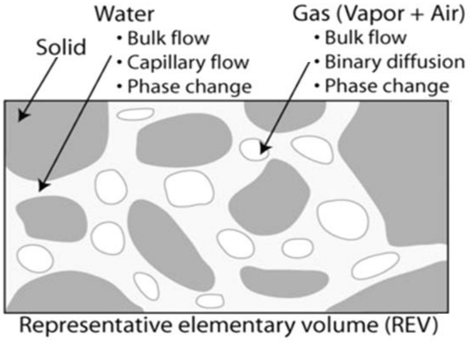

Fig. 4. The porous media and the associated modes of transport [108]

When the porosity of the product was changed, the structure of it was changed due to deformation. The volume of two phases (liquid water and gas) was changes. In contrast to the volume of the solid phase, it was constant [116]. So, the porosity, Φ at any time in a deforming medium can be calculated by Eq. (2).

$$\Phi = 1 - \frac{(1-\Phi_0)\Delta V_0}{\Delta V} \tag{2}$$

where Φ_0 is the initial porosity and ΔV_0 is the volume of a REV.

For the transportable phases, mass balance equation (MBE) includes the effects of binary diffusion (gas phase), capillary flow (liquid phase) and bulk flow (convection). For the liquid water phase, mass conservation equation (MCE) includes the phase change, capillary flow, and bulk flow. So, the equation is as Eq. (3) [117-118].

$$\frac{\partial c_w}{\partial t} + (v_w - v_s)\cdot\nabla c_w + c_w \nabla\cdot v_w = \nabla\cdot(D\nabla c_w) - \dot{N} \tag{3}$$

where c_w is the water phase concentration, v_w is the velocity of water phase, v_s is the velocity of solid phase, D is the capillary diffusivity and $\dot{N}$ is the volume of the evaporation rate.

Similarly, the continuity equation for the gas phase can be given by Eq. (4).

$$\frac{\partial c_g}{\partial t} + (v_g - v_s)\cdot\nabla c_g + c_g \nabla\cdot v_g = \dot{N} \tag{4}$$

where c_g is the gas phase concentration, and v_g is the velocity of gas phase.

Thus, MBE for the vapor component of the gas phase includes the phase change, binary diffusion and bulk flow can be written as Eq. 5) [118-119].

$$\frac{\partial c_v}{\partial t} + (v_g - v_s)\cdot\nabla c_g + c_g \nabla\cdot v_g = \nabla\cdot\left[S_g\left(\frac{C^2}{\rho_g}\right)\Phi M_a M_v D_g \nabla v\right] + \dot{N} \tag{5}$$

where c_v is the vapor concentration, S_g is the gas phase saturation, C is the molar density, ρ_g is the gas phase density, M_a is the molecular weight of air, M_v is the molecular weight of vapor, D_g is the effective gas diffusivity and v is the total velocity.

The vapor concentration, c_v is calculated from the relationship between the mass fraction of vapor with respect to total gas, ω_v and the gas phase concentration, c_g. The equation can be given by Eq. (6).

$$c_v = \omega_v c_g \tag{6}$$

Similarly, c_a is the concentration of air and it is calculated by Eq. (7).

$$c_a = \omega_a c_g \tag{7}$$

where ω_a is the mass fraction of air with respect to total gas and it is calculated by Eq. (8).

$$\omega_a = 1 - \omega_v \tag{8}$$

The report of Halder *et al.,* [120] showed that the porous media incorporates the change of phase between liquid water and vapor. This phenomenon was called condensation or evaporation. The equation can be given by Eq. (9).

$$\dot{N} = k_{ev}(p_v^* - p_v)\frac{M_v}{RT} \tag{9}$$

where k_{ev} is non-equilibrium evaporation constant, p_v is the vapor pressure, p_v^* is the equilibrium vapor pressure, R is the universal gas constant and T is the condensation or evaporation temperature.

Studied by Yutthana Tirawanichakul and Supawan Tirawanichakul [44], the author presented for MBE of the crumb rubber producing STR20. During drying, their report assumed that the shrinkage of samples was negligible and the moisture was transferred by liquid diffusion. Thus, the mass transfer between crumb rubber and an inlet hot air can be calculated by Eq. (10).

$$W_f = (M_i - M_f)r + W_{mix} \tag{10}$$

where W_f is humidity at final condition, W_{mix} is humidity of an inlet air temperature at the chamber, M_i is the initial moisture content, M_f is the moisture content at final condition and r is the ratio of dry sample mass to dry air mass.

The report of Tekasakul *et al.,* [121] showed that the equations governed mass and heat transfer of rubber sheets. During drying, their report assumed that the radiation effects were negligible as same as the shrinkage and deformation of the rubber sheets. Therefore, the equations were as follows [122-125].

$$\frac{1}{\alpha_r}\frac{\partial(\overline{T})}{\partial t} = \nabla\cdot(\nabla\overline{T}) \tag{11}$$

$$\frac{1}{D_r}\frac{\partial(\overline{M})}{\partial t} = \nabla\cdot(\nabla\overline{M}) \tag{12}$$

where α_r is the thermal diffusivity of rubber sheet; $\overline{T}$ is the temperature; D_r is the water diffusivity; and $\overline{M}$ is the moisture content.

For the water diffusivity of rubber sheets, it can be calculated by Eq. (13) [126].

$$D_r = 4\times10^{-5}\exp(-6158.47/T) \tag{13}$$

For energy conversation equation (ECE), it is derived from the first law of thermodynamics. For the gas phase, energy balance equation (EBE) is as Eq. (14) [125, 127-128].

$$\frac{\partial(\rho I)_g}{\partial t} = -\nabla\cdot\left[(\rho I)_g V_g + (I_v + I_a) j_v + j_h\right] - \rho_g \nabla\cdot V_g \tag{14}$$

where $(\rho I)_g$ is the specific internal energy (SIE) of the gas per unit volume of gas, ρ_g is the gas pressure, V_g is the gas volume, j_v is the sum of diffusive fluxes (SODF) of the vapor and j_h is the sum of diffusive heat fluxes (SODH).

$$(\rho I)_g \equiv (\rho I)_a + (\rho I)_v \tag{15}$$

where $(\rho I)_a$ and $(\rho I)_v$ is the sum of the corresponding specific energies of dry air and vapor, respectively.

$$j_a + j_v = 0 \tag{16}$$

where j_a is the SODF of air.

For Eq. (15) and Eq. (16), this equation can be assumed by Eq. (17).

$$p_a\nabla\cdot V_a + p_v\nabla\cdot V_v = p_g\nabla\cdot V_g \tag{17}$$

where p_a is air pressure, p_v is vapor pressure, V_a is the air volume and V_v is the vapor volume.

Similarly, EBE for the water phase can be calculated by Eq. (18) [125, 127-128].

$$\frac{\partial\theta_w(\rho I)_w}{\partial t} = -\nabla\cdot\left[(\rho I)_w q_w + \theta_w j_{hw}\right] - \dot{N} \tag{18}$$

where $(\rho I)_w$ is the SIE of the water per unit volume of water, j_{hw} is the SODH of the water, θ_w is the unit volume of water and q_w is the diffusive heat fluxes in water phase.

And EBE for the solid phase can be calculated by Eq. (19) [125, 127-128].

$$\frac{\partial\theta_s(\rho I)_s}{\partial t} = -\nabla\theta_s j_{hs} \tag{19}$$

where $(\rho I)_s$ is the SIE of the solid per unit volume of solid, j_{hs} is the SODH of the solid and θ_s is the unit volume of solid phase.

For EBE of the crumb rubber producing STR20 producing STR20, the report assumed that the temperature difference between air and grain of the sample was negligible (this sample was constant internal temperature). Thus, EBE can be calculated by Eq. (20) [44].

$$c_a^* T_{mix} + W_{mix}(h_{fg} + c_v^* T_{mix}) + r c_r^* T_0 = c_a^* T_{eq} + W_{mix}(h_{fg} + c_v^* T_{eq}) + r c_r^* T_{eq} \tag{20}$$

where h_{fg} is the specific evaporated enthalpy, c_v^*, c_a^* and c_r^* is the specific heat capacity (SHC) of vapor, air and wet product, respectively. T_{eq} is an equilibrium temperature and T_{mix} is an inlet air temperature at the chamber.

6. The Rubber Drying Kinetics

The drying of agricultural products such as rubber, vegetables and fruits, always produces a considerable shrinkage effect. This phenomenon considers in the physical of the product, such as the diffusion coefficient, mass and heat transfer. For some research reports [31, 37, 129-131], presented that shrinkage effect could be considered as directly related to the water volume removed during drying process. Other authors [38, 40, 42-43] had proposed a further component to the shrinkage phenomenon during drying besides the volume reduction due to the loss of moisture such as the mechanical forces [39, 132-134]. However, the mechanical shrinkage could be neglected if the delineation was focused on the drying kinetics [133, 135-137].

The basic method which describes the interaction between an agricultural material and water is the relationship between the water content of the mixture of water and material and the water activity, at a certain pressure and temperature [138-141]. This relationship is called the moisture sorption isotherm. It is an essential means for predicting the quality changes of products and evaluating chemical, physical and micro-biological stability during processing [142], as well as during storage. Moisture sorption isotherms are generally sigmoid in shape, a result of several basic interacting mechanisms of water binding. Some typical moisture sorption isotherms are shown in Figure 5. As temperature increases the amount of water adsorbed by a product increase and the isotherms shift upwards [143]. Experimental data can be fitted into models or equations that describe the moisture sorption isotherms in different ranges of relative humidity. Many researchers have determined the moisture humidity relationship for the rubber product, such as crumb rubber [144]; rubber stick [145] and rubber sheets [141].

Several methods are available for determination of water sorption isotherm (WSI). These methods may be divided into three categories: gravimetric, hygrometric and Nano-metric. Among these methods, gravimetric method is simplest to implement in a laboratory [141-142]. The gravimetric method involves the measurement of weight changes that can be measured both discontinuously and continuously in static and dynamic systems [145-146].

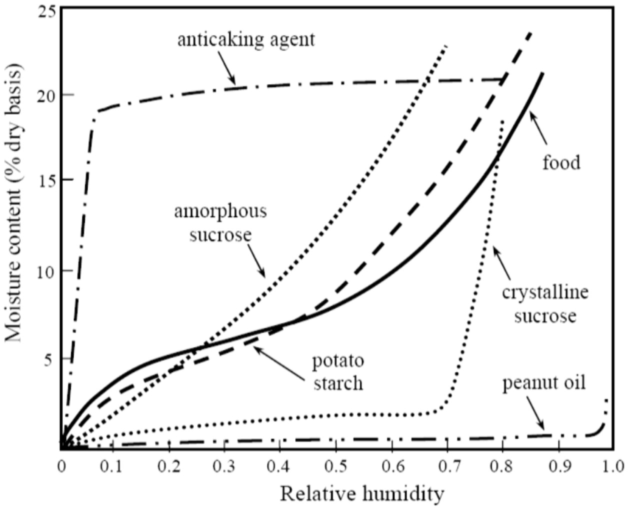

Fig. 5. The typical moisture isotherms for agriculture products [143]

The moisture content of the product in equilibrium with air is known as the equilibrium moisture content (EMC) and the relative humidity of air as equilibrium relative humidity (ERH). The relationship between vapor pressures, water activity and ERH is expressed as Eq. (21).

$$a_w = \frac{p}{p_0} = \frac{ERH}{100} \tag{21}$$

where a_w is water activity, p is the partial pressure of water vapor and p_0 is the vapor pressure of pure water at the same temperature.

For EMC of the rubber product, many researchers are determined by using static systems method; SSM [141, 147]. EMC is determined using saturated salt solution (SSS) such as Lithium-Chloride (LiCl), Sodium-Chloride (NaCl) and Potassium-Iodide (KI), etc. Equations for fitting WSI in agriculture products are of interest in many aspects of products preservation. Many Researchers attempted to describe the WSI mathematically. While some models have been derived theoretically based on thermodynamic concepts and others are extended or modified forms of these models. Table 1 presents the widely mathematical models. In this section, the review of the study of the EMC of the crumb rubber is presented. The reports of researchers are discussed as follows.

In 2007, Tirawanichakul *et al.,* [126] studied and developed the mathematical model of important parameters which affecting the crumb rubber producing STR20 drying process in terms of EMC. The EMC isotherms of crumb rubber were calculated by using SSM at temperature of 35, 40, 50, 55, and 60°C. The SSS were Ammonium-Sulfate ($(NH_4)_2SO_4$); Lithium-Chloride (LiCl); Potassium-Nitrate

(KNO_3); Sodium-Chloride (NaCl); Magnesium-Nitrate Hexahydrate ($Mg(NO_3)_2 \cdot 6H_2O$) and Magnesium-Chloride Hexahydrate ($MgCl_2 \cdot 6H_2O$). Four mathematical models; Chung and Pfost [148], Halsey [149], Henderson and Kaleemullah [150-152]; were used for prediction of the experimental values. This report presented that the model, namely Henderson was the most appropriate model for explaining the EMC isotherms. In contrast to Tirawanichakul *et al.,* [44, 147] studied and developed the mathematical model of important parameters which affecting the STR20 drying process in terms of EMC. The EMC isotherms of crumb rubber were calculated by SSM among surrounding temperature of 40-60°C provided to RH surrounding of 10-90%. The SSS were LiCl; ($MgCl_2 \cdot 6H_2O$); ($Mg(NO_3)_2 \cdot 6H_2O$); (KNO_3) and (NaCl). This report also depicted that modified Halsey model was suitable in used for developing the crumb rubber producing STR20. The report of Tirawanichakul *et al.,* [153] studied and developed the mathematical model; namely Freundlich, B.E.T., Oswin, Smith, Chung and Pfost, Iglesias and Chirife, Modified Henderson, Modified Halsey, Modified Oswin and Modified GAB; of important parameters which affecting the STR20 drying process in terms of EMC. The report presented when decreasing of temperature at constant relative humidity (RH), the EMC value increased. The EMC curves of STR20 presented that the EMC value was dependent on temperature. This report also depicted that the model, namely Modified Oswin was the most appropriate model for explaining the EMC isotherms. For rubber sheets, the report of Tirawanichakul *et al.,* [126] used Four mathematical models, namely Chung and Pfost, Halsey, Henderson and Kaleemullah; for prediction of the experimental values. The result presented that the model, namely Halsey was the most appropriate model for explaining the EMC isotherms.

In 2011, Tasara *et al.,* [154] presented that the EMC of rubber sheets using SSM at temperature of 40, 50, 55, and 60°C, all of the SSS provided to RH surrounding of 10-90%. The six mathematical models were Halsey, Oswin, GAB, Modified Oswin, Henderson and Smith which using to fit with the data of experiment. The result showed that the model, namely Modified Oswin was the most appropriate model for explaining the EMC isotherms. The report of Dejchanchaiwong *et al.,* [155] used four mathematical models, namely Modified Halsey, Modified Oswin, Modified Henderson and Modified Smith; for prediction of the experimental values. The EMC isotherms of rubber sheets were calculated by SSM among surrounding temperature of 40-60°C provided to RH surrounding of 10-90%. The SSS were LiCl; $MgCl_2 \cdot 6H_2O$; $Mg(NO_3)_2 \cdot 6H_2O$; KNO_3 and NaCl. The result indicated that the model, namely Modified Halsey was the most appropriate model for explaining the EMC isotherms. Jeentada *et al.,* [141] studied and developed the mathematical model of important parameters which affecting the rubber sheet drying process in terms of EMC. The EMC isotherms of rubber sheets were calculated by SSM among surrounding temperature of 40-70°C. The SSS were LiCl; $MgCl_2 \cdot 6H_2O$; $Mg(NO_3)_2 \cdot 6H_2O$; $(NH_4)_2SO_4$; NaCl and Potassium-Iodide (KI). Twelve mathematical models, namely Oswin, Modified Oswin, Halsey, Modified Halsey, Henderson, Modified Henderson, Peleg, Chung and Pfost, Modified Chung-Pfost, GAB, Modified GAB and Henderson–Thompson, were used for prediction of the experimental values. This report showed that the model, namely Peleg was the most appropriate model for explaining the EMC isotherms.

The researchers accept that the drying phenomenon of products during the falling rate time is controlled by the mechanism of vapor and liquid diffusion [39, 134]. A mathematical modeling of thin-layer drying models describe the drying phenomenon of these materials mainly fall into three categories namely, empirical, semi-theoretical and theoretical. Empirical and semi-theoretical drying equations consider only external resistance to moisture transfer resistance between air and product while theoretical drying equation takes into account only internal resistance to moisture transfer [150, 156].

Table 1
Equations describing the sorption equilibrium isotherms

Name	Equation*	Reference
BET	$EMC = \frac{ABa_w}{\left[(1-a_w)+(B-1)(1-a_w)a_w\right]}$	[157]
Chen	$EMC = -\frac{1}{A}\ln\left[\frac{1}{B}\ln(a_w - C)\right]$	[158]
Chung and Pfost	$EMC = -\frac{1}{A}\ln\left[\frac{(T+B)\ln(a_w)}{-C}\right]$	[148]
Modified Chung and Pfost	$EMC = \frac{\ln(A)}{C} - \frac{1}{C}\ln\left[-(T+B)\ln(a_w)\right]$	[148]
GAB	$EMC = \frac{ABCa_w}{\left[(1-Ca_w)+(1-Ca_w+BCa_w)\right]}$	[159]
Halsey	$EMC = \left[\frac{-A}{T\ln(a_w)}\right]^{\frac{1}{B}}$	[149]
Modified Halsey	$EMC = \left[\frac{-\exp(A+BT)}{\ln(a_w)}\right]^{\frac{1}{C}}$	[160]
Henderson	$EMC = \left[\frac{\ln(1-a_w)}{-AT}\right]^{\frac{1}{B}}$	[152]
Modified Henderson	$EMC = \left[\frac{\ln(1-a_w)}{-A(T+B)}\right]^{\frac{1}{C}}$	[152]
Iglesias-Chirife	$\ln\left[EMC + \left(EMC^2 + \frac{ECM}{2}\right)^{\frac{1}{2}}\right] = Aa_w + B$	[160]
Oswin	$EMC = A\left[\frac{a_w}{1-a_w}\right]^B$	[161]
Modified Oswin	$EMC = (A+BT)\left[\frac{a_w}{1-a_w}\right]^{\frac{1}{C}}$	[162]
Peleg	$EMC = A(a_w)^B + C(a_w)^D$	[163]
Smith	$EMC = A - B\ln(1-a_w)$	[164]

* A, B, C and D are the equation constants.

Many Researchers attempt to describe the drying phenomenon based on empirical drying concepts. Thus, some of the widely used mathematical models for empirical drying equation are presented in Table 2. In this section, the study of the mathematical models for NR drying is reviewed. Many reports of researchers are discussed as follows.

For crumb rubber producing STR20, the report [44] presented that the mathematical model; namely Henderson and Pabis was the most appropriate model for explaining the rubber sheet drying. Moreover, the mathematical model, namely Logarithmic was the best fitted model for thin-layer crumb rubber producing STR20 drying by hot air temperature of 110-130°C. The study of Xiang Ng *et al.,* [165] showed that the mathematical model, namely modified Henderson and Pabis was the best fitted model for thin-layer crumb rubber producing STR20 drying. Their result was different to the result from Yutthana Tirawanichakul and Supawan Tirawanichakul [44] when this crumb rubber in their experiment was dried by vacuum oven at temperature of 80-100°C.

Table 2
Equations describing the thin-layer models

Name	Equation*	Reference
Herderson and Pabis	$MR = a\exp(-kt)$	[151]
Modified Herderson and Pabis	$MR = a\exp(-kt)+b\exp(-gt)+c\exp(-ht)$	[166]
Logarithmic	$MR = a\exp(-kt)+b$	[167]
Newton	$MR = \exp(-kt)$	[168]
Page	$MR = \exp(-kt^n)$	[169]
Sharma	$MR = a\exp(-k_1 t)+b\exp(-k_2 t)$	[44]
Two-term exponential	$MR = a\exp(-k_1 t)+b\exp(-k_2 at)$	[170]
Verma	$MR = a\exp(-kt)+b\exp(-gt)$	[44]
Wang and Singh	$MR = 1+at+bt^2$	[171]

* MR is moisture ratio. a, b, c, k, k_1, k_2, g, h and n are the equation constants.

For rubber sheet, Yutthana Tirawanichakul and Supawan Tirawanichakul [44] studied the drying kinetics of rubber sheets. The report showed that the mathematical model; namely Page was the most appropriate model for explaining the rubber sheet drying. The report of Dejchanchaiwong *et al.,* [155] used twelve drying kinetics of mathematical models, namely Page, Overhults, Approximation of diffusion, Two term, Weibull Distribution, Midilli *et al.,* Verma *et al.,* Hii *et al.,* Two terms exponential, Modified Henderson and Pabis, Modified Page and Modified Overhults; for prediction of the experimental values. The report showed that the mathematical models; namely Weibull Distribution and Modified Henderson and Pabis were the most appropriate models for explaining the rubber sheet drying by hot air and green-house gas drying at temperature of 45-55°C, respectively. Ekphon *et al.,* [172] studied the drying kinetics of rubber sheets using hot air and conventional natural drying. An initial moisture content of the experimental sheet ranging of 23-40% dry-basis was dried until the final moisture content reached to 0.15% dry-basis by air flow rate of 0.7 m/s and temperature of 40-70°C. Ten mathematical models, namely Page, Overhults, Logarithmic, Weibull Distribution, Approximation of diffusion, Hii *et al.,* Verma *et al.,* Modified Page, Modified Henderson and Pabis and Modified Overhults were used for prediction of the experimental values. The results presented that the mathematical models; namely Verma *et al.,* and Approximation of diffusion were the best fitting models for describing the drying kinetics of rubber sheets with the hot air and conventional natural drying, respectively. In 2013, Jeentada *et al.,* [173] studied and developed the mathematical model of important parameters which affecting the drying kinetics of the rubber sheet. Ten mathematical models, namely Newton, Page, Henderson and Pabis, Logarithmic, Weibull Distribution, Midilli *et al.,* Verma *et al.,* Two terms exponential, Modified Henderson and Pabis and Wang and Singh were used for prediction of the experimental values. This report showed that the model, namely Modified Henderson and Pabis, Midilli *et al.,* Verma *et al.,* Weibull Distribution and Page were the suitable model for explaining the drying kinetics. Studied by Pianroj *et al.,* [174], a hybrid solar-electrical dryer was built for studying the drying kinetics of rubber sheets. The report presented that the mathematical model; namely modified Henderson and Pabis was the best fitted model for thin-layer rubber sheet drying at temperature of 40-60°C. The report of Dejchanchaiwong *et al.,* [175], the apparatus in their research was indirect solar and mixed-mode dryers. Ten mathematical models, namely Page, Henderson and Pabis, Modified Page, Modified Henderson and Pabis, Weibull Distribution, Logarithmic, Diffusion approach, Verma *et al.,* Two terms and Hii *et al.,* were used for prediction of the drying kinetics of the rubber sheet. The report showed that the mathematical model, namely Hii *et al.,* was suitable model for explaining the drying kinetics of rubber sheet.

For effective diffusion coefficient, D_{eff}, determination, the assumption that the resistance to moisture flow is uniformly distributed throughout the interior of the homogeneous isotropic material. D_{eff} is independent of the local moisture content and if the volume shrinkage is negligible. From Fick's second law, the equation can be determined by Eq. (22).

$$\frac{\partial M}{\partial t} = D_{eff}\nabla^2 M \tag{22}$$

where M is the moisture content and t is the adsorption time.

Crank [176] studied the analytical solutions of this equation for various regularly shaped bodies such as an infinite slab, a cylinder and a sphere. The partial differential equation of moisture diffusion for the drying product considered the geometric of the material that was diffused inside the body and they were calculated by Eq. (23) to Eq. (25).

For infinite slab shape:

$$\frac{M_t - M_{eq}}{M_i - M_{eq}} = 1 - \frac{8}{\pi^2}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^2}\times\exp\left[-\frac{\pi^2(2n+1)^2}{4L^2}D_{eff}t\right] \tag{23}$$

where M_{eq} is moisture content when it is in equilibrium at the given relative humidity level, M_i is the initial moisture content, M_t is the moisture content at time, n is the number of terms of the series, and L is the slab thickness.

For finite cylindrical shape:

$$\frac{M_t - M_{eq}}{M_i - M_{eq}} = 1 - \sum_{n=0}^{\infty}\frac{4}{(r^2\lambda_n^2)}\times\exp\left[-\lambda_n^2 D_{eff}t\right] \tag{24}$$

where r is the radius and λ_n is the positive roots of the first Bessel function of order zero.

For spherical shape:

$$\frac{M_t - M_{eq}}{M_i - M_{eq}} = 1 - \frac{6}{\pi^2}\sum_{n=0}^{\infty}\frac{1}{n^2}\times\exp\left[-\frac{\pi^2 n^2}{r^2}D_{eff}t\right] \tag{25}$$

D_{eff} is conventionally described by the Arrhenius-type equation as Eq. (26).

$$D_{eff} = D_0\times\exp\left[\frac{E_a}{RT_{abs}}\right] \tag{26}$$

where D_0 is the Arrhenius-factor, R is the ratio of dry sample mass to dry air mass and T_{abs} is the absolute temperature.

In 2007, the study of Tirawanichakul *et al.,* [126] showed that the diffusion models of crumb rubber producing STR20 drying could be determined by non-linear regression analysis. When their resulting data were substituted into the analytical solution of Crank [176] and Luikov [177], D_{eff} of this crumb rubber could be defined as Eq. (27) to Eq. (29).

For infinite slab shape:

$$D_{eff} = 69.365 \times \exp\left[\frac{-2246.601}{T_{abs}}\right] \qquad r^2 = 0.8530 \tag{27}$$

For finite cylindrical shape:

$$D_{eff} = 2.443 \times 10^{-3} \times \exp\left[\frac{4.973}{T_{abs}}\right] \qquad r^2 = 0.7176 \tag{28}$$

For spherical shape:

$$D_{eff} = 1.0 \times 10^{-6} \times \exp\left[\frac{2995.5}{T_{abs}}\right] \qquad r^2 = 0.7348 \tag{29}$$

From the results of a correlation coefficient, r^2, they suggested that the estimations of the effective diffusion coefficient obtained for infinite slab shape had a good relation to the experimental value. This result agreed with Yutthana Tirawanichakul and Supawan Tirawanichakul [44] report.

For other initial moisture content and rubber product study, Tirawanichakul *et al.,* [147] studied the diffusion models of natural rubber drying namely, crumb rubber producing STR20 and rubber sheet. In this experiment, they assumed these samples had an infinite slab shape. D_{eff} equation of these samples that had the initial moisture content ranging of 30-65% dry-basis could be written as Eq. (30) to Eq. (31).

For crumb rubber producing STR20:

$$D_{eff} = 41.3857 \times \exp\left[\frac{-6158.473}{T_{abs}}\right] \qquad r^2 = 0.971 \tag{30}$$

For rubber sheet:

$$D_{eff} = 4.0 \times 10^{-5} \times \exp\left[\frac{-1192.368}{T_{abs}}\right] \qquad r^2 = 0.923 \tag{31}$$

These experiments suggested that the initial moisture content had effect on D_{eff}.

7. The Energy Consumption

There are many ways to reduce the energy consumption in NR production. In the latex production such as SL and CL, 90% of the electricity is used in the centrifugation process (Thai Department of

Industrial Work (TDIW), 2001). A report by TDIW showed that an installation of inverters to centrifugal machines could reduce the energy consumption. Addition, this report presented that changing the clutch and gear systems machines (old machines) with the variable pulleys machines (new machines) could reduce 20% of electricity consumption.

For RSS production, the position and size of gas supply ducts and ventilating lids are often not optimal in the smoking chambers [14, 32, 34, 178]. This problem has significantly affected to the hot gas circulation in the rubber smoking chambers. The resulting large temperature variations and non-uniform flow in the chambers lead to increase the energy consumption [14, 178]. The energy requirement can decrease by moisture control of the Para-rubber fuel wood and installation of insulators in drying chambers. Installation of insulators at the chamber can decrease the fuel consumption (the para-rubber wood) about 770 kg per times [14]. Designing the drying chambers with appropriate hot gas circulation and temperature distribution can decreased the energy consumption in the rubber sheets drying process [14, 33]. In this section, the study to reduce the energy consumption in RSS production is reviewed. Many reports of researchers are discussed as follows.

Kalasee [14] is the first report to improve on energy consumption in RSS production. The report showed that improvement the RSS chamber by Computational Fluid Dynamics Technique (CFD) could be reduced the energy consumption in RSS production. The uniform flow and temperature in the RSS chamber led to decrease the energy consumption about 30%. Investigated by Promtong [32], CFD technique was used to improve temperature and air velocity flowing in the RSS chamber. The results showed that the uniform flow and temperature could decrease the energy consumption. It was similar to the results from Kalasee [14]. The studies of Promtong and Tekasakul [178] presented that non-uniform flow and temperature in RSS chamber had affected to the addition of energy consumption. Improvement the RSS chamber by using CFD Tekasakul and Promtong [178] could help the RSS cooperatives (low-cost rubber factory in Thailand) to achieve at least a 31.25% saving in energy. The report of Tasara *et al.,* [154] presented that a new design chamber, namely two-stage drying strategy could be decreased the energy consumption by the exhaust air recycling. For their chamber, the exhaust air recycling in RSS process was 80-95%. Dejchanchaiwong *et al.,* [179] presented that CFD technique modelling of single-phase and multi-phase flow could be used to simulate the relative humidity and temperature in the RSS chamber. Dejchanchaiwong *et al.,* [180] designed and constructed a new RSS chamber according to the modified RSS room by using CFD. The results showed that the better air flow and temperature distribution could be reduced the drying time from 72 hours in an older RSS room to 48 hours in this new design chamber. In 2019, Dejchanchaiwong *et al.,* [180] presented that the modified RSS chamber was able to save the fuelwood; Para-rubber wood; consumption. This chamber was saving the wood consumption 67% by comparison with a conventional RSS room (the 1994 model). The report suggested that this chamber was suitable for the rubber cooperative, represented a good SEC and had the financial return. The report of Promtong [181] presented that a new design chamber, namely the new two-stage forced convection system could save 35% of fuel consumption and 25% of RSS production time compared with the original an older RSS chamber.

For STR 20 production, the report by Office of Industrial Economics [182] presented that the regular maintenance of shredder machines and cutters and using the high efficiency motors could decrease electricity energy consumption about one-third. The energy requirement could reduce by controlling of the temperature drying and the moisture of crumb rubber [44, 182]. The report of Khongchana *et al.,* [45] suggested that drying the crumb rubber for 40 minutes at 130^{o}C and followed by 180 minutes drying at 110^{o}C has a lower specific energy consumption and better quality as compared to current practice in many factories.

Recommendation of this review, CFD technique is very interesting method to improve the rubber processing room (RPR). Temperature and air flow distribution inside Industrial scale solar dryer using CFD simulation was investigated by Noh *et al.,* [183]. Eakvanich *et al.,* [184] used the CFD technique to improve the performance and the temperature distribution of a commercial domestic refrigerator with the latent heat storage materials. Furthermore, the experimental and CFD simulation results shown that the electricity consumption and the human thermal comfort in the radiant cooling room with the solid desiccant dehumidifier were better [185]. The CFD technique can be used to decrease the energy consumption and production time. The study of the relative humidity, air flow and temperature distribution on modification of the RPR by CFD technique field is very interesting in the future.

8. Conclusions

This review focuses on literature findings the major constituents of rubber latex, rubber latex particles, particle interaction of rubber latex, the rubber porous structure, the rubber drying kinetics and the energy consumption of RSS and STR20 block rubber. The following are the conclusions and recommendations from the review:

I. Latex that flows out of the Para-rubber tree is a complex form of cytoplasm. It is containing a suspension of rubber and non-rubber particles in an aqueous serum [3, 5-6, 55-56]. Using ultra-centrifugation method, latex can be mainly divided into four fractions an upper white layer consists of rubber particles, Frey-Wyssling particles in a yellow or an orange layer, C-serum phase and lutiods particles in the bottom fraction [1-3].

II. The major component of Para-rubber latex is rubber hydrocarbon particles. The particles are usually spherical [6]. The averages of these particles have diameters of 0.02 to 3.0 micron, and they are protected by a complex film containing lipids and proteins [2, 5, 8]. Para-rubber latex has variable composition depending on several factors, for example, age of the tree [65], season [14, 66-67], soil conditions, tapping and frequency [68].

III. Fresh NR latex from the Para-rubber tree is a thixotropic neutral liquid of density around 0.98 g/cm^3. It has a pH of 6.5 to 7.0 and molecular weight distribution 10^5 to 10^7 g/mol [75-76]. Besides rubber polymers $(C_5H_8)_n$, NR latex contains carbohydrates, fatty acids, glycolipids, phospholipids, proteins, lutoids, inorganic salts and other components [6, 77-79]. Latex can be maintained by the main forces of attraction between neighboring rubber particles that are encased with lipids and proteins [80]. The main forces of attraction between neighboring rubber particles in latex system can be divided into five force types; SF, VWI, EF, EI and PPI [9].

IV. The porous model technique is utilized to describe moisture transfer [98-101]. This technique is based on the existence of two different regions (non-hygroscopic and hygroscopic).

V. The drying of agricultural products such as rubber, vegetables and fruits, always produces a considerable shrinkage effect. This phenomenon considers in the physical of the product, such as the diffusion coefficient, mass and heat transfer. In this review was able to conclude that the different in an initial moisture content of raw material, the experimental temperature and the drying equipment had affect to the EMC isotherms and the drying kinetics.

VI. There are several ways to reduce the energy consumption in rubber production. For RSS production, the resulting large temperature variations and non-uniform flow in the chambers lead to increase the energy consumption [14, 178]. The energy requirement could decrease by moisture control of the Para-rubber fuel wood and installation of insulators in drying chambers [14, 178]. Installation of insulators at the chamber could decrease the fuel consumption (the para-rubber wood) about 770 kg per times [14]. Designing the drying chambers with appropriate hot gas circulation and temperature distribution could decreased the energy consumption in the rubber sheets drying process [14, 33]. For STR20 production, the report by Office of Industrial Economics [182] presented that the regular maintenance of shredder machines and cutters and using the high efficiency motors could decrease electricity energy consumption about one-third. The energy requirement could reduce by controlling of the temperature drying and the moisture of crumb rubber [182, 145].

This review article would be helpful to the researchers, scientists and students who are working and studying on a field in forms, structure, physiology and biochemistry of NR and the drying kinetics and energy consumption of NR drying product.

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