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Method for producing natural rubber powder from rubber latex by spray drying process

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Abstract

This paper presents the method for producing natural rubber powder from natural rubber latex by a spray drying process under hot gas jet generated from a pulse combustor. The pulse combustor is valveless Helmholtz type, having single tailpipe and single air inlet. In the experiments, the flow rate of LPG fuel was fixed at 29 l/min. The rubber latex was sprayed from an air spray nozzle into the drying chamber. The latex droplets were heated by hot gas jet and dried in a chamber. The temperature of the hot gas jet in the chamber inlet was about 180 ^{o}C and the chamber outlet temperature was about 80 ^{o}C. The latex dispersion was spray-dried to obtain rubber particles. After the drying process, the morphology and particle size of natural rubber powders were analyzed by scanning electronic microscopy (SEM) and electron micro analyzer, respectively. The range of particle size was from approximately 50 µm to 300 µm.

Keywords: *natural rubber powder, nozzle configuration, particle size, pulse combustor, spray drying*

1. Introduction

Natural rubber (NR) is an important agricultural product of Thailand. In 2016, Thailand produced more than 4 million tons of natural rubber (Markets Insider, 2017). According to the International Rubber Study Group (IRSG) (2015), the sharing of natural rubber from Thailand accounted for about one-third of the world production. Most of it was exported in several forms such as rubber sheets, rubber block and concentrated latex.

At the present time, natural rubber latex is being produced manually from rubber trees, followed by filtering, then coagulating and drying the latex, then shipping the natural rubber in the form of a sheet or block from the countries of origin. In this way, even at the present, where the need of natural rubber is rather continuing to rise, rubber sheet and block are produced from rubber latex relying on manual labor. Further, the latex coagulating, rinsing and drying steps in particular the drying step, have a large influence on the viscosity of the rubber product. The current method for production of rubber from the latex cannot be said to be sufficient in view of the variation in rubber quality. Natural rubber is mainly used for tires, gloves, condoms, balloons and other relatively high-value products (Praktikantin, 2017).

Natural rubber powder (NRP) has advantages in further production. It is better to mix with some chemical solutions or powders prior to vulcanization. So it also will be possible to make good compounds with different plastics and fillers. Beside this, it will save a lot of energy because will not grind the big chunks of rubber sheets or blocks.

Natural rubber powder will be widely used as an additive in polymer industries because of its mechanical properties with high tensile modulus. Natural rubber powder can be successfully prepared by a spray drying method. Spray drying processes, the generation of small and uniform droplets and the efficient precipitation of submicron particles is challenging (Sosnik & Seremeta, 2015). For the production of droplets in micrometer size there are many methods, such as ultrasonic atomization (Arpagaus, Collenberg, Rutti, Assadpour, & Jafari, 2018), de Laval type atomization in the supersonic spray dryer (Eggersdorfer, Koren, Stolovicki, Amstad, & Weitz, 2017) and liquid atomization by electrospraying (Jaworek & Sobczyk, 2008) were developed. The advantages of a spray drying method will be to control the particle size, reproducible and scalable (Arpagaus & Schwartzbach, 2008). The produced powders are high in quality and have typically low moisture contents (Schmid, 2011), resulting in high shelf life stability (Anandharamakrishnan & Ishwarya, 2015). However, it is difficult to prepare rubber nanoparticles using conventional methods due to inherit properties of rubber that were adhered together (Li et al., 2007). In the recent literature, there are several works that demonstrate the possibility to obtain ultrafine rubber powder, particles from micrometer to nanometer dimensions, based on rubber lattices, such as styrene-butadiene, acrylonitrile-butadiene and natural rubber. Some examples of works will be presented in the next paragraphs.

According to the literatures, they have reported the preparation of natural rubber powder by spray drying of Zinc oxide pre-vulcanized latex (Sae-Oui, Sirisinha, Sa-nguanthammarong, & Thaptong, 2010). However, instead of irradiation, they used a chemical reaction with sulfur and other additives, at 70 °C for 24 h. After this period, the latex was sprayed and dried to obtain the particles. It was also described to prepare natural rubber powder pre-vulcanized with Maltodextrin for reducing particle size. It was found that the starch of maltodextrin was used to encapsulate the latex particle which the free flowing and non-stick of rubber powder could be prepared. The inlet air temperature of 120 ºC was found to be suitable to make the rubber powder. The mass ratio of rubber to maltodextrin should not be higher than 9:1 to obtain the free flowing rubber powder (Sopanon & Soottitanawat, 2011). It has been reported that research into the preparation of rubber powder from styrene-butadiene latex modified with a colloidal oxide. The SBR rubber latex can be prepared from the ratio of latex mixed with methyl methacrylate and silica colloid, spray drying, solid rubber latex. 40% by weight with a flow rate of 250 ml/h. The air inlet temperature of 120 °C and air outlet temperature of 75-80 °C in a Mini B-190 Buchi Labortechnik AG dryer. The particle size of rubber powder analyzed by SEM is from approximately 1 μm to 10 μm. Differential scanning calorimeter analysis indicated that the glass transition temperatures did not change significantly, and thermogravimetric analysis showed thermal stability until approximately 200 °C (Paiva, Oliveira, & Gavioli, 2014). In addition, natural rubber powder was used as an additive in PLA (polylactid) because of its mechanical properties and crystallisation ability of PLA. The ductility of PLA has been significantly improved by blending with natural rubber, elongation break, the incorporation of natural rubber not only increased the crystallisation rate but also enhanced the crystallisation ability of PLA (Bitinis, Verdejoa, Cassagnau, & Lopez-Manchado, 2011).

Another interesting work about a method for producing natural rubber latex or synthetic rubber latex synthesized from emulsion polymerization is dried to produce rubber using a pulse combustor generating a shock wave. In the application, such a pulse combustor is used to spray and dry latex, having a solid concentration of 60% by weight or less in a drying chamber, under conditions of a frequency of 250 to 1200 Hz, more preferably 300 to 1000 Hz, and a temperature of not more than 140°C, more preferably 40 to 100°C (Daisuke, Tetsuji, & Yousuke, 2007).

2. Objectives

The aim of this present study relates to a method for producing natural rubber powder from a rubber latex by a spray drying process inside a drying chamber under hot gas jet generated from pulse combustor. Furthermore, the morphology and particle size of natural rubber powder were investigated.

3. Materials and methods

3.1 Experimental setup

The experimental setup with the instrumentation is shown in Figure 1. The schematic diagram of the experimental setup consists of a Helmholtz-type pulse combustor, ignition controller, a drying chamber, blower, digital data logger, thermocouple (Type K), rotameter, a high compressor tank, peristaltic pump, rubber latex tank, spray nozzle and feed talcum powder. The K-type thermocouple uncertainty is ± 0.5 ºC. The uncertainty of air flow meter is about 3.1%.

A drying chamber has a volume of about 1.95 m³ (0.45 m radius and 3 m high). Inside the drying chamber thermocouple type K for measuring temperature variation was installed in the center of the chamber. The distance between measuring positions (T_1-T_{15}) is 200 mm as shown in Figure 1.

For details of the spray drying nozzle, the fluid was contained within a flexible tube (silicone rubber tube) fitted inside a circular pump casing. A rotor with a number of rollers attached to the external circumference of the rotor compresses the flexible tube. As the rotor turns, some part of the tube under compression is pinched thus forcing the fluid to be pumped through the tube. As the tube opens to its starting point the fluid is drawn to the pump.

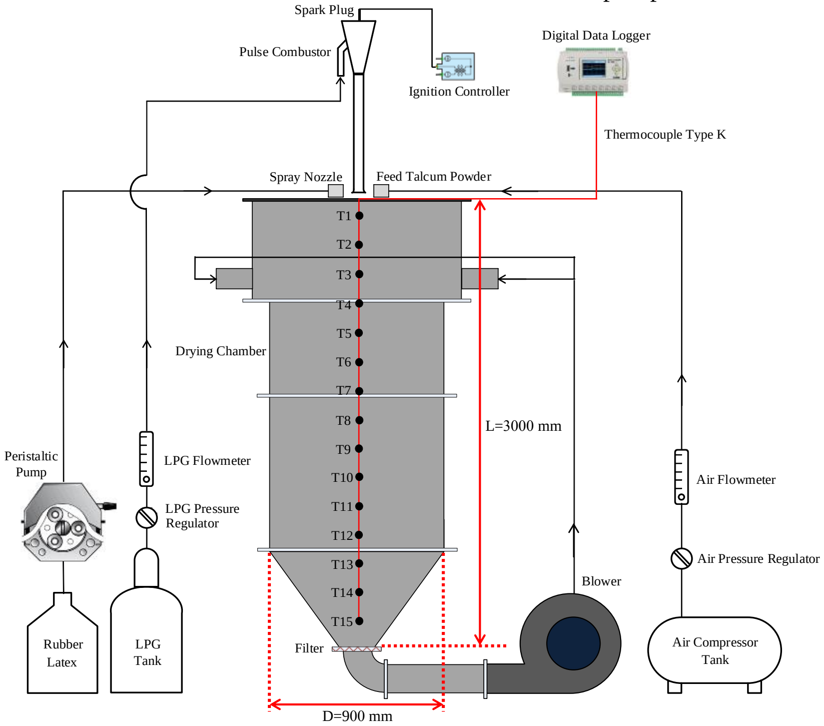


Figure 1 Experimental setup for producing natural rubber powder from a rubber latex by spray drying process

Figure 2 The photograph of drying chamber

3.2 Model and parameters of pulse combustor

The pulse combustor used in this study is a Helmholtz-type without mechanical valves. A schematic of this combustor is shown in Figure 3. The pulse combustor consists of a combustion chamber, a single tailpipe and a single air inlet pipe. The dimension of the combustion chamber is 110 mm in diameter and 300 mm in length. The pulse combustor is made of stainless steel. A spark plug was installed at the bottom of combustion chamber for ignition during starting. The dimension of the air inlet pipe is 29 mm in diameter and 165 mm in length. The dimension of the tailpipe is D=47 mm in diameter. The length of tailpipe is 752 mm (16D).

3.3 Nozzle configurations

Figure 4 shows the configuration and dimensions of the spray nozzle which used in this study. The spray nozzle consists of the air inlet and rubber latex inlet tubes. The air inlet tube has 2 mm in diameter and the rubber latex inlet has 4 mm in diameter. The spray angle of the nozzle orifice was 40 degrees. The flow rate of the latex is adjusted by the peristaltic pump through silicone rubber tube. The air from the compressor tank was introduced through the air inlet of spray nozzle. The flow rate was controlled by a rotameter. Simultaneously, the air is flowing through the nozzle and then impinges through the rubber latex and breaking latex into small particles. Figure 5 shows the models of the spray nozzles at different air inlet positions used in this present study.

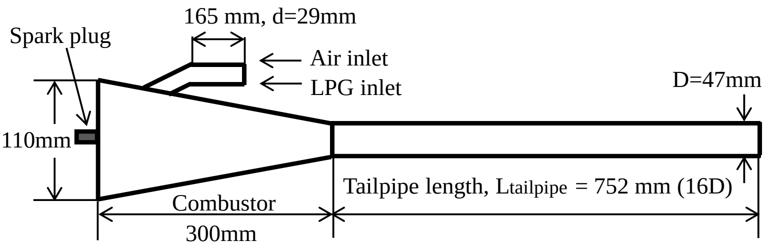


Figure 3 Schematic diagram of a Helmholtz-type pulse combustor

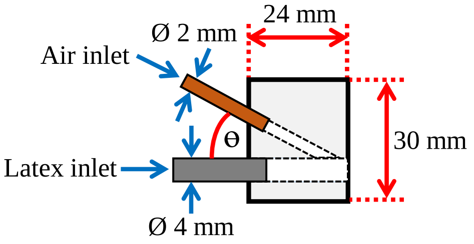


Figure 4 Model and parameters of spray nozzle

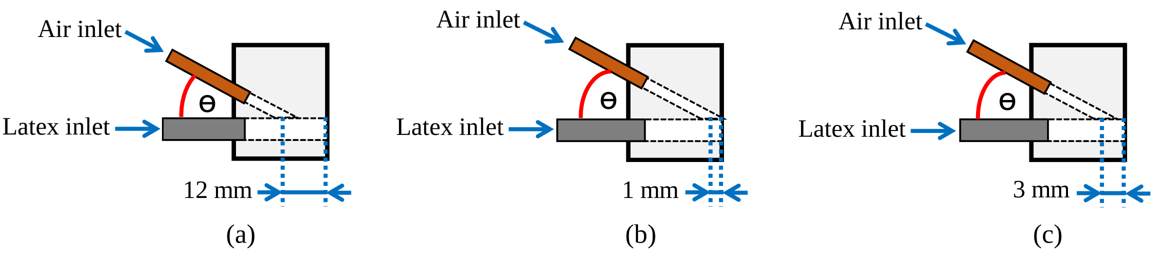


Figure 5 The model of spray nozzles at different air inlet positions: (a) position 1, (b) position 2 and (c) position 3

3.4 Materials and procedure

High ammonia natural rubber latex (HA-60% by weight) was purchased from Chalong latex industry co., Ltd. (Songkhla, Thailand). The formulation of natural rubber latex was shown in Table 1. The talcum powder was obtained from the Department of Materials Science and Technology, Prince of Songkla University (Hatyai, Thailand).

In the experiments, the supplied flow rate of LPG fuel was fixed at 29 l/min. The drying process starts with the LPG rotameter and air rotameter used to measure the flow rates of fuel and air, respectively. The LPG fuel and compressed air were injected into the center of the air inlet pipe for mixing inside the combustion chamber. A spark plug was installed at the bottom of the combustion chamber for starting the system. Both spark plugs and compressed air were used only in the beginning for starting the process.

The air was blown with a blower through a stainless steel pipe into the air inlet on the top of the drying chamber. The inverter controls the speed of the blower. The temperature variation inside the drying chamber was measured by using a thermocouple type K connected to digital data logger storage.

Conditions of the fixed nozzle air flow rate at 90 l/min, the inlet air temperature of 180 ^{o}C and relative humidity in the drying chamber of 38-55% by weight, fixed feed talcum powder flow rate at 60 ml/min and the mass flow rate of rubber latex fed into the spray nozzle varied in range of (Q_L) 20-60 ml/min were studied. After feeding latex into the spray nozzle, rubber particles dispersed around inside the drying chamber and dropped to the bottom of the chamber. At the near outlet drying chamber a filter was installed for the collection of rubber particles.

3.5 Micrograph and size analysis of rubber powder particles

The morphology of rubber particles prepared by spray drying depends on the drying conditions and the feed properties. Dense, hollow, porous, and encapsulated structures with spherical, wrinkled, shriveled outcomes are possible (Arpagaus, John, Collenberg, & Rutti, 2017). The morphology of the samples was observed using a scanning electron microscopy (SEM), Quanta 400, FEI, (Czech Republic) at 20 kV. The particle size of rubber was determined by an electron micro analyzer.

3.6 Mechanical properties testing

The mechanical testing is one of the major elements in describing the properties of natural rubber production for application. This research investigates the mechanical properties of two different natural rubber. The tensile testing of the samples was measured according to the ASTM D412 (Die C) standard. It was used for measuring the hardness of natural rubber by piercing the needle into the testing samples by Shore A according to ASTM D2240 standard.

Table 1 Formulation of the HA 60 % by weight of natural rubber latex

Properties	Test Results	H.A. Limits
Total solids content	61.73 %	≥ 61.50
Dry rubber content	60.14 %	≥ 60.00
Ammonia content (on total weight)	0.70 %	≥ 0.60
Ammonia content (on water phase)	1.829 %	≥ 1.60
Non rubber solid	1.59 %	≥ 2.00
pH	10.56	≥ 10.50
KOH number	0.56	≤ 1.00
Volatile fatty acid number	0.029	≤ 0.20
Mechanical stability time @55% T.S. (ASTM)	1,100 Secs	≥ 650
Mg^{2+}	17.00	≤ 30.00 PPM
Colour of latex	White	White
Colour of film	Normal	Normal
Odour of latex	Sweet	Sweet

4. Results and discussions

4.1 Temperature variation inside drying chamber

Figure 6 shows the temperature variation inside the drying chamber at the different measuring positions measured with thermocouples probe type K. It was found that the position near the tailpipe outlet at T_1 gives the highest temperature at 180 °C and the measuring position near the drying chamber outlet at T_{15} gives the lowest temperature at 80.5 °C. The temperature distribution of hot air decreased linearly in the drying chamber.

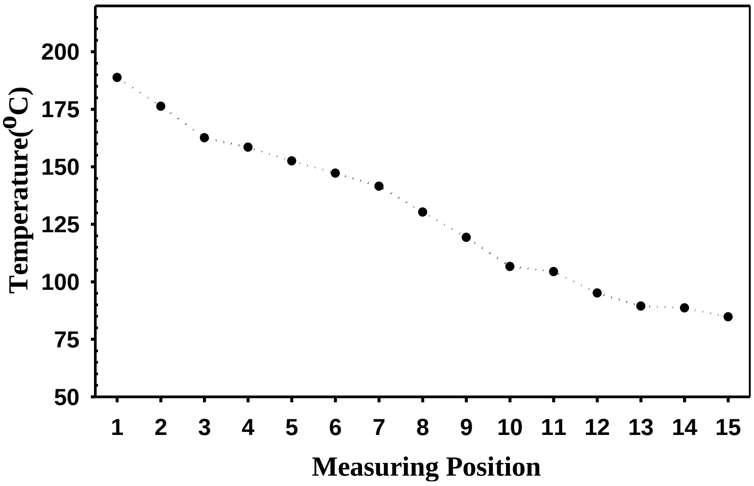


Figure 6 Temperature variation inside the drying chamber according to the measuring position

4.2 Effect of spray nozzle

Figure 7 shows the photographs of latex spraying profiles for different nozzle configurations of different air inlet positions. The liquid and air flow rate fed into the nozzle were set at 20 ml/min and 90 l/min, respectively.

Figure 7(a) shows the sprayed latex near the outlet of nozzle. It obviously generated large droplets and cannot be controlled. In the Figure 7(b) it can be seen that the latex droplets from the spray nozzle gets more aerosol characteristics compared to the air inlet position 1. It was found that the small droplets make a fog in the middle of the beam. But the diameter of the latex droplets is relatively large. So the present study decided to use the nozzle model c (see Figure 5(c)) because all the beams consist of small droplets.

As the results are shown in the Figure 7(b) and (c), smaller droplets of latex spray was obtained because the air inlet position was placed near the outlet of the nozzle. These results indicated from the air inlet position 3, Figure 7(c) the sprayed latex was smoother than air inlet position 2, Figure 7(b) and could be controlled droplet sizes. The optimum position for the air inlet position of the spray nozzle configuration was position 3, as seen in Figure 7(c). This was because the latex spray does not hit the surface of the drying chamber at this position.

4.3 Morphology of natural rubber powder

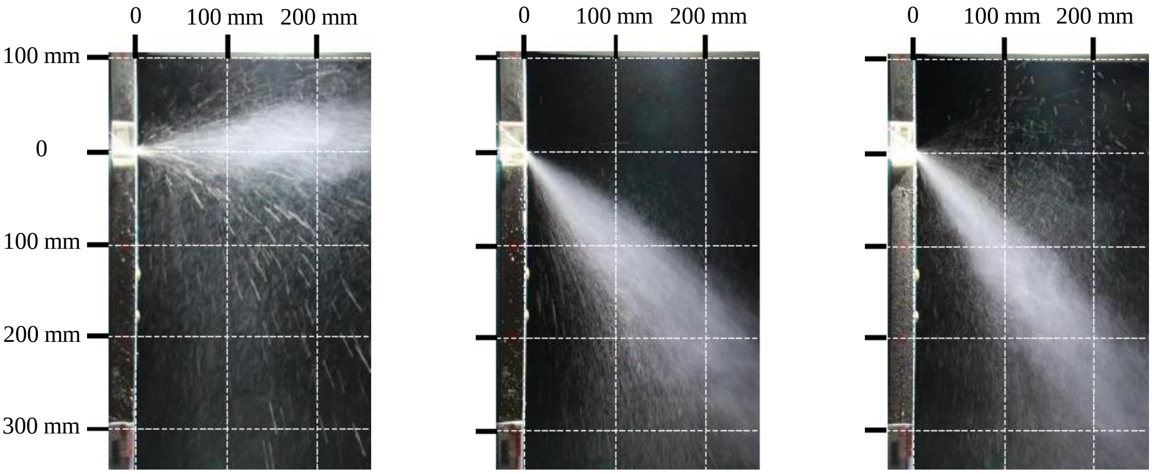
The morphology obtained from the SEM was shown in Figure 8 which a particle size of about 50-300 μm. The comparison for SEM micrographs under ×500 of magnification of Figure 8(a) the natural rubber particles which were produced by spray drying without talcum powder and Figure 8(b) the natural rubber particles coated with talcum powder. These results showed that the particles without talcum powder stuck together. Meanwhile, characteristic of the natural rubber particles coated with talcum powder were spherical. It is found that the talcum powder could be used for coating or encapsulating the surface of natural rubber particles.

The morphology of natural rubber particles was characterized by scanning electronic microscopy (SEM) and the photomicrographs. The samples are shown in Figure 9(a-h). Figure 9 shows the SEM micrographs for the natural rubber particle at different magnifications.

It was clearly shown in Figure 9(a) and (b) that from the SEM for spray dried particles without talcum powder they were sticking together.

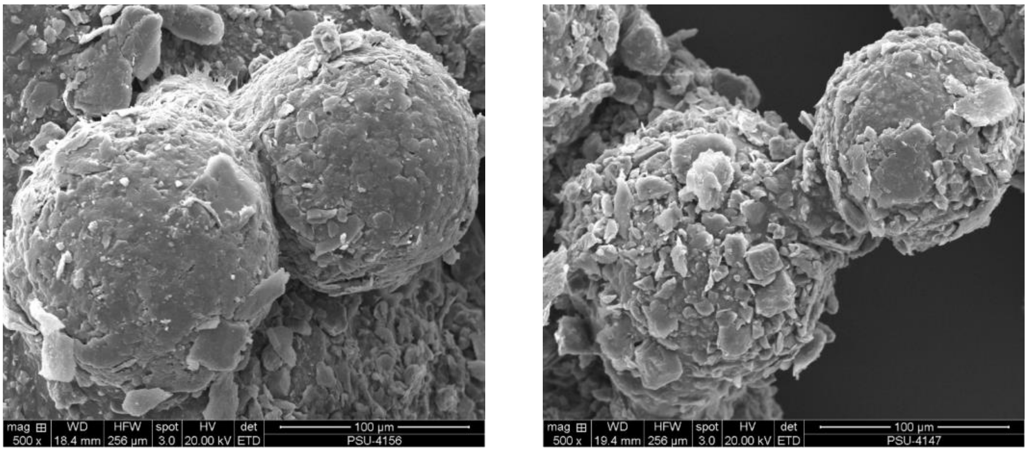
Besides this previous result, the morphology of spray dried rubber latex and talcum powder at different mass ratio was shown in Figure 9(c-h). The natural rubber particles from spray drying were spherical in shape and had more agglomeration observed at the higher flow rate of the feed latex ratio.

Meanwhile, the flow rate of fed latex was elevated to 60 ml/min, the morphology of particles changed, Figure 9(g). Spherical particles and a large size distribution of about 100 µm and above 300 µm were observed.



(a) (b) (c)

Figure 7 Effect of nozzle configurations at different air inlet positions: (a) position 1, (b) position 2 and (c) position 3



(a) (b)

Figure 8 Comparison of SEM micrographs under ×500 magnification (a) natural rubber particles and (b) natural rubber particles coated with talcum powder

4.4 Mechanical properties

The results of mechanical properties is summarized in Table 2. It can be observed that the tensile strength of natural rubber powder was strongly dependent on coated with talcum powder. Moreover, the Young's modulus obtained with natural rubber powder is slightly higher than that obtained with rubber block STR 5L. The elongation reduced due to the coating of natural rubber powder with talcum powder, which indicates the quality of the material that may be talcum powder mixed in natural rubber particles. The tensile strength of natural rubber powder and Young's modulus were 16.07 and 2.22 MPa, respectively, while the elongation at break and hardness of natural rubber powder equal to 578 % and 54.40 respectively.

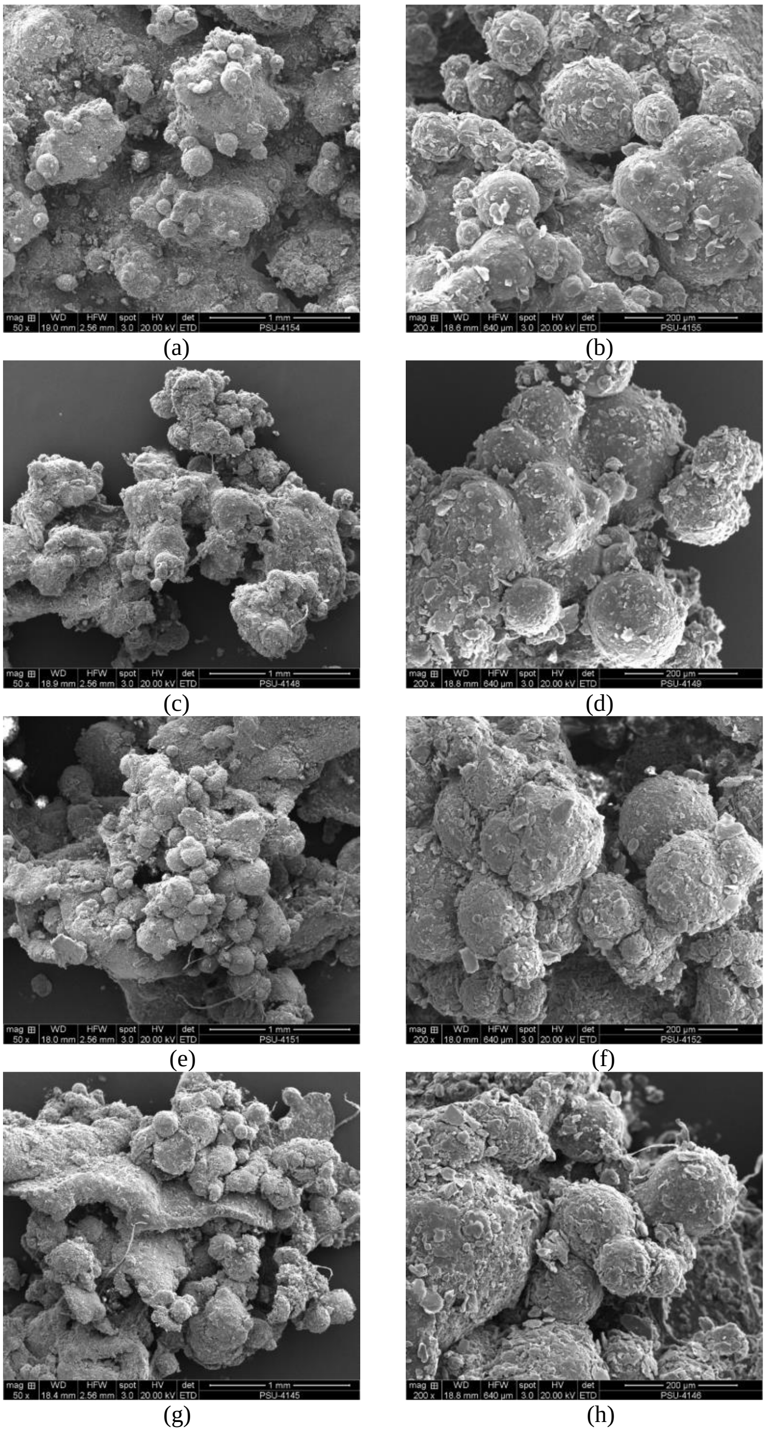


Figure 9 The SEM micrographs for the natural rubber (NR) particles at different magnifications containing: (a) NR particles at magnification ×50 for Q_L = 20 ml/min (b) NR particles at magnification ×200 for Q_L = 20 ml/min (c) NR particles coated with talcum at magnification ×50 for Q_L = 20 ml/min (d) NR particles coated with talcum at magnification ×200 for Q_L = 20 ml/min (e) NR particles coated with talcum at magnification ×50 for Q_L = 40 ml/min (f) NR particles coated with talcum at magnification ×200 for Q_L = 40 ml/min (g) NR particles coated with talcum at magnification ×50 for Q_L = 60 ml/min (g) NR particles coated with talcum at magnification ×200 for Q_L = 60 ml/min

Table 2 The comparison of mechanical properties between rubber block (STR 5L) and natural rubber powder (NRP)

Mechanical properties	STR 5L	NRP
Tensile strength (MPa)	10.52±1.32	16.07±0.94
100% Modulus (MPa)	0.60±0.05	2.22±0.05
Elongation at break (%)	788±76	578±23
Hardness (Shore A)	29.00±0.71	54.40±0.54

5. Conclusion

This paper has presented a method for producing natural rubber powder from a rubber latex. The high ammonia natural rubber latex could produce natural rubber powder followed by spray drying of resulting dispersions with the technique coating or encapsulating the surface of rubber particle with talcum powder. Talcum powder could be used to coat the surface and separate the natural rubber powder while drying. The spherical rubber particles with micrometric sizes in the range were 50 μm to 300 μm.

6. Acknowledgements

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7. References

Anandharamakrishnan, C., & Ishwarya, P. S. (2015). Chapter 8: *Spray drying for nanoencapsulation of food components. Spray Drying Techniques for Food Ingredient Encapsulation* (pp. 180-197), Chichester, UK: John Wiley & Sons Ltd.

Arpagaus, C., & Schwartzbach, H. (2008). Scale-up from the Buchi Mini Spray Dryer B-290 to the Niro MOBILE MINOR, best@buchi Information Bulletin, 52/2008.

Arpagaus, C., John, P., Collenberg, A., & Rutti, D. (2017). Chapter 10: *Nanocapsules formation by nano spray drying* (pp. 346-401) S. M. Jafari, Nano encapsulation Technologies for the Food and Nutraceutical Industries, Inc. DOI: 10.1016/B978-0-12-809436-5.00010-0

Arpagaus, C., Collenberg, A., Rutti, D., Assadpour, E. & Jafari, S. M. (2018). Nano spray drying for encapsulation of pharmaceuticals. *International Journal of Pharmaceutics, 546*(1-2), 194-214. DOI: 10.1016/j.ijpharm.2018.05.037

Bitinis, N., Verdejoa, R., Cassagnau, P., & Lopez-Manchado, M.A. (2011). Structure and properties of polylactide/natural rubber blends. *Journal of Materials Chemistry and Physics, 129*, 823-831. DOI 10.1016/j.matchemphys.2011.05.016

Daisuke, K., Tetsuji, K., & Yousuke, K. 2006, Method for producing rubber from rubber latex, *United States Patent*, 0189732 A1

Eggersdorfer, M. L., Koren, V., Stolovicki, E., Amstad, E., & Weitz, D. A. (2017). Rapid production of submicron drug substance particles by supersonic spray drying. *Crystal Growth and Design, 17*(4), 2046-2053.

International Rubber Study Group (IRSG). (2015). Statistical Summary of World Rubber Situation. Retrieved November 30, 2017, from http://ap.fftc.agnet.org/ap_db.php?id=819.

Jaworek, A., & Sobczyk, A. T. (2008). Electrospraying route to nanotechnology: An overview. *Journal of Electrostatics, 66*(3-4), 197-219.

Li, D., Xia, H., Peng, J., Zhai, M., Wei, G., Li, J., & Qiao, J. (2007). Radiation preparation of Nano powdered styrene-butadiene rubber (SBR) and its toughening effect for polystyrene and high-impact polystyrene, *Radiation Physics and Chemistry, 76*(11-12), 1732-1735.

Markets Insider. (2017). Global and China Natural Rubber Industry Report. Retrieved October 12, 2017, from http://markets.businessinsider.com/news/stocks/Global-and-China-Natural-Rubber IndustryReport-2017-2021-1002266986.

Paiva, L. B. de, Oliveira, A. M. de, & Gavioli, R. R. (2014). Preparation and properties of rubber powder from modified-SBR latex by spray drying process, *Powder Technology, 264*, 507-513. Retrieved from https://www.sciencedirect.com/science/article/pii/S0032591014005270?via%3Dihub

Praktikantin. (2017). Rubber Farm Economics in Thailand. Retrieved October 12, 2017, from https://pugnatorius.com/rubber-farm-investments/.

Sae-Oui, P., Sirisinha, C., Sa-nguanthammarong, P., & Thaptong, P. (2010). Properties and recyclability of thermoplastic elastomer prepared from natural rubber powder (NRP) and high density polyethylene (HDPE). *Journal of Polymer Test, 29*(3), 346-351. DOI: 10.1016/j.polymertesting.2009.12.010

Schmid, K. (2011). *Spray drying of protein precipitates and evaluation of the nano spray dryer B-90.* (PhD's thesis, Ludwig-Maximilians-University, Munich). Retrieved from https://edoc.ub.uni-muenchen.de/13132/1/Schmid_Katja.pdf

Sopanon, P., & Soottitanawat, A. (2011). Preparation of natural rubber powder by spray drying. *TIChE International Conference*, November 10 – 11, 2011 at Hatyai, Songkhla THAILAND, 004-006.

Sosnik, A., & Seremeta, K. P. (2015). Advantages and challenges of the spray-drying technology for the production of pure drug particles and drug-loaded polymeric carriers. *Advances in Colloid and Interface Science*, *223*, 40-54. DOI: 10.1016/j.cis.2015.05.003