J. Rubb. Res., **6(1)**, 12–35

Concentration of NR Field Latex Using Tubular Cross Flow Ultrafiltration System

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This study presents an alternative method of concentrating field latex (FL) that produced latex concentrate and a clear serum as a by-product. The serum obtained could be utilised for the extraction of useful biochemicals; and as it was not contaminated by sulphuric acid, value-added products from waste could also be obtained. A tubular cross flow ultrafiltration system, one of the pressure-driven membrane separation techniques, was used to examine the concentration process of FL. A suitable preservation system to increase the stability of latex to withstand mechanical shear stresses during ultrafiltration was investigated Fouling of membrane could be reduced by a cleaning-in-place (CIP) technique and also by adopting suitable cleaning protocols Results showed that FL with a suitable composite preservation system could be concentrated from 30% DRC to 46% DRC, by applying CIP with transmembrane pressure of 2 5 bar.

Key words: field latex, latex concentrates, serum; latex-stability; ultrafiltration; membrane separation, transmembrane pressure

NR latex as an industrial raw material is supplied as concentrated latex where the DRC is increased from ±30% to ±60%. It serves as the main raw material for manufacturing NR latex products. Three methods of concentration currently used are centrifugation, creaming and evaporation. Centrifugation is widely used[1] and accounts for some 95% of the total concentrates produced in Malaysia. Centrifugation produces skim latex as a by-product. Skim rubber processing involves the recovery of 4%–5% of dry rubber from skim latex by cheap grade sulphuric acid coagulation that leaves the serum portion contaminated by the acid. The bulk of the serum contaminated by sulphate ion together with the washings is eventually discharged into effluent ponds. This sulphate ion releases hydrogen sulphide gas while undergoing microbial breakdown in anaerobic effluent ponds causing malodour and environmental degradation[2 3].

The single largest component of latex serum (about 23% by weight of the total non-rubbers) is a water-soluble carbohydrate quebrachitol[4–5], which is a chemical feedstock for the synthesis of a range of bioactive materials. An economically feasible method for extracting this product from the serum

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was developed[6]. If all the serum from NR latex processing could be recovered uncontaminated by sulphuric acid, the biochemical extraction of quebrachitol[4–6] can be carried out. This would very much boost the income from latex and may well exceed that obtained from the sale of rubber

A suitable processing technology needs to be developed to separate serum from latex to avoid obtaining skim latex so that both products can be economically utilised *i e* latex concentrate for manufacturing latex products and serum for biochemical extraction This process could eventually pave the way for isolating value-added products from serum and discharging minimal amount of effluent, thus making latex processing environment-friendly[7] Membrane separation technology, in particular, ultrafiltration, could become an alternative method of concentrating NR latex, which unlike centrifugation, produces concentrated latex and a clear serum that is not contaminated The process of biochemical extraction would be a worthwhile option and further separation processes would be carried out to isolate them once the concentration process is perfected. Engineers in this field have long recognised ultrafiltration as a possible separation method. The major difficulties encountered in this endeavour were the instability of latex and non-availability of suitable membrane material capable of withstanding the highly alkaline feed[8]

Membrane can selectively separate components over a wide range of particle sizes and molecular weights, from macromolecular materials such as starch and protein to monovalent ions Membrane processes are classified according to the driving force used in the process. Driving force widely applied includes pressure, electrical potential, partial pressure and concentration gradient[9]

Membrane separation process which uses pressure as the driving force are reverse osmosis, nanofiltration, ultrafiltration (UF) and microfiltartion. These processes differ from each other depending on the pore size of the membrane used and magnitude of the applied pressure (transmembrane pressure) UF was found more suitable for concentration of NR latex due to its wide range of molecular distribution[10] and high solid content.

In this study UF experiments of NR latex were carried out to identify a suitable composite preservation system between two available options. [ammonia, ammonium laurate, TMTD and zinc oxide (Option 1); ammonia and Terric[8] (Option 2)], and to study the effects of feed velocity and TMP on permeate flux Attempts were also made to identify the optimum TMP for the concentration process and the degree of concentration achievable was also determined

MATERIALS AND METHODS

Material

For experiments involving UF the latex samples of a multi-clonal type were obtained from the Rubber Research Institute Experiment Station, Sg Buloh, Malaysia. The latex samples were stabilised and preserved with either one of the two preservation systems mentioned above

Preservation System

The most important factor for a successful concentration of NR latex is the stability of latex. To avoid fouling of membrane caused by premature coagulation and to minimise possible destabilisation of latex on encountering mechanical shear stresses during passage through the membrane, a suitable preservation must be used The preservation system used was very similar to the system used during centrifugation[11] The mechanical shear stresses during passage of latex through the membrane is comparable to average centrifugal force equivalent of 8000 g exerted on the latex particles during centrifugation[1]

A composite preservation system consisting of a primary preservative (ammonia) and secondary preservatives [tetra methyl thiuram disulphide (TMTD) and zinc oxide (ZnO)] and a stabiliser [ammonium laurate or Terric® (non-ionic surfactant)] to boost the mechanical stability of latex was formulated[11]

The following preservation systems were prepared by mixing chemicals with compositions based on w/w

- *Preservation system 1 (PS 1)* Ammonia, TMTD, ZnO, and ammonium laurate
- *Preservation system 2 (PS 2)* Ammonia and Terric®

One of the above preservation system was added 20 kg of field latex, with *ca* 30% DRC and left overnight for maturation to allow the preservation chemicals to interact with the latex particles and stabilise it

METHODS

Introduction

UF membrane element configurations There are a number of UF membrane element configurations available such as plate and frames, spiral wound, hollow fiber and tubular forms[12] From previous work done on the concentration of epoxidised natural rubber (ENR)[13], cross flow tubular UF system was found more suitable for the concentration of NR latex

Tubular crossflow UF system In the tubular crossflow UF system, transmembrane pressure (TMP) is the driving force which causes permeation to occur The cross-flow pressure-driven process requires only the pumping of the feed-stream tangentially across the appropriate membrane, *i e* parallel to the membrane surface The membrane splits the feed stream into two one stream is the permeate, consisting of components small enough to pass through the membrane pores, the other stream is the concentrate (retentate) consisting of components large enough to be retained by the membrane The retentate stream is usually recirculated through the membrane module because one passage through the membrane may not deplete the feed significantly Important operating variables applied are TMP and cross-flow velocity Cross-flow velocity is the average rate at which the process fluid flows parallel to the membrane surface Velocity has a major effect on the permeate flux The permeate flux is dependent on the applied TMP for a given surface area up to a threshold TMP Above this pressure, which has to be experimentally determined for each application, higher pressures have little or no effect In fact, too high a pressure may aggravate fouling of the membrane[9]

Hagen-Poiseuille Law Fluid flows through microporous membranes by the Hagen-Poiseuille Law for streamline flow and is the function of pressure drop, viscosity, density and channel dimensions (such as diameter of a tube) to flow rate through a channel

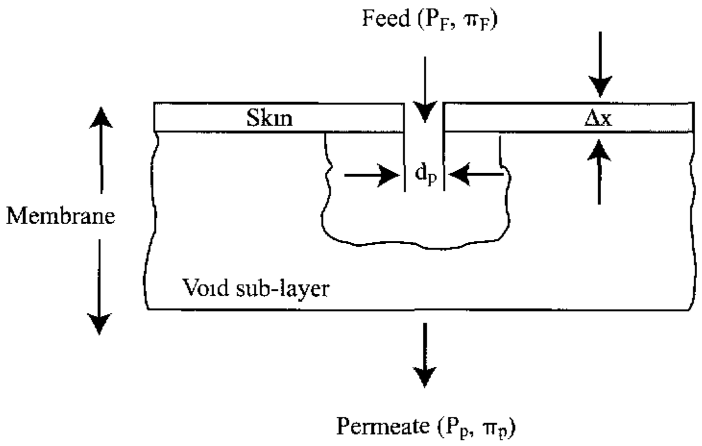

Figure 1 Schematic representation of the cross section of a typical asymmetric UF or MF Membrane P_F *is the applied pressure* P_p *is back-pressure on the permeate side (this is zero if the permeate side is open to atmosphere)* π_F *and* π_p *are osmotic pressures of the feed and permeate solutions (Adapted from Cheryan*[12]*)*

In *Equation 1*, J is the flux rate (L/m²/h) through a membrane, d_P (*Figure 1*) is the channel diameter, P_T is the applied transmembrane pressure μ is the viscosity of the fluid permeating the membrane and x is the length of the channel (the membrane 'skin' thickness) and ε is the surface porosity of the membrane[12]

$$J = \frac{\varepsilon d_P P_T}{32\Delta x\mu} \qquad 1$$

Darcy's Law Pressure controlled region (resistance-in-series) NR latex being a colloidal suspension contains particles that are too large to penetrate the membrane pores, thus a sieving mechanism is dominant and a polarised gel layer of rejected particles forms on the membrane surface The gel layer provides an additional resistance to filtration, so the permeate flux declines with time The polarised gel layer and membrane may be considered as two resistances in series (*Equation 1*), then it reduces to *Equation 2* and the permeate flux is then described by Darcy's Law and takes the form of

$$J = \frac{1}{Am}\,\frac{dVp}{dt} = \frac{|\Delta P|}{(Rm + Rc)\mu} \qquad 2$$

If the membrane is only exposed to pure solvent, the above equation reduces to

$$J = \frac{|\Delta P|}{Rm\mu} \qquad 3$$

Where J = permeation rate (flux)
Am = membrane filtration area
dVp = total volume of permeate
ΔP = transmembrane pressure
Rm = intrinsic membrane resistance (total of membrane and fouling resistances)
Rc = resistance of polarised gel layers deposited on the membrane (filter cake, gel foulants)
μ = viscosity of permeate
t = filtration time

Where, $\Delta P = \frac{P\ + P_i}{2}$, P_i and P are the inlet pressure the outlet pressure gauges, respectively

During the UF of NR latex, if significant fouling occurs because of specific membrane-solute (protein and latex particles) interactions, the intrinsic membrane resistance (*Rm*) would increase. For polymeric membranes, *Rm* was found to increase with increase in TMP due to membrane compaction[14].

Mass transfer model: Pressure independent region. The polarised gel layer resistance (*Rc*) combined with the *Rm* when reaching a limiting value whereby the applied TMP would just cancel off the combined resistances. When this happens it is known as the pressure independent region. This is explained and illustrated in *Figure 2*.

As NR latex is ultrafiltered, solute is brought to the membrane surface by convective transport at a rate, defined as:

$$J_s = J\,C_b \qquad \ldots 4$$

Where: J = permeate flux
C_b = bulk concentration of the rejected solute.

The resulting concentration gradient causes the solute to be transported back into the bulk of the solution due to diffusion effects. Neglecting axial concentration gradients, the rate of back-transport of solute would be:

$$J_s = D\frac{dC}{dX} \qquad \ldots 5$$

Where: D = diffusion coefficient
$\frac{dC}{dX}$ = concentration gradient over a differential element in the boundary layer.

At steady state, the two mechanisms will balance each other; both *Equations 4* and *5* can

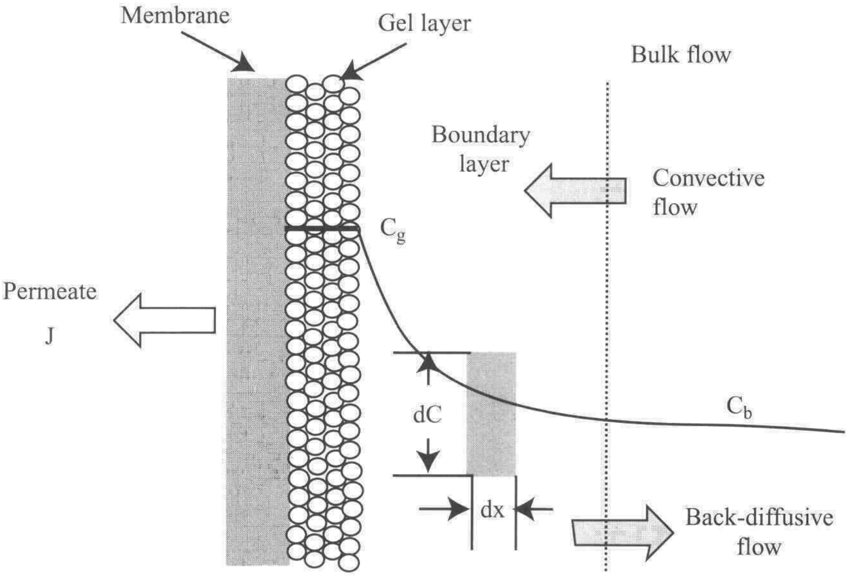

Figure 2. Concentration of polarisation during UF of NR latex the colloidal and macromolecular solutes, showing the built-up of the polarised (gel) layer and associated boundary layer (Adapted from Cheryan[12].)

be equated and integrated over the boundary layer to give

$$J_s = \frac{D}{\delta} \ln \frac{C_g}{C_b} \qquad 6$$

Where C_g = gel concentration
C_b = bulk concentration of the rejected solute
δ = thickness of the boundary layer (D/k)
k = mass transfer coefficient, having the same units as the flux J and calculated as

$$k = \frac{D}{\delta} \text{ where } \quad J = k \ln \frac{C_g}{C_b} \qquad 7$$

This model (*Equation 7*) is valid only in the pressure-independent region The flux will be controlled by the rate at which solute is transferred back from the membrane surface into bulk fluid In most operations C_g and C_b are fixed physicochemical properties of the feed, flux can only be improved by enhancing *k* as much as possible, such as by reducing the thickness of the boundary layer Flux will become independent of pressure and only increase if the back-transport mechanism reduces the gel layer thickness by increasing feed velocity and creating turbulent flow Both these mechanisms increase the shearing action against the membrane thus reducing gel layer resistance[12]

Predicting flux for UF of NR latex During UF of NR latex, two regions in the process are known to exist, these are pressure dependent and independent regions The variations between these are functions of filtration time, nature of filtrating sample and its hydrodynamic characteristics of the feed[14] Since NR latex has a high solid content, the filtration time increases the [Hagen-Poiseuille Equation (*Equation 1*)] and is not applicable as resistances originating from the initial *Rm* with membrane fouling resistance added on to it but is still termed as *Rm* and *Rc*[14] The limiting value would be reached when the driving force, which is the transmembrane pressure, has little or no effect on the flux This is termed as the pressure independent region (*Equation 7*) The available literature does not completely describe the behaviour of NR latex during UF None are completely satisfactory, their biggest failing is the inability to completely describe all regions, from pressure-control to the mass transfer-control region and to the cake controlled region This is due to the complexity of NR latex as the feed which comprises of many different constituents such as rubber particles (*ca* 35%), aqueous phase (*ca* 55%) and lutoid phase (*ca* 10%)[15]

This study was carried out basically by manipulating operating variables such as TMP, feed velocity with the feed pump operating from a fixed compressed air supply of 6 bar Developing mathematical model and describing the behaviour of NR latex fully during UF was not the scope of the study but it is essential to quantify various factors affecting flux so as to maximise the use of UF to concentrate NR latex

Experimental Design

A schematic diagram of the experimental set-up and a pilot plant is shown in *Figures 3* and *4* The process fluid, which is a highly stabilised NR latex, was fed into the membrane module (PCI Model MICRO 240 FP 110) unit *via* a Sand Piper Model (EB ½ – A Type) diaphragm pump[16] It works on air supply pressure not exceeding 100 p s i (6 89 bar) and was connected to a compressed air supply of six bar The permeate was collected directly onto a digital balance and the reading was recorded in a PC The retentate was recycled into the feed tank The feed flow rate was measured manually as it entered the feed tank The system was

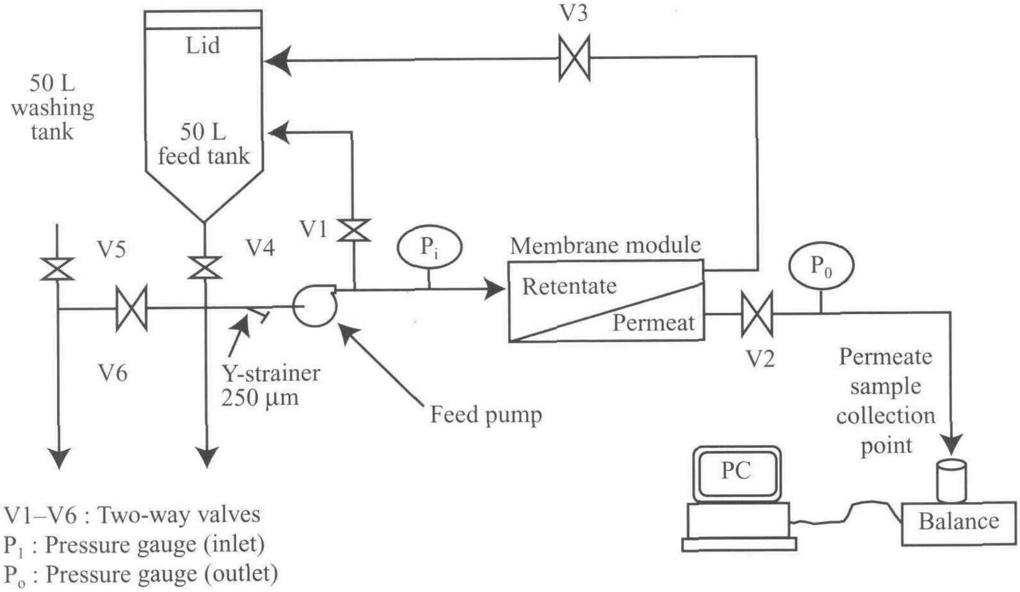

Figure 3. Schematic diagram of the tubular ultrafiltration system.

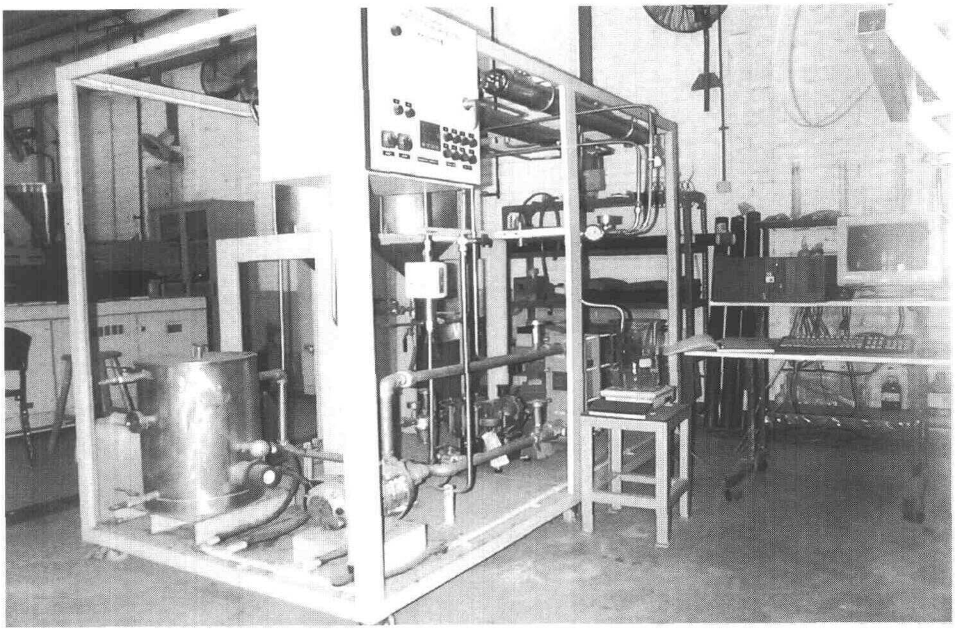

Figure 4. A pilot plant fabricated at the Chemical Engineering Department, University of Malaya where the experiments were conducted.

fitted with two pressure gauges of oil-filled type used to measure the inlet and outlet pressure of the feed as it entered and left the membrane module. The plant was equipped with a 'Y' strainer in the line from the feed tank to feed pump. It is necessary to remove or pre-filter coagulants that were usually found in latex (feed solution) before it entered the membrane module. The unit was equipped with two different tanks: A feed tank of 50 L capacity and a back-wash tank of also 50 L capacity for storing de-ionised (DI) water for cleaning purposes. A two-way valve connected both the tanks. The membrane system consisted of a tubular module (model MICRO 240-FP110) encased in a stainless steel housing obtained from PCI Membrane System Ltd., United Kingdom.

Membrane Characteristics

The membrane used was made of Polyvinylidene Fluoride Type (FP100 PVDF) from PCI Membranes UK, with an apparent retention character of 100 000 MWCO and an effective membrane area of 0.024 m^2.

It consisted of two tubes connected in series by special cap ends with each tube measuring 0.3 meter in length and an internal diameter of 100 mm. It could be ideally used with a pH range of between1.5 – 12. This took care of cleaning solutions of 0.3% nitric acid with a pH of 2 and 0.2% (0.05 M) sodium hydroxide (NaOH) with pH 12. It functioned well with the transmembrane pressure not exceeding 10 bar and temperature below 80°C. The hydrophilicity is rather low which is a disadvantage factor[17].

Membrane storage. When using the membrane for the first time, the preservative chemical was cleaned by running the membrane fitted to the system with 0.3% nitric acid (approximately pH 2). The cleaning process involved running the system with this cleaning solution with a TMP of three bar for about an hour followed by 0.2% (0.05 M) NaOH for 20 min. Membranes in the preservative solution could be stored for up to a year in temperatures ranging from – 4°C to 30°C. Used membranes were cleaned and preserved in 0.25% metabisulphate solution. This solution would be able to preserve the membrane for a month[18].

The membrane must never be left to dry, for it might lead to structural stress that could cause the internal membrane pore structures to collapse. If the membrane was required to be stored in the module, it must be ensured that the membrane was wet and that there was liquid in the module. When left in the system for the following day's run, the membrane module containing the membrane must be packed with DI water by closing the respective valves. For membrane storage over the weekends, it must be removed and submerged in DI water.

Ultrafiltration Run Procedure

Water flux test. Before the start of any experiment, the membrane should be chemically cleaned with 0.2 of sodium hydroxide solution followed by a water flux test. This test was done on an unused membrane and was used as a control and was compared with water flux test done after using the membrane[19]. Variation in water flux of more than 20% indicated that the membrane was fouled[20]. Further cleaning was necessary or the membrane be replaced.

Cleaning of membrane system. After the end of each run, the retentate from the feed tank was drained into a container to be stored. Valves V2, V3, and V4 were fully opened and drainage connection to the 50 L tank was disconnected so that excess latex trapped in the pipes of system was drained out DI water which was stored in the back-wash tank, was immediately let into the system with the feed pump turned on to flush out the system from any remnant latex After 20 min of circulation the feed pump was stopped and the recycled DI water was drained This process was repeated twice with fresh supply of DI water until the recycled water was free of latex As a final cleaning process, 0 2% sodium hydroxide solution was circulated into the system followed by DI water which would be trapped in the membrane system by closing off the necessary valves This was to ensure that the membrane was wet and ready for use the following day

Ultrafiltration Experiments

Experiment UF 1 To identify a suitable composite preservation system (Appendices 1a–d) Once the cleaning and water flux tests were completed, preserved latex was fed into the feed tank after being sieved manually using 200-micron sieve to remove any small pieces of coagulum The feed pump was then turned on to circulate the latex Circulation was maintained for 20 min before settling the system to a pre-selected TMP The system has to be stabilized for about 20 min at the required TMP so that the membrane was fully soaked with the latex and compacted Permeate collection was carried out after a 20 min wait For each TMP the permeate was collected for a predetermined time and was directly collected into a container, which was placed on a digital balance and attached to a PC where the mass was recorded The process was repeated at various values of TMPs and the corresponding permeate mass was recorded in the PC At the beginning and the end of each ultrafiltration run, one liter of sample was removed from the prepared samples and from the retentate, respectively for carrying out DRC, TSC, pH and viscosity test

Experiment UF 2 To study the effects of TMP on feed velocity and permeate flux (Appendix 2) For this experiment, the TMP was fixed for each run Once it was adjusted to the required TMP, series of different feed flow rates (feed velocity) were obtained by adjusting valve V1 (*Figure 3*) with corresponding variation in permeate flux For each TMP, four pairs of feed velocity and permeate flux were obtained Starting with a TMP of two bar, the experiment was repeated for three, four and five bar

Experiment UF 3 To identify optimum TMP for concentration process (Appendices 3a–b) The procedure for this experiment was the same as for *Experiment UF 1* For a predetermined TMP, permeate flux was obtained but before going for the next TMP the process pump was stopped and the latex was drained and the cleaning procedure for the system and membrane was conducted Starting with a TMP of one bar, the whole procedure was repeated for two, three, four and five bar, and corresponding permeate fluxes were obtained An optimum TMP was determined where the permeate flux had the maximum value This TMP value obtained was used in the following experiment

Experiment UF 4 To determine the degree of NR latex concentration (Appendix 4) After performing the necessary cleaning procedure and water flux test, 20 kg of preserved latex (*PS 1*) was fed into the feed tank The feed pump was turned on to circulate the feed Optimum TMP value obtained from *Experiment UF 3* was set The system was left to stabilise for 20 min before commencing the experiment Reading of permeate was taken hourly for seven hours Retentate was drained off into a container after the seventh hour The whole procedure was repeated for the second and third day The concentration process was carried out for a total of 20 h covering a period of 3 days Throughout the duration of the experiment, the TMP was maintained at the optimum value

RESULTS AND DISCUSSION

Suitable Composite Preservation System Between Two Available Options (*Experiment UF 1*)

PS 1 was found to be more effective then *PS 2* Using *PS 2* there were more small pieces of coagulum retained during manual sieving (using 200 μm sieves) During the UF runs the 200 μm 'Y'- strainer attached to the system was blocked *PS 1* was more stable, there were no blockages and little or almost none of the coagulum was retained during sieving The only disadvantage was that the high level of ammonia in the preservation system caused some frothing in the feed tank as the temperature during the day increased

Effect of TMP on Feed Flow Rate (Feed Velocity) and Permeate Flux (*Experiment UF 2*)

High flow rate (feed velocity) caused turbulence and high shear force on the membrane which swept away accumulated solute, reducing the hydraulic resistance of the cake and reducing the thickness of the boundary layer When the system was in the pressure-controlled status, this effect was insignificant as the concentration polarisation and cake built up on the membrane surface was minimal At higher TMP values the effect of velocity on permeates flux was prominent UF carried out at TMP values of three, four and five bar are known to be in the pressure independent regions *Figure 5* shows an increase in feed-flow rate at these TMP values which prominently increased the permeate flux as shown from the slopes of the graphs for feed-flow rate ranging from 180 mL/s to 360 mL/s Therefore, high feed flow rate increased permeat flux in the pressure independent regions

Optimum TMP for NR latex concentration process (*Experiment UF 3*)

From *Figure 6*, it could be seen clearly that at low TMP and low feed concentration, the permeate flux varies linearly with TMP Deviation from these phenomena occurred at high TMP The membrane was chemically cleaned before the flux reading was taken for a new TMP value, so that the value obtained for flux would be free of any gel layer interference from previous filtration Nevertheless, the *Rm* value from *Equation 2* for polymeric membrane (PVDF) was found to increase with increasing TMP and operating time due to membrane compaction[14] This coupled with low feed-flow rate (feed velocity) at high TMP caused the *Rc* value to increase as well As the UF run proceeded, the pressure controlled region was turned into pressure independent region whereby the polarised *Rc* combined with the *Rm* reaching a limiting value coupled with the high pressure acting against the gel layer compacting and blocking the pores of the membrane caused the flux to decrease At three bar the limiting value mentioned in *Methods* was attained

Reduction in flux was thought to occur also due to an increase in solid concentration on the membrane surface that resulted in a higher osmotic pressure, causing a decrease in the driving force ($P_T - \Delta\pi$) and flux Osmotic pressure was generally valid for reverse osmosis of small molecules in solutions but for

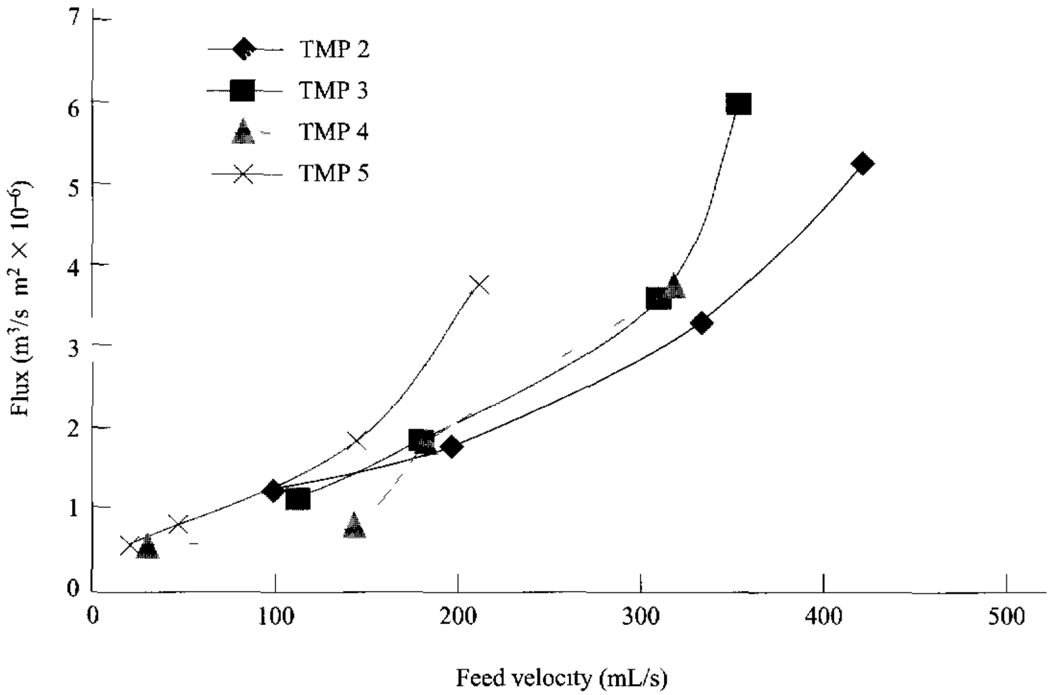

Figure 5 Variations of permeate flux with feed-flow rate (feed velocity) at different TMP

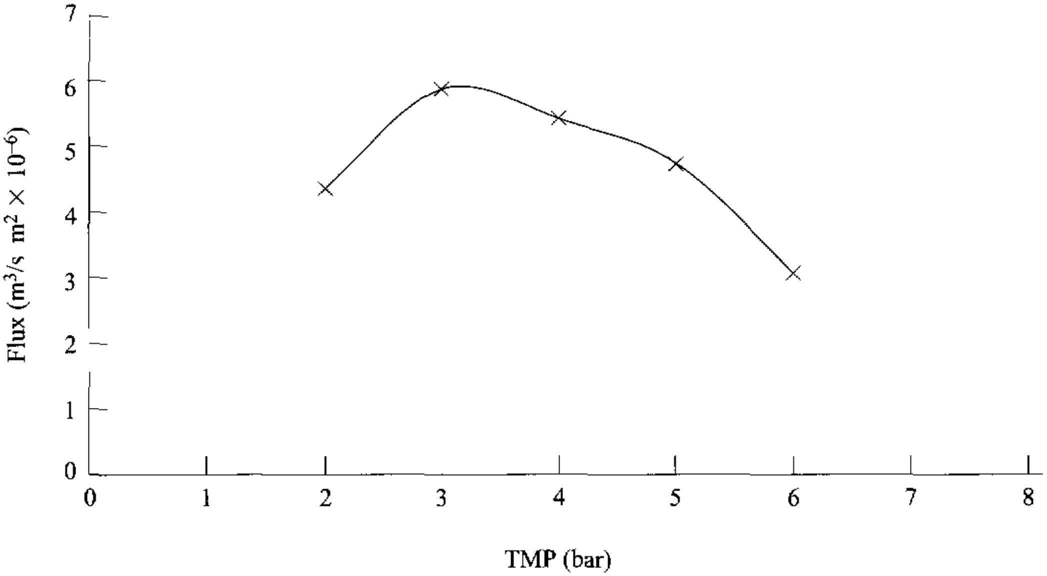

Figure 6 Variation of permeate flux against TMP

UF and microfiltration the effect is still prominent within the polarised layer and could become important if the solute concentration was high enough This was because of the importance of the second and third 'viral coefficients' in the osmotic pressure equation of van't Hoff model

$$\pi = A_1C + A_2C^2 + A_3C^3 + \tag{8}$$

where A_1, A_2 are known as the 'virial coefficients' and C are the concentration of each constituent which make up the NR latex

Increasing TMP beyond 3 bar caused the high TMP to act against the gel layer, making permeation difficult as such the flux decreased Therefore the ideal TMP for concentration should be 2 5 bar below the limiting value of 3 bar

Degree of Concentration Achievable (*Experiment UF 4*)

The concentration process was carried out for three days covering a period of 20 h This was to evaluate the longest period the membrane could withstand fouling and the maximum concentration that could be achieved The rate of decline of the permeate flux was uniform for the first 15 h From the 18th hour onwards it reached a constant value (*Figure 7*)

The concentration process was interrupted by the presence of DI water that was left in the system which could not be drained off completely It diluted the feed when *the run was resumed the following day* Some rubber particles were deposited at the inlet ball valve of the pump The initial and final DRC was 29 61% and 46 09%, respectively The calculation of percentage concentration per square metre per hour is as shown

$$\frac{\text{Final DRC} - \text{initial DRC}}{\text{Membrane area} \times \text{Concentration time}} = \frac{(46\ 09 - 29\ 61)}{0\ 024 \times 20} = 34\ 33\%\ \text{m}^{-2}\ \text{h}^{-1}$$

Assuming the area of membrane to be 0 024 m^2 the increase in DRC value per square metre membrane area per hour (m^2/h) was 34 33%

The rinsing of the membrane with DI water could only remove the gel layer on the surface of the membrane where as the membrane matrix was fouled continuously during the 20 h of UF run The initial flux (J_V) of 6 71 × 10^{-6}(m^3 s^{-1} m^{-2}) dropped to 4 67 × 10^{-6}(m s^{-1} m^{-2}) after an eight hour of UF run The three cycles of DI water rinsing could not improve the flux as the calculated flux value after the first hour of UF run was 4 6 × 10^{-6}(m^3 s^{-1} m^{-2}) when the UF run resumed the following day There was a drop of 31% from the initial flux value after 8 h of UF run After 20 h of UF run the final flux was calculated to be of 0 86 × 10^{-6}(m^3 s^{-1} m^{-2}) which was a drop of 87% from initial flux value

The alternative objective of *Experiment UF 4* was to obtain the worse condition that could lead to membrane fouling The flux that was reduced to about 13% of its initial value was the result of severe fouling of the membrane as washing with DI water could only remove the gel layer on the surface of the membrane The NR latex protein which had a molecular weight distribution ranging from 5 kD to 50 kD coupled with the presence of macro-molecules (100 kD –200 kD) which accounted for *ca* 85% v/v of the total dispersed rubber could have easily blocked up the pores of hydrophobic PVDF[17] membrane wall

The flux reduction was also partly due to the increase in viscosity of the permeate

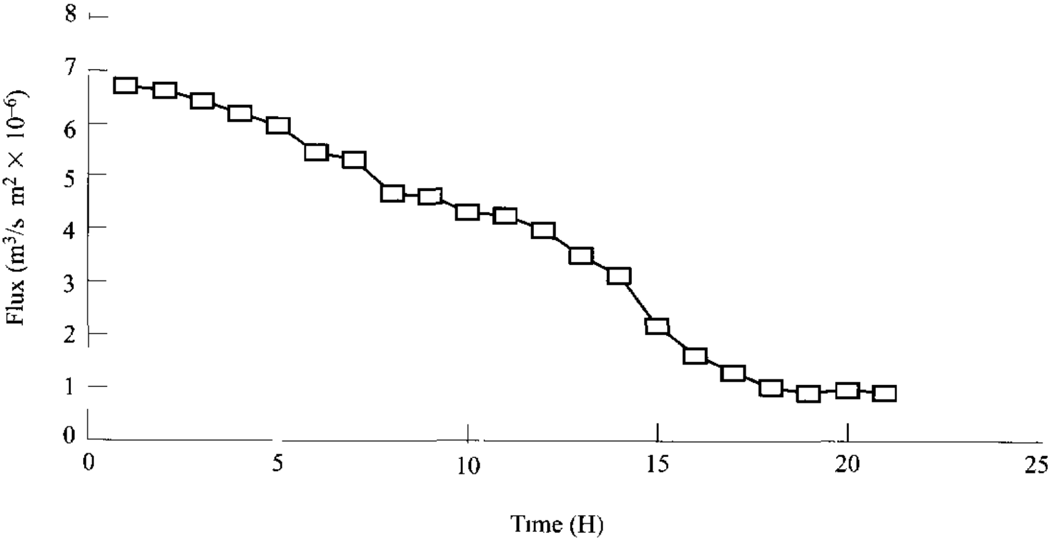

Figure 7 Variations of flux with time during the concentration process

The permeate consisted of water, carbohydrate resins and proteins, and the selectivity varied inversely with molecular mass. Higher molecular-mass and more viscous portions were derived at later stages of the filtration, thus the level of permeate flux dropped.

CONCLUSION

Concentration of NR latex by UF could become a viable option to solve environment-related problems with the current concentration mode of centrifugation. UF could become a method of obtaining a clear serum as permeate which contains mainly uncontaminated quebrachitol and other useful biochemicals[21]. The serum, currently discharged as waste would be worth millions if the entire latex concentrating factories switched to UF. This could pave the way for zero discharge and cleaner production.

For a successful concentration of NR field latex, the tubular ultrafiltration module was the most suitable, as the latex had a high solid content[17]. As the latex particles were negatively charged and highly alkaline in nature with a pH of 10–11, hydrophilic membrane material was a better choice[10]. With a limited choice of membrane at hand, 100 kD PVDF type of membrane was found to be a suitable alternative, although it was rather hydrophobic in nature but it can be utilised at a wide pH range of 1.5–11.

The membrane cleaning procedure, which included 3 rinsing cycles of DI water followed by 0.2% NaOH circulation of cleaning solution with a feed pump for 30 min after every UF run of NRL, could at least restore the flux to 80% of value when the membrane was new. More effective cleaning procedures need to be developed in the future to obtain better flux recovery after membrane cleaning. The membrane left overnight should be packed with DI water instead of cleaning solution to prevent bleeding or rupture of the membrane matrix.

The NR latex preservation system (*PS 1*) [1.0 % ammonia, 0.1% ammonium laurate (from 10% Solution) and 0.025% TMTD/ Zinc Oxide (from 50% dispersion with a ratio of 1:1)], was found to be a better alternative, although the pH was rather high (10.5 – 11.00), but this could arrest the microbial action and keep the latex stable for UF runs. The feed tank should be kept closed so that the ammonia was not released into the factory surroundings. The feed pump should be dismantled after every 20 h of UF run to remove any coagulated rubber attached to the diaphragm or other cavities and the entire pump should be cleaned.

During UF runs, the TMP was at its optimum value of 2.5 bar, to prevent any membrane compaction and also to work at the pressure dependent region to prolong the time taken for polarisation of concentration and gel layer formation to occur. The cross flow velocity should be ideally 250 mL/s–300 mL/s so as to create a turbulent flow that could impart more shearing action against the membrane which could prolong gel layer formation to maintain a good permeate flux. This could hasten the process of concentration.

During the 20 h of UF run, the concentration of NR field latex was increased from its initial DRC of 29.61% to a final DRC of 46.09%. Assuming the area of membrane to be 0.024 m^2, the increase in DRC value per square meter of membrane area per hour (m^2/h) was 34.33%. During the concentration process, the daily UF run lasted for 7 h with a rise in temperature of 16°C. Increasing the membrane area would increase the level of concentration within a shorter period and could possibly reach 60% concentration while taking precaution to control the rise in temperature with a suitable cooling system. Future studies should be focused on this area. A complete mathematical model describing the behaviour of NR field latex during UF or membrane separation during concentration should also be considered.

ACKNOWLEDGEMENT

The authors would like to thank the Director General of Malaysian Rubber Board (MRB) for having granted the permission to carry out this Project at the Chemical Engineering Department, University of Malaya and for permission to publish this paper. The Chemical Engineering Department (University of Malaya) for Vote F Grant, is also acknowledged. The authors also wish to thank and appreciate the following from MRB: Lee Thong Hoe and R. Selvaraj for their assistance during the UF runs, the Material Characterisation Unit, and Dr Nordin Kadir Bakti for his excellent technical guidance.

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APPENDIX 1A
TO IDENTIFY A SUITABLE COMPOSITE PRESERVATION SYSTEM
(*EXPERIMENT* UF 1)

Water flux test for new membrane after preservatives were removed.

TMP (bar)	Calculated permeate flux (J_V) ($m^3/s.\ m^2$) × 10^{-5}
1	1.73
2	2.65
3	5.4
4	6.74
5	7.32
6	8.73

Results of UF using preservation system: Ammonia, ammonium laurate, ZnO and TMTD.

TMP (bar)	Calculated permeate flux (J_V) ($m^3/s.\ m^2$) × 10^{-6}	Cross-flow rate (mL/s)
2.50	2.94	253
3.00	2.90	251
4.00	2.87	197
5.00	1.56	179
5.50	1.29	131

Characteristics of latex sample.

Testing parameters	DRC (% mass)	TSC (% mass)	pH	Viscosity (cPs)
Before UF run	31.73	34.26	10.73	3.73
After UF run	29.57	32.00	10.59	3.65

APPENDIX 1B
TO IDENTIFY A SUITABLE COMPOSITE PRESERVATION SYSTEM
(*EXPERIMENT* UF 1)

Water flux test for membrane which had been used once (1st run) with NR latex and chemically cleaned.

TMP (bar)	Calculated permeate flux (J_V) ($m^3/s.\ m^2$) × 10^{-5}
1	1.83
2	3.45
3	4.78
4	5.88
5	6.68
5.5	7.28

Results of UF using preservation system: Ammonia and Terric®.

TMP (bar)	Calculated permeate flux (J_V) ($m^3/s.\ m^2$) × 10^{-6}	Cross-flow rate (mL/s)
1.00	4.77	320
1.50	4.73	288
2.00	4.52	270
2.50	3.72	266
3.00	3.67	250
3.50	3.18	200
4.00	2.24	158

Characteristics of latex sample.

Testing parameters	DRC (% mass)	TSC (% mass)	pH	Viscosity (cPs)
Before UF run	23.67	25.30	6.54	3.4
After UF run	23.11	24.59	6.13	3.2

APPENDIX 1C
TO IDENTIFY A SUITABLE COMPOSITE PRESERVATION SYSTEM
(*EXPERIMENT* UF 1)

Water flux test for membrane which had been used twice (2nd run)
with NR latex and chemically cleaned.

TMP (bar)	Calculated permeate flux (J_V) ($m^3/s.\ m^2$) × 10^{-5}
1.25	3.75
1.75	4.88
2.25	6.22
3.00	8.06
3.60	8.70
4.00	9.02
4.50	9.20

Results of UF using preservation system: Ammonia, ammonium laurate, ZnO and TMTD
(Repeat of using *PS 1*).

TMP (bar)	Calculated permeate flux (J_V) ($m^3/s.\ m^2$) × 10^{-6}	Cross-flow rate (mL/s)
1.75	4.78	313
2.25	5.23	327
2.50	5.55	341
3.00	5.44	336
3.50	5.07	307
4.00	4.37	295
4.50	2.68	222

Characteristics of latex sample.

Testing parameters	DRC (% mass)	TSC (% mass)	pH	Viscosity (cPs)
Before UF run	31.88	34.52	10.69	3.7
After UF run	29.62	32.06	10.59	3.3

APPENDIX 1D
TO IDENTIFY A SUITABLE COMPOSITE PRESERVATION SYSTEM
(*EXPERIMENT* UF 1)

Water flux test for membrane which had been used thrice (3rd run) with NR latex and chemically cleaned.

TMP (bar)	Calculated permeate flux (J_V) ($m^3/s.\ m^2$) × 10^{-5}
2.00	6.90
2.25	8.35
3.00	11.49
3.60	12.45
4.00	12.87
4.50	13.86
5.00	14.01

Results of UF using preservation system: Ammonia and Terric® (Repeat of using *PS 2*).

TMP (bar)	Calculated permeate flux (J_V) ($m^3/s.\ m^2$) × 10^{-6}	Cross-flow rate (mL/s)
1.75	4.54	308
2.20	4.16	322
3.00	4.34	312
3.50	4.06	310
4.00	3.45	290
4.50	2.91	251
5.00	2.35	218

Characteristics of latex sample.

Testing parameters	DRC (% mass)	TSC (% mass)	pH	Viscosity (cPs)
Before UF run	28.44	30.89	6.95	3.79
After UF run	28.66	30.86	6.49	3.45

APPENDIX 2

TMP ON PERMEATE FLUX AND FEED VELOCITY (CROSS-FLOW RATE)

(*EXPERIMENT* UF 2)

Water flux test for New Membrane after cleaning the preservations.

TMP (bar)	Calculated permeate flux (J_V) (m^3/s. m^2) × 10^{-5}
1.80	2.97
2.75	5.09
3.50	9.75
4.00	10.53
4.50	11.10
5.00	11.80
5.50	12.80

Results of the UF using preservation system: Ammonia, ammonium laurate and TMTD.

TMP (bar)	Calculated permeate flux (J_V) (m^3/s. m^2) × 10^{-6}	Cross-flow rate (mL/s)
2.00	5.29	421
	3.34	333
	1.80	196
	1.24	98
3.00	6.01	353
	3.64	309
	1.86	179
	1.16	112
4.00	6.00	318
	4.00	184
	1.85	142
	79.22	29
5.00	3.76	211
	1.85	144
	80.78	47
	56.59	20

Characteristics of latex sample.

Testing parameters	DRC (% mass)	TSC (% mass)	pH	Viscosity (cPs)
Before UF run	31.54	33.71	10.59	3.90

APPENDIX 3A
TO IDENTIFY OPTIMUM TMP FOR NR LATEX CONCENTRATION PROCESS (*REPLICATE 1*).
(*EXPERIMENT* UF 3)

Water flux test for New Membrane after been cleaned from preservations.

TMP (bar)	Calculated permeate flux (J_V) ($m^3/s. m^2$) × 10^{-5}
1	2.89
2	4.51
3	5.98
4	7.00
5	8.30

Results of the UF using preservation system: Ammonia, ammonium laurate, TMTD and ZnO.

TMP (bar)	Calculated permeate flux (J_V) ($m^3/s. m^2$) × 10^{-6}	Cross-flow rate (mL/s)
1.00	5.52	359
2.00	5.90	335
3.00	5.45	304
4.00	4.75	268
5.00	3.10	230

Characteristics of latex sample.

Testing parameters	DRC (% mass)	TSC (% mass)	pH	Viscosity (cPs)
Before UF run	30.93	33.23	10.59	5.0

APPENDIX 3B

TO IDENTIFY OPTIMUM TMP FOR NR LATEX CONCENTRATION PROCESS (*REPLICATE 2*). (*EXPERIMENT* UF 3)

Water flux test after 1st run with NR latex.

TMP (bar)	Calculated permeate flux (J_V) ($m^3/s.\ m^2$) × 10^{-5}
1	2.77
2	4.35
3	5.67
4	6.59
5	7.80

Results of the UF using preservation system: Ammonia, ammonium laurate and TMTD.

TMP (bar)	Calculated permeate flux (J_V) ($m^3/s.\ m^2$) × 10^{-6}	Cross-flow rate (mL/s)
1.00	7.40	365
2.00	7.64	349
3.00	6.86	330
4.00	4.85	270
5.00	2.63	174

Characteristics of latex sample.

Testing parameters	DRC (% mass)	TSC (% mass)	pH	Viscosity (cPs)
Before UF run	28.77	31.29	10.60	4.7

APPENDIX 4

TO DETERMINE THE DEGREE OF NR LATEX CONCENTRATION (*EXPERIMENT* UF 4)

Water flux new membrane after preservatives were removed.

TMP (bar)	Calculated permeate flux (J_V) ($m^3/s. m^2$) × 10^{-5}
2.00	2.93
2.75	5.00
3.50	9.23
4.00	10.49
4.50	11.09
5.00	10.91

Results of the UF using preservation system: Ammonia, ammonium laurate, TMTDand ZnO
Concentration process–*Experiment UF 4* TMP maintained at 2.5 bar (Day 1)

Time	Temperature °C	Volume of permeate (mL)	Cumulative volume of permeate	Permeate flow rate (mL/s)	Calculated permeate flux (J_V) ($m^3/s. m^2$) × 10^{-6}
9.00 am/0	28	–	–		
10.00 am/1	33	569	569	0.16	6.71
11.00 am/2	37	560	1129	0.16	6.60
12.00 pm/3	40	544	1673	0.15	6.41
1.00 pm/4	41	523	2196	0.15	6.17
2.00 pm/5	42	504	2700	0.14	5.94
3.00 pm/6	43	461	3161	0.13	5.43
4.00 pm/7	44	450	3611	0.13	5.31
4.30 pm/7.5	44	198	3809	0.11	4.67

Characteristics of latex sample.

Testing parameters	DRC (% mass)	TSC (% mass)	pH	Viscosity (cPs)
Before UF run	29.61	31.09	10.41	5.10

APPENDIX 4
TO DETERMINE THE DEGREE OF NR LATEX CONCENTRATION (EXPERIMENT UF 4) *CONT.*

Concentration process – *Experiment UF 4* TMP maintained at 2.5 bar (Continued from day 1)

Time/h	Temperature °C	Volume of permeate (mL)	Cumulative volume of permeate	Permeate flow rate (mL/s)	Calculated permeate flux (J_V) (m^3/s. m^2) × 10^{-6}
9.00 am/7.5	27	–	–		
10.00 am/8.5	34	390	4199	0.11	4.60
11.00 am/9.5	38	364	4563	0.10	4.29
12.00 pm/10.5	40	359	4922	0.10	4.23
1.00 pm/11.5	45	336	5258	0.09	3.96
2.00 pm/12.5	41	295	5553	0.08	3.48
3.00 pm/13.5	42	263	5816	0.07	3.10
4.00 pm/14.5	42	182	5998	0.05	2.15
4.30 pm/15	43	67	6065	0.04	1.58

Concentration process – *Experiment UF 4* TMP Maintained at 2.5 bar (continued from day 2).

Time/h	Temperature °C	Volume of permeate (mL)	Cumulative volume of permeate	Permeate flow rate (mL/s)	Calculated permeate flux (J_V) (m^3/s. m^2) × 10^{-6}
9.00 am/15	27	–	–		
10.00 am/16	33	105	6170	0.03	1.24
11.00 am/17	36	82	6252	0.02	0.97
12.00 pm/18	37	78	6330	0.02	0.92
1.00 pm/19	37	74	6404	0.02	0.87
2.00 pm/20	37	73	6477	0.02	0.86

Characteristics of latex sample.

Testing parameters	DRC (% mass)	TSC (% mass)	pH	Viscosity (cPs)
After UF run (20 h)	46.09	47.86	10.04	8.30