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Permeation of salicylic acid through skim natural rubber films

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ABSTRACT

The skim natural rubber latex (SNRL) with about 4 wt% dry rubber content was incubated with 0.012 wt% urea and 0.025 wt% ethanol both without and with stirring for various times to yield deproteinized rubber films (DSNR). The extractable protein (EP) content was determined by modified Lowry method. The salicylic acid permeation through rubber films was checked by measuring the concentration on the sink side at various times by using spectrophotometer at the wavelength of 344 nm. The studies show that after SNRL were incubated both without and with stirring, the up-and-down patterns of EP contents were seen along the incubation time. Incubation with stirring significantly reduced the EP content and the minimum was seen when SNRL was incubated for 1 h with 52% reduction. The findings also suggest that the SNRL film with a higher EP content yielded the higher the flux. The permeation kinetics through SNR films followed Fickian diffusion. Apart from being more easily peeled of glass plates, SNR films containing higher EP contents were superior to HANR films for their higher permeation fluxes. In addition, the flux of the SNR/HANR blend increased with increasing SNR fraction so the permeation rate could be controlled by adjusting the blend ratio.

1. Introduction

Nowadays, the natural rubber obtained from *Hevea brasiliensis* is a major raw material in the rubber product industry. Because of its excellent flexibility, the natural rubber has been applied to many consumer products such as pacifiers, gloves, condoms, shoe soles, vehicle tires and medical products. The field natural rubber latex (FNRL) contains about 30 wt% rubber particles (30% DRC or dry rubber content). After the FNRL is centrifuged, the main product of the concentrated rubber latex (HANRL or high ammonia natural rubber latex) with 60% DRC is obtained together with the skim natural rubber latex (SNRL) with 4–6% DRC as a by-product. While the HANRL is applied either in the latex form for the dipped products or in the solid form for the molded products, the SNRL is formed into skim blocks by coagulating the rubber particles with a sulfuric acid solution at a high concentration. Skim blocks are, therefore, a low-grade rubber used in the mixing process with a higher standard rubber to lower the viscosity during the mixing.

Thailand ranked first as an exporter of rubber blocks and rubber latex in South East Asia in 2015. It was reported that the concentrated latex was produced in a large quantity of 795,098 tons in 2015, along with 62,600 tons of skim rubber, which was double the previous year's production. The production rate is still increasing every year but the value of the rubber as a raw material in the market is decreasing. To sustain its value in the market, as indicated in the national policy, Thai government has attempted to promote more uses of the natural rubber against the current widespread uses of synthetic rubber. The policy, thus, motivates and supports the findings of novel potential applications of the rubber and all components in the latex, including skim rubber and the left-over serum, to boost the value of natural rubber as a whole. In addition, to implement the sustainability concept of using biopolymers which are a large renewable resource, this research investigates a new application of SNRL whose use is still limited.

Subject to the high force in centrifugation, the SNRL contains smaller particles than the HANRL does, leading to differences in film properties. Rippel et al. (2003) studied the film formation of the concentrated and the skim latices. The results of infrared spectroscopy showed that the rubber particles were composed of various functional groups including isoprene, proteins and phospholipids. In addition, the protein contents of the rubbers in the skim latex were higher than those of the particles in concentrated one due to the larger specific surface area of skim rubber particles. The results of atomic force microscopy (AFM) also showed that fluidity of particles in the skim latex was higher than that of particles in the concentrated latex so the skim natural rubber particle could spread to become flatter. Moreover, when both latices were formed into rubber films, skim rubber particles were aggregated more easily, resulting in a smoother film.

The SNRL could be applied extensively in the form of a film.

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However, the investigations of rubber films have usually been done for the concentrated latex. They were applied in the control release of a compound fertilizer prepared by granulation machine and being coated with various polymers (Hanafi et al., 2000). As a drug release application, ketoprofen pseudolatex gel from ethyl cellulose and deproteinized natural rubber latex (DPNRL) for transdermal drug delivery systems with the different enhancers was investigated (Suksaeree et al., 2014). The rubber films could also be applied as transdermal films for controlling the release of nicotine which was used in the treatment of smoking addiction. The factors investigated include the thickness of the rubber layer and the concentration of nicotine (Pichayakorn et al., 2013), the blend of the natural rubber and other polymers (Suksaereea et al., 2012; Pichayakorn et al., 2013) and the effect of plasticizers (Pichayakorn et al., 2013). Recently, the membrane of the blend of the deproteinized natural rubber (from the FNRL) grafted with poly(dimethylaminoethyl mathacrylate) and poly(vinyl alcohol) has been investigated for the release behavior of rhodamine-B as a model drug in water after the membrane was in equilibrium with rhodamine-B (Jayadevan et al., 2018).

Since medical films may be applied on the human skin, the release of allergenic proteins from natural rubber products in some cases could cause problems to users. Thus, there have been many works investigating the removal of allergenic proteins from natural rubber latex to obtain deproteinized natural rubber latex (DPNRL). Some of reported methods to remove proteins from the surface of rubber particles in latices are the treatment of latex with denaturants such as urea and sodium dodecyl sulphate (SDS) (Kawahara et al., 2004), the enzymatic treatment (Kawahara et al., 2004; Sakdapipanich et al., 2015), the treatment of latex with polar organic solvents such as acetone, ethanol and 2-propanol in the presence of SDS (Chaikumpollert et al., 2012), the saponification of the latex with sodium hydroxide (Sakdapipanich et al., 2015), and the protein extraction with polyethylene glycol (PEG) with different molecular weights in batch and flow systems (Danwanichakul et al., 2014). Not only were the investigations of the protein removals attempted in the latex phase but protein removals from the rubber films were also studied. Kalapat et al. (2009) studied the protein extraction from skim rubber films by considering three factors, which were the type of extracting medium, the number of extraction steps and the film area.

This work focuses on the effect of the non-rubber components in the skim rubber, among which proteins were of interest, on the permeation behavior of a model drug through rubber films. To prepare the rubber films with different protein contents, the deproteinization of SNRL was incubated with urea and ethanol. Such films were then tested for the permeation of salicylic acid, a major active ingredient in acne treatment, which was chosen as a model drug in this study. Even though the deproteinized rubber films were successfully applied for controlled drug releases, there has been no discussion about the effect of proteins on the release. This study might in some way be related to the study of the effect of the enhancer in polymer matrix (in our case, removing the enhancer). Additionally, there has been no investigation of using the skim rubber for this application before so the comparison between the skim and concentrated rubber was made in this work. Such body of knowledge could somehow boost the use of natural rubber, especially for the skim rubber in this particular application.

2. Materials and methods

2.1. Materials

The skim natural rubber latex (SNRL) with 4.18%DRC was obtained from Rubber Estate Organization, Nakhonsrithammarat, Thailand. Sulfuric acid and acetic acid were supplied from J.T. Baker Ltd. Sodium deoxycholate (DOC) and polysorbate were purchased from Sigma-aldrich. Ovalbumin, salicylic acid and phosphotungstic acid (PTA) were purchased from Himedia Laboratories Pvt., Ltd. Trichloroacetic acid (TCA) was purchased from Fisher Chemical. Sodium carbonate was purchased from a division of Nuplex Industries (Aust) Pty., Ltd. Sodium hydroxide was purchased from Merck. Folin was purchased from Loba chemie Pvt., Ltd. Propylene glycol was purchased from Ajax Finechem Pty., Ltd. Toluene was purchased from RCI Labscan limited. Sodium tartate was purchased from Qrec, New Zealand. Copper (II) sulfate was purchased from Suksapanpanich, Thailand.

2.2. Methods

2.2.1. Determination of dry rubber content (DRC)

A 5.0 g sample of the rubber latex was poured into a Petri dish and diluted with 10 ml of distilled water. A 2 wt% sulfuric acid solution was slowly added until the rubber particles were coagulated. The coagulum was removed and the remaining serum was filtered with nylon cloth 3 times so more rubber was obtained. The rubber coagulum was made into a rubber film and the film was washed with water several times to remove sulfuric acid. It was dried at 50 °C for 24 h and weighed.%DRC is determined as the percentage of the mass of dried rubber in total mass of original latex used in the experiment.

2.2.2. Determination of alkalinity of latex

A 5.0 g sample of the natural rubber latex was mixed with 200 ml of distillated water in Erlenmeyer flask and 3 drops of the indicator was added. After that, the solution was titrated with a 0.1 N HCl solution until the color of the mixture was changed from yellow to pink. Then, the alkalinity was calculated from Eq. (1)

$$\%Alkalinity\ (\%\ NH_3\ in\ one\ unit\ of\ rubber\ latex\ mass) = 1.7NV/m \quad (1)$$

Where, N is the normality of HCl solution, V is the volume of the acid solution used in the titration (ml) and m is the mass of the rubber latex (g).

2.2.3. Preparation of deproteinized natural rubber films (from SNRL and HANRL)

The SNRL was diluted to yield a 4%DRC suspension and incubated with a solution of 0.012 wt% urea and 0.025 wt% ethanol for various incubation times including 0.5, 1, 2, 4 and 24 h. The effect of stirring with an overhead stirrer during the incubation was also observed. In case of stirring, it was stirred at 150 rpm at room temperature. After that, the suspension was centrifuged at 7000 rpm for 5 min to obtain the cream fraction. It was then coagulated with a 2 wt% sulfuric acid and the coagulum was dried at 60 °C. A 1 g sample of the rubber coagulum was dissolved completely in toluene and the solution was cast into a film. The procedure was the same for the preparation of the films from HANRL but only the incubation with stirring was performed.

2.2.4. Preparation of rubber films from deproteinized HANRL/SNRL blends

The rubber blend was obtained by mixing HANRL and SNRL, of which the composition was varied to be 75:25, 50:50 and 25:75. Before mixing, both HANRL and SNRL were diluted to yield 4%DRC suspensions. The mixture was mixed with urea and ethanol as discussed in Section 2.2.3 by using an overhead stirrer for 1 h. Then, the suspension was coagulated by a 2 wt% sulfuric acid solution and the coagulum was dried at 60 °C. The dried coagulum was dissolved in toluene and cast into the film.

2.2.5. Determination of extractable protein content by modified lowry method according to ASTM D5712

The calibration curve of the standard protein (ovalbumin) was prepared first. The phosphate-buffered saline (PBS) was prepared by dissolving 3 g of PBS in 1000 ml of distilled water and 0.1 g of ovalbumin was then added into the 50 ml of PBS solution to yield a 2000 ppm ovalbumin solution. To construct a calibration curve, the ovalbumin solution was diluted to be 2, 4, 6, 8 and 10 ppm and was

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