Malaysian Journal of Chemistry, 2023, Vol. 25(3), 212-220
(Special Issue: *Rubbercon2022*)

Mini Review on the Preservatives for Natural Rubber Latex (NRL)

Adhikari, A. M. K. S. P.[1], Abdul Rahman, N.[1,2*], Ismail, M. F.[1], and Zainuddin, N.[1]
[1]Department of Chemistry, Faculty of Science, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia
[2]Nanomaterials Processing and Technology Laboratory, Institute of Nanoscience and Nanotechnology, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia
*Corresponding author (e-mail: a_norizah@upm.edu.my)

Natural rubber latex (NRL) harvested from the *Hevea Brasiliense* tree contains colloidal rubber particles, carbohydrates, and various plant proteins. The non-rubber substances in NRL lead to spontaneous coagulation that cause the rubber to rot within 3-5 hours. Many chemicals are used for the preservation of NRL to suppress spontaneous coagulation and putrefaction and improve the stability of the latex for long-term storage. Ammonia (NH_3) is the most commonly used and most effective preservative of NRL. However, it has the potential to cause health problems. Recently several naturally occurring substances, synthetic chemicals, and nanomaterials have been introduced to NRL for preservation purposes. These preservatives are reviewed and their advantages and disadvantages are summarized in this mini-review.

Keywords: Natural rubber; field latex; coagulation; preservatives; ammonia; property enhancement; health effect

Received: January 2023; Accepted: March 2023

While exploring America in 1493, Christopher Columbus came across the indigenous people of Haiti playing with hard bouncing balls [1,2]. At the time, some Latin American countries of the Amazonian region had utilized the same materials for certain simple applications [1,2]. Subsequently, the Europeans adopted the materials for use in Europe. The name "Rubber" was introduced by the Europeans to describe the material because of its ability to erase pencil marks, a minor application of rubber [3]. With continuous research, more unique properties of rubber were discovered and utilized by the Europeans [4,5]. In 1870, the Brazilian rubber seeds were germinated and planted in South and South East Asian British colonies such as Malaysia, Sri Lanka, and Singapore [6]. Today, Malaysia and Sri Lanka are among the countries that significantly contributed to fulfilling the world rubber requirements [7].

Rubber has to be harvested from the rubber tree in the form of natural rubber latex (NRL). The NRL has a short lifetime and can easily rot. However, the NRL product industry has flourished with the discovery of latex preservatives that are able to keep the latex from rotting and coagulating for a long period of time. Thus, NRL preservatives have become an important subject of the NRL product industry. In the literature, a limited number of journal articles reported on NRL preservatives where most of them were written in the mid-19th century [8]. The preservatives and techniques established in the field are also limited with the same techniques and preservatives are being used for over 100 years and only a few new modifications were introduced [6]. The most commonly used preservative for NRL is ammonia which has been used for more than a hundred years. A few non-ammonia-based preservatives have also been introduced. To the best of our knowledge, there is no review paper or a critical evaluation of those preservatives published in the literature, thus, this review will benefit those who want to start working on natural rubber latex preservatives.

1. Composition of NRL

NRL is obtained from the bark of the genus *Hevea-Brasiliense* trees as a milky white color sap (Figure 1) with a distinct odor [9]. NRL contains about 35% rubber hydrocarbon as 1,4-*cis*-polyisoprene (Figure 2), about 60% of water, and 5-6 % of non-rubber substances (Table 1) such as carbohydrate, sugar, phospholipids, proteins, yellow pigments, and metal ions [10,11].

†Paper presented at Rubbercon2022

Figure 1. Tapped rubber tree.

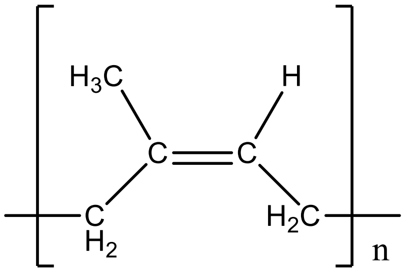

Figure 2. Chemical structure of rubber; (*cis*-1,4-polyisoprene).

Table 1. Typical composition of Natural Rubber Latex (NRL).

Ingredients	Approximate Quantity (% w/v fresh field latex)
Rubber hydrocarbon	35.0
Protein	1.5
Carbohydrates, Sugar	1.5
Phospholipids	1.0
Organic substances, pigments	0.5
Inorganic substances (Mg^{2+}, Ca^{2+})	0.5
water	60.0

The collected latex can be processed in two ways through dry rubber processing or concentrated latex processing. Dry rubber processing leads to a limited number of applications. Thus, concentrated latex processing is the preferable method for processing. However, concentrated latex processing requires long-term latex storage. The presence of the other non-rubber components in the NML leads to latex degradation and causes rotten latex [14]. Because of this, latex preservation is an essential step to keep the latex from rotting. until the completion of its various forms of production. It is possible to keep NRL for up to six months or even more with the correct preservation of NRL [12]. Over the years, different chemicals have been used to preserve NRL. However, some of the chemicals used are hazardous to humans and to the environment, and only a few of them are user-friendly [13].

2. Coagulation of Latex

The process of latex rotting involves a critical step called coagulation. The coagulation process is essentially the combination of the rubber components in the latex that caused these rubber components to be inseparable [14]. The rubber components of latex are typically in the coil conformation protected by a membrane with net negative charges. Because of the net negative charges, the rubber components repel each other. This in turn prevents the rubber components from combining with each other, thus preventing coagulation [15,16]. Figure 3 illustrates how the spontaneous coagulation of latex occurs.

Essentially, the non-rubber components that are present in latex provide a good medium for the growth of microorganisms in the latex [15]. Sugar and carbohydrates are ideal media for the growth of bacteria [16, 17]. These airborne bacteria gradually break down the protein, producing volatile fatty acids (VFA) such as formic, and acetic acid that will decrease the pH level of the latex. The acid is then ionized, which creates hydrogen ions that neutralize the negative charges on the rubber components of latex. This process leads to the formation of neutral rubber particles and allows a combination of rubber particles to take place [18]. When these neutral rubber particles collide, their outer membrane layers break up and free the rubber polymers from it. Once the rubber polymers are released from rubber particles, the rubber polymers combine with each other to form large lumps that precipitate out of the latex.

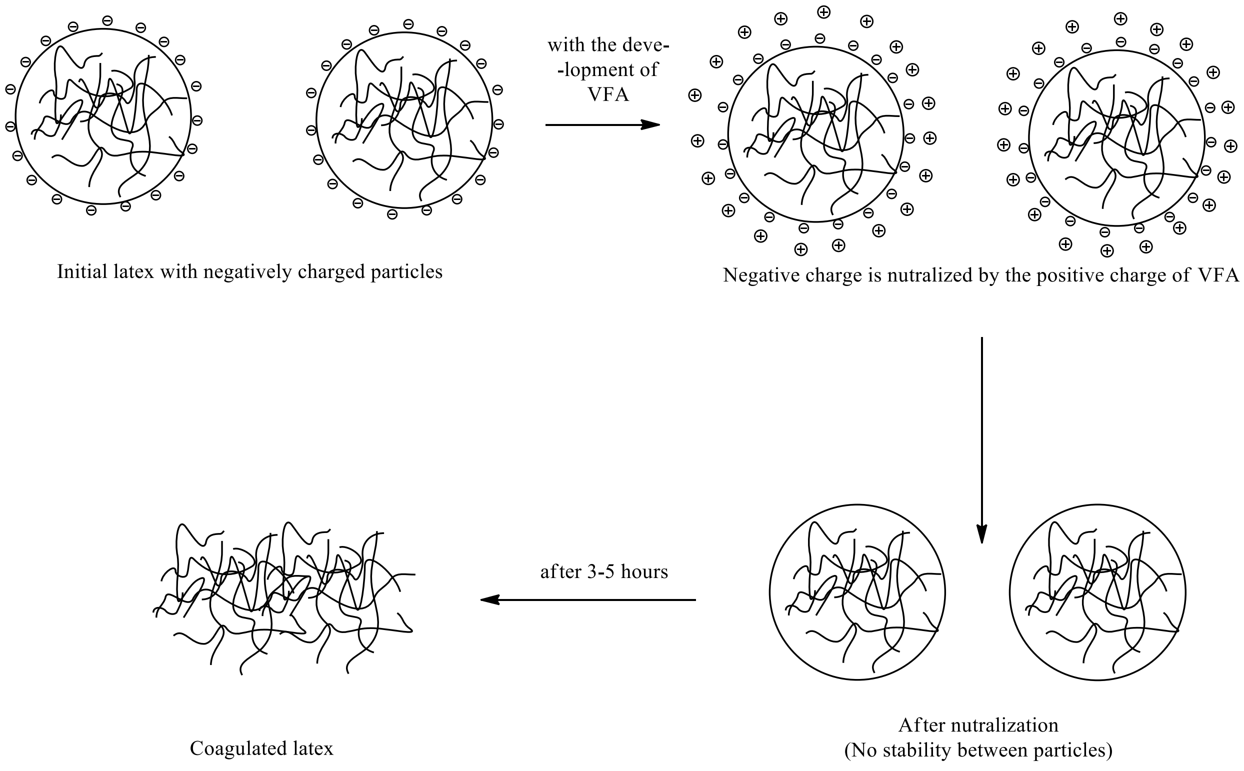

Figure 3. The mechanism for the spontaneous coagulation of NRL.

Besides the non-rubber components of the latex, the contaminated latex receiving cups, collecting buckets, tapping panel bark, and rain guards also have some influence to enhance bacterial growth [19]. This is a non-desirable process because it disrupts further processing of NRL [20]. Generally, pre-coagulation can be identified by the visible clots in the collecting cups, clots in the collected latex, fermentation bubbles in the latex coagulum, etc., giving a bad odor and discoloration. Therefore, it is a must to prevent pre-coagulation until the final product is formed and this is also a major challenge to the latex preservation process [21].

From the mechanism of latex coagulation, it is obvious that latex preservation is a process of maintaining the electrostatic stability of the latex within a period of time, so that latex remains stable in its aqueous solution [22]. Latex preservation can be classified into two categories based on the ammonia level applied [23]. The first type is long-term preservation, which involves 0.6-0.7 % of ammonia that enables the transportation and storage of latex until it is ready to be processed by the consuming countries. Another type of latex preservation is short-term preservation, which keeps the natural rubber latex (NRL) in a latex state for a limited period, usually a few days, before processing. This method is commonly used in the production of dry rubber. [16].

3. Anticoagulants

Different chemicals are used as anticoagulants to prevent spontaneous coagulation in the rubber field or in the transportation tanks for short-term preservation before applying the proper preservative system. They are usually called anticoagulants and are added to the collecting vessels by the tappers in the rubber field [23]. The main purpose of adding anticoagulant is to stop coagulation until the latex reaches the processing factories.

Sodium sulfite (Na_2SO_3) is a mild alkali compound that has been used as an anticoagulant for NRL. Sodium sulfite is occasionally used to inhibit pre-coagulation for brief periods [23]. The recommended level of usage is usually between 0.05 to 0.15% by weight of latex in the field and 0.03-0.05% by weight of latex in the transportation tanks [24]. In addition to prevention from coagulation, it can support the prevention of enzymatic discoloration that happens due to various reasons such as tapping after wintering, new tapping cuts, high tapping intensities, etc. [24]. Sodium sulfite is one of the preservatives recommended to preserve the latex intended to be used for crepe rubber manufacture.

Another common anticoagulant used is sodium carbonate (Na_2CO_3), which is also called washing soda.

It is typically used by adding 0.025 % by weight to the latex in the field and 0.005% to the latex in the transportation tank. The usage of sodium carbonate requires careful application as over-addition will cause bubble formation in the latex in the transportation tank.

Ammonia is also used as an anticoagulant in the field and in transportation tanks to prevent coagulation. For short-term preservation, ammonia is normally added in the field at 0.01 to 0.05% by weight to the latex in the field, and 0.005 to 0.01% by weight to the latex in the transportation tanks [24].

Sodium bicarbonate ($NaHCO_3$) has also been used as a short-term preservative. Mostly, it is applied in crepe rubber manufacturing as an alternative to ammonia since it does not stain crepe-like ammonia does. However, the quantity of $NaHCO_3$ required is relatively higher than that of ammonia and subsequently, the cost is also higher. Therefore, this is only applied to the latex used for white crepe rubber manufacturing [25].

4. Preservation of Field Latex by Ammonia

The industrially viable and highly consumed preservative system for NRL preservation is ammonia. Ammonia plays several roles in latex. It acts as an antibacterial, as an alkali, and as a metal ion deactivator [23]. It can perform as a bactericide to suppress the bacterial growth of latex [26]. This helps to prevent the generation of VFA and acidity in the latex. When NRL protein is attacked by bacteria, lactic acid is produced which generates hydrogen ions. However, hydroxide ions produced from the reaction of ammonia with water neutralize these hydrogen ions. As a result, no hydrogen ions exist to neutralize the negative charges on the rubber particles, preventing them from combining and coagulating.

Ammonia also deactivates metal ions like magnesium and calcium present in latex by forming an ion complex. Ammonia also helps to keep the colloidal stability of the latex for a longer time [23]. According to US Patent 5840790, the level of ammonia used for effective preservation is 0.1 % to 0.5 % by weight of typical field latex [27]. Ammonia preservation can be done using two methods; a high ammonia method and a low ammonia method. In the high ammonia method, 0.7% of ammonia (NH_3) by weight is added to the latex [16]. In the low ammonia method, 0.2% of ammonia and two other ingredients which are tetramethylthiuram disulfide (TMTD) (0.025% by weight) and zinc oxide (ZnO) (0.025% by weight) are added to the NRL [16].

Although ammonia is the most commonly used for latex preservation and seem harmless to human, there are some disadvantages of using ammonia such as air pollution [28], corrosive effect on human skin, eyes, and respiratory systems, and unpleasant odor that is harmful to the human brain cells [29]. In addition, it can also damage rubber itself by discoloration of latex and due to higher pH needs more acids to coagulate in the latter stages of processing. It also causes corrosion to the equipments [30].

5. Preservation of Centrifuged Latex by a Dispersion of TMTD/ZnO

Centrifuged latex is the primary form of preserved latex that can be kept for a long time and is the form of latex used for exportation to different parts of the world for various dipped products and other latex-based product manufacturers. In centrifuged latex, there are two different systems that are manufactured by varying the concentration of ammonia in latex. If the ammonia concentration is 0.2 to 0.4 % by weight, the centrifuged latex is called low ammonia (LA) preserved latex and it can be kept for two to three months without destabilization. If the ammonia concentration is 0.6 % to 0.7 % by weight, it is called high ammonia (HA) preserved latex which can be kept for more than six months without putrefaction [23]. Unsurprisingly, usually, centrifuged latex manufacturers have a preference for HA latex.

In a typical centrifuge, machine latex rotates between 7,000 to 10,000 rpm and generates a force of about 8,000 times of gravity, enabling fast separation. Since the heavier particles are forced outward and the lighter particles inward, at the top of the centrifuge the output runs into two streams. One consists of rubber which has 60% dry rubber content (DRC) and the average volume is about 45% of the total stream throughput. The other 55% parts of the stream throughput usually called skim rubber which contains 3-6 % of DRC.

TMTD and ZnO dispersion which is a bactericide is used as the common secondary preservative in addition to ammonia in centrifuged latex production. Usually, this dispersion is added 0.02 to 0.05 % by weight in the latex for effective preservation [23]. However, TMTD is known to generate carcinogenic nitrosamine at elevated temperatures which are harmful to humans and the natural environment [31]. This has been a huge drawback to the rubber industry for centuries. ZnO itself has an issue that leads to the thickening and destabilization of the latex. Therefore, scientists and rubber technologists have been conducting research to find alternative chemicals for ammonia (NH_3) and TMTD/ZnO.

6. Latex Preservation by Other Chemicals

Broeck and co-workers introduced a mixture of formaldehyde (HCOH) and alkali metal hydroxides such as NaOH, and KOH for preservation in early 1957. They found that a mixture of HCOH 0.2 % to 1.0 % by weight on latex and NaOH/KOH 0.1% to 0.4% by weight on latex can preserve NRL efficiently

[32]. Usually, formaldehyde kills bacteria along with enzymes [33]. However, it decreases the pH level of latex to 6.5 which leads to partial coagulation and thickening of latex with time. This is one of the reasons for the poor mechanical stability of the latex preserved with this mixture. Later it was found that the mechanical stability can be increased by adding polyvinyl methoxy ether or polyvinyl methyl ether in the range between 0.01% and 0.2% by weight on latex along with HCOH [34]. Therefore, it is not recommended to use HCOH alone as a preservative.

Some amines, such as ethylamine, ethanolamine, ethylenediamine, and isopropanolamine have also been used to preserve latex instead of ammonia [35]. However, the cost of those amines is comparatively higher than that of ammonia, and preservative properties were also relatively poorer. Therefore, those amine compounds were not popular as preservatives for NRL.

The preservative potential of methylol compounds such as imidazolidinyl urea, diazolidinyl urea, hydroxymethylaminoacetate, and bicyclic oxazolidine was studied by researchers in Thailand [36]. In their study, they proved that 0.1 to 2.0 g of the methylol compounds on 100 g of latex along with 0.01 g of anionic surfactants can obtain a good preservation effect.

Another chemical preservative was introduced by Wang and his team for use in centrifuged latex. They used a 3 % solution of S-triazine instead of TMTD/ZnO. The preservative properties, changes in the mechanical properties, and cytotoxicity properties of a latex film were studied. They also found that the preservative system can preserve latex for six months effectively [37].

7. Latex Preservation by Natural Substances

Due to the various health problems associated with the toxic chemicals used in the preservatives, many naturally occurring non-toxic substances have been investigated for producing eco-friendly preservation techniques. In 2010, a study on chitosan and its derivatives in order to preserve latex has been conducted by researchers in Thailand [38]. Chitosan is a polysaccharide extracted from the outer skeleton of shellfish such as crabs, lobsters, and shrimps [39]. In their study, they used a series of low to high-molecular-weight chemically modified chitosan derivatives and they observed that the activity of preservation is high with the low molecular weight of chitosan derivatives. However, there is confusion with their study period of 14 hours on field latex preservation since the VFA values were recorded within 14 hours.

Another interesting research has been conducted by a group of researchers in China. They treat bamboo charcoal (in which the particle size is less than 100 nm) with fresh latex without ammonia or with a low amount of ammonia. The treated latex has shown better preservation with longer storage time while enhancing the mechanical properties [40]. However, their dispersant media contain chemicals such as sodium carboxymethylcellulose, polyvinyl alcohol, Tween 80, sodium lauryl sulfate, and dodecyl benzenesulfonate due to the poor solubility of bamboo charcoal in NRL. The main application in the research was natural rubber latex foam. This preservative system is suitable only for some specific applications since bamboo charcoal treatment badly affects the color of the latex.

Five different extracts of biological microbial surfactants were isolated and they were named MRB02, MRB12, MRB16, MRB18, and MRB25 by Hassan & Singh (2010) [41]. The extracts were treated with NRL and it was found that the extracts are capable of using as latex stabilizers while suppressing the bacterial growth of latex. MRB25 showed better performance among the five surfactants. It was also discovered that the MRB25 surfactant can deactivate the metal ions like Ca^{2+} and Mg^{2+}. They also found that the Brookfield Viscosity of the treated latex was also similar to ammonia-treated latex. Therefore, MRB25 has the potential to be used as a natural preservative for NRL preservation. However, it has not been shown that the properties of the MRB25-treated latex products are similar to or better than that of conventionally preserved latex products.

Hydrophobically modified fructan (saccharide), hydrophobically modified starch hydrolysate, and a mixture of them were separately studied by Booten and co-workers (2010). They discovered that the aqueous dispersion of each one of the above and mixture also can preserve NRL effectively and obtained a patent for their finding [35]. According to the discovery 0.2% by weight of ammonia needs to be added to the latex first before 0.001 % to 0.35 % by weight of hydrophobically modified saccharide is added. It is claimed that this system can exist for more than 20 weeks without destabilization. However, the method has not yet been applied in large-scale productions in the industry.

8. Latex Preservation by Nanomaterials

With the massive advancement and expansion of the nanomaterials applied in the polymer industry and especially with rubber-related applications in the last two decades, it has been shown that the enhancement of physicomechanical, electrical, and thermal properties of rubbers can be achieved with the incorporation of nanomaterials in the latex. Since nano-metals have shown their antibacterial activity towards biological systems they have also been introduced for NRL preservation. However, only a few studies were found using nanomaterials for the preservation purposes of latex. Silver nanoparticles have shown antibacterial activity towards NRL. With the enhancement of mechanical properties and the antibacterial activity of silver nanoparticles, it is very suitable to be used as a preservative for NRL. It has also shown better dispersion in the latex matrix [42].

An increment of the antibacterial activity towards both gram-positive and gram-negative bacteria with silver nanoparticle-incorporated foam rubber has been observed in laboratory-level experiments [43]. A similar study has confirmed that silver nanoparticles can inhibit the growth of bacteria and minimize the formation of VFA which results in the latex stability in latex foam rubber [44]. However, one drawback of silver nanoparticles is their comparatively expensive price compared to conventional preservatives. Hence it is not an industrially feasible material to preserve latex.

Nano metal oxides have also been shown antibacterial properties against bacteria in rubber latex. Recently, a study on nano zinc oxide (ZnO) was carried out by a researcher in Thailand and highlighted the better preservative properties of nano ZnO and a mixture of urea, isothiazolone, and nano ZnO against NRL. It is claimed that adding 0.2 % to 0.8% by weight of the mixture on dry latex weight can effectively preserve the dry latex. Application on centrifuged latex uses 0.1% to 0.3% by weight of the solution can also preserve the centrifuged latex well. However, the ratio was not revealed in the paper. It is also proposed that the higher level of oxidative stress of the nanostructure of ZnO will encourage the death of bacteria cells owing to the interactions between reactive oxygen species and cell membrane, proteins, or DNA causing an adequate number of termite death. [45].

A similar study has been conducted by a group of Sri Lankan researchers with low ammonia TMTD/ZnO system in centrifuged latex preservation (LATZ) [46]. In their study, they claimed that nano ZnO preserved LATZ latex can exist for 75 days without destabilization while showing similar mechanical properties with the conventional LATZ systems. There were numerous preservative systems were introduced by different researchers. Their advantages and disadvantages are summarized in Table 2.

Table 2. A summary of the advantages and disadvantages of the preservatives.

No	Preservative System	Advantages	Disadvantages
1	Ammonia [24, 25, 26]	Perform as a bactericide and deactivate metal ions like Mg^{2+} and Ca^{2+} present in latex. Helps to keep the colloidal stability of the latex.	Cause polluted air, corrosive effects on human skin, eyes, and respiratory systems, and unpleasant odor damage to human brain cells.
2	TMTD/ZnO [25,27]	A good bactericide, Not affect the color. Use in smaller quantities.	Cause to generate carcinogenic nitrosamine at higher temperatures.
3	Mixture of formaldehyde (HCOH) and alkali metal hydroxides such as NaOH, KOH [24, 25, 32]	Efficient preservative	Decrease the pH level of latex to 6.5 which leads to partial coagulation and thickening of latex. Poor mechanical stability.
4	Some amines, such as ethylamine, ethanolamine, ethylenediamine, isopropanolamine [25,33]	Preserve latex.	The cost is high and dropping latex properties.
5	Modified chitosan derivatives [38, 39]	Not hazardous to humans.	Not a simple method and the synthesis cost is higher.
6	Nano scale bamboo charcoal [40]	better preservation with longer storage time, a natural substance	Dispersant media comprises few chemicals and charcoal affected badly towards the color of the product.
7	Extracted biological microbial surfactants [39]	capable of using as latex stabilizers while suppressing the bacterial growth of latex, also deactivate metal ions like Mg^{2+} and Ca^{2+}	The latex properties after the treatment have not yet been proven.

8	Hydrophobically modified fructan (saccharide), hydrophobically modified starch hydrolysate, and a mixture of them [35]	Preserved latex can exist for more than 20 weeks without destabilization.	Not feasible in industry-level applications.
9	Silver nanoparticles [42,43,44]	Better preservation, good dispensability in the latex matrix, and enhancement of mechanical properties.	The production cost is higher when compared to ammonia.
10	Nano ZnO [45]	Can preserve latex and keep it for 75 days without destabilization.	Not yet proven for industrial-level applications.

Although a considerable amount of novel preservative systems were successfully studied and applied for NRL, none of those systems were feasible to use in industrial-level applications. Therefore, ammonia is still the dominant chemical as the ideal primary preservative to preserve field latex despite some of its drawbacks. The common secondary preservative system is TMTD/ZnO even though it is known to generate carcinogenic nitrosamines compound.

Some of the preservative systems such as silver nanoparticles, and nano zinc oxide have shown enhancement of other properties such as thermal properties, electrical conductivity, physicomechanical properties, and dynamic mechanical properties of the latex-based products in addition to preservation.

CONCLUDING REMARKS

Natural rubber latex (NRL) is one of the most important natural polymers for human society. NRL has been extensively used as a renewable raw material in the production of a wide range of products. The applications include medical devices, surgical gloves, aircraft and car tires, pacifiers, clothes, toys, etc. Latex preservation is directly linked with the rubber industry, without proper preservation, the NRL will coagulate and cannot be maintained at the highest quality. The number of studies conducted on the preservation of natural rubber latex is still considered low and the major preservative system is ammonia despite the health consequences posed by it. New research innovation is required to find not only environmentally friendly and human-safe natural or synthetic compounds that can preserve NRL. It also must be economically feasible in order to be applied in the factories. Otherwise, the research findings are only for publications and have no real-world value.

ACKNOWLEDGEMENTS

The authors are grateful for the financial support from the Fundamental Research Grant Scheme (FRGS) from the Ministry of Higher Education Malaysia (FRGS/1/2019/STG01/UPM/02/7).

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