Development of Latex Foam Pillows from Deproteinized Natural Rubber Latex

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Currently there is significant demand for pillows with improved pressure-relief features made from natural materials, alternatives to petrochemical-based foams. In order to meet the requirements, this study's approach is to develop latex foam pillows from deproteinized natural *rubber (DPNR) latex with a unique dual-density cervical-shaped structure. In this work, DPNR latex foam pillows were produced at three different density levels which are high-density, medium-density and low-density. Extractable protein content of latex foam made from DPNR was confirmed lower than latex foam made from low ammonia NR latex (LATZ) and of commercial NR latex foams, making DPNR pillows more hypoallergenic than others. The physical properties of the produced DPNR latex foams were examined in accordance with* Malaysian Standard *MS679, and were found to comply with all requirements stipulated in standard specifications. A novel dual-density cervical-shaped DPNR latex foam pillow prototype was produced where the pillow has lower density at the upper part and higher density at the lower part. Pressure-mapping was used to visualize the pressure distribution patterns and to measure the average peak pressure when a mannequin head was placed on top of the pillow. The study observed that decreasing the density increases the softness of the DPNR latex foam. Softer latex foams led to larger surface contact area, and hence a reduced average peak pressure value. This cervical-shaped structure further increased the surface contact area between the pillow and mannequin head, and thus reduced further the average peak pressure value.*

Keywords: deproteinized natural rubber latex foam; physical properties; pressure-relief; dual density; cervical-shaped pillow

INTRODUCTION

Sleep is important to rejuvenate human physical and mental health, thus indirectly affecting each person's performance capacity[1]. Sleep environments, including mattress and pillow, play a major role in influencing sleep quality[2–4]. According to Lin and Wu[2], to improve sleep quality it is important to reduce sleep-disruptive events such as neck pain, snoring and awakening. Sleeping on a pillow that does not properly support head and neck can create tension in neck muscles, and cause neck and shoulder pain[1–4]. Thus, development of pillows that support the head and neck joints at the correct positions during overnight sleep is an important consideration for researchers and industry alike[5].

High quality "memory foam" pillows have been recommended as therapeutic pillows that could offer better sleep quality[2,6–8]. However, memory foam pillows exhibit shorter lifespans than regular polyurethane foams[6,7]. Both memory foams and regular polyurethane foams are made from petrochemicals, in particular a mixture of isocyanates and polyols, but memory foams are typically more expensive than the regular polyurethane foams due to the additional chemical ingredients required to impart the slow recovery behaviour[9]. According to a previous study[10], isocyanates are a well-known cause of occupational asthma caused by high exposure, at work during manufacturing, or by sensitization. This has raised awareness among users of the possibility that both memory foam and regular polyurethane foams could, over time, release toxic gases which may cause health hazards[10–13]. Further to that, it is well-known that petrochemical-based foam materials contribute to health and environmental issues as well as challenging waste management and disposal problems[6,14–16]. Moreover, with the growing awareness concerning the rising risk of global warming and fossil fuel depletion, as well as new legislation that has been implemented by several countries to encourage the use of "green materials" in product manufacturing, it is both timely and necessary to develop pillows that not only offer pressure-relief features but also that are made from less hazardous materials.

Natural rubber (NR) latex foam is the best alternative material, not only because NR is a natural material, but also because of its intrinsic anti-microbial and dust-mite resistant properties, and has been identified as suitable for people susceptible to asthma[6,17]. NR latex foam pillows are also more durable, exhibit a high degree of elasticity, and better breathability compared to petrochemical-based foam pillows, and are therefore categorized as premium bedding products[18]. Currently there is significant demand for NR latex foam pillows with improved hygiene properties. This is due to the increased prevalence of clinical sensitivity to NR latex products[19,20]. NR latex products contain extractable rubber proteins, which, when in contact with human sweat, can cause latex allergy or *Type I* (immediate) hypersensitivity reactions[21,22]. Latex allergy symptoms can be mild, such as redness of skin and itching, or severe, such as wheezing, mucosal swelling and anaphylactic shock[21,23]. Although there is no evidence reported of NR latex foam pillows causing latex allergies, as is the case with NR latex gloves, the latex allergy issue has raised awareness among users of the potential risks of using other products, including pillows, made from NR latex foam[20,24–26].

Deproteinised natural rubber (DPNR) latex is a purified form of NR latex from which most of the ash and protein components have been removed. This is achieved by treating the NR latex with a proteinase enzyme to hydrolyse proteins in the latex system, which are then eliminated through a concentration process[27]. Previous studies[5,28] found that the resultant end products exhibit reduced levels of extractable proteins as well as being odour-less when compared with normal NR products. However, to date there is limited information concerning the utilization of DPNR in latex foam bedding products. Therefore, the primary objective of the study is to investigate the feasibility of producing pillows from DPNR latex. For the specific purpose of the study, the DPNR latex is prepared directly from freshly tapped NR latex, to differentiate its source from traditional DPNR latex prepared from LATZ. In order to accomplish this objective, compounding, foaming and gelling formulations to produce pillows from the DPNR latex will be developed. The second objective of the study is to evaluate the effect of foam density levels on physical properties, on the morphological structure, and ultimately on the pressure-relief performance of the pillows. Finally, factors influencing pressure-relief performance such as pillow design and structure and softness of the material are discussed. Knowledge gained from this study is important not only to provide methods and formulations to produce DPNR latex foam pillows, but also information on the advantages of using DPNR latex in beddings industry.

EXPERIMENTAL

Materials

In this work, DPNR latex concentrate of 60% total solid content (TSC) prepared from freshly tapped NR latex was used. The method and composition of DPNR latex concentrate was described in Patent Application Number PI2020004246[29]. In brief, the composition to produce DPNR latex comprises of freshly tapped NR latex, proteinase enzyme and non-ionic surfactant. The deproteinization of freshly tapped NR latex was carried out via heat enzymatic hydrolysis reactions in a jacketed reactor. The resultant DPNR latex was then subjected to a centrifugation process to increase the concentration of the DPNR latex concentrate from 30% to 60% TSC. All chemicals used in this work were commercially available, purchased from Alpha Nanotech Sdn. Bhd. For the specific purpose of the study, a commercial grade low ammonia NR latex (LATZ) was purchased from Getahindus (M) Sdn. Bhd as a control.

Compounding and Production Process

There are two options to produce NR latex foam pillows: the Dunlop batch foaming process, and the continuous foaming process[30,31]. In this study, the batch foaming process was chosen because this technique is better suited to small-scale production and research and development purposes, especially when density is being varied[30,32]. Table 1 shows the compounding formulation used in this study, which is the commonly-used sulphur-vulcanized system[31]. All vulcanizing agents were mixed first and allowed to activate at room temperature for 30 minutes.

During the compounding process, potassium oleate (P.O.) was added first into the NR latex, followed by the pre-mixed vulcanizing agents, all while the NR latex was being stirred at 100 rpm. The mixing process was carried out for two hours. After this, the speed of the stirrer was reduced from 100 rpm to 45 rpm and left at room temperature for a further 16 hours to allow efficient maturation of the NR latex compound[30]. The NR latex compound should be continuously stirred to prevent sedimentation of the vulcanizing agents, but at low speed so as to avoid latex destabilization (coagulate) due to shear.

Table 1. Compounding formulation used in this study

Ingredient	TSC (%)	Dry weight (phr)
Latex [a, b]	60	100
Potassium oleate	20	1.50
Sulphur dispersion	60	2.50
Zinc oxide (ZnO) dispersion	60	0.15
Zinc diethyl dithiocarbamate (ZDEC) dispersion	50	0.75
Zinc dibutyl dithiocarbamate (ZDBC) dispersion	50	0.25
Zinc 2-mercaptobenzothiazole (ZMBT) dispersion	50	1.0
Antioxidant dispersion (Wing stay-L)	50	1.0

[a] = LATZ latex; **[b]** = DPNR latex; phr = parts per hundred rubber

Figure 1 shows the different stages in the pillow production process using the Dunlop batch foaming technique. The compounded NR latex is whipped in a Hobart mixer where the stirrer rotates in a planetary motion at high speed (400 rpm) to entrap air into the NR latex compound. It should be noted that DPNR latex is a modified form of NR latex in which the physiochemical properties (i.e. the viscosity and mechanical stability time) of the DPNR latex differ from those of normal NR latex due to the additional chemical treatment. Further to that, in the absence of proteins (which are naturally occurring latex stabilizers), there is the possibility of DPNR latex foam collapsing during the production process[33]. Thus, the study expected to face some difficulties or dissimilarities of foaming behaviour during the latex foam production process. For example, when the density is too low, the foam collapses. Therefore, the study's approach was first to produce DPNR latex foam with high-density (HD), followed by medium-density (MD) and finally, if possible, low-density (LD) latex foam. For the specific purpose of this study, both DPNR latex and LATZ latex foam pillows were produced at three targeted wet density levels: 0.16 g/cm^3 (HD); 0.12 g/cm^3 (MD); and 0.09 g/cm^3 (LD). This was done by controlling the volume expansion of the latex foam during the foaming process. In this work, the volume of the latex foam bowl mixer is 15,000 ml. Therefore, approximately 2400 g, 1800 g and 1400 g of compounded DPNR latex was whipped until the latex foam reached the marked level of the bowl, to produce the HD, MD and LD latex foam respectively.

Figure 1: The different stages involved in the production of NR latex foam pillows using the Dunlop batch foaming process

After the latex foam achieved the targeted wet density level, the gelling ingredients listed in Table 2 were added. It should be noted that sodium silicofluoride (SSF) dosage was varied due to the different types of latex, alkalinity, density levels and the speed of gelation required. As mentioned above, DPNR latex is a modified form of NR latex in which the physicochemical properties (i.e. the viscosity and mechanical stability time) of the DPNR latex are different to those of normal NR latex (LATZ) due to the additional chemical treatment. Therefore, a different quantity of SSF is required for each type of latex. It should also be noted that the SSF hydrolyses into silicic acid and hydrofluoric acid, which gel the NR latex foam[27,34,35]. This presence of hydrofluoric acid causes the gradual drop of pH value. Normally, the gelling process of NR latex foam occurs at pH 8.0 - 8.5[31]. Consequently, 5-6 minutes are required to set the foam cell structures. Therefore, in this work different quantities of SSF were used for different types of latex and density levels to ensure that the latex foams are able to be transferred into the pillow moulds within that period. These quantities were arrived at after a small number of laboratory trials. After the gelling process was completed in an aluminium pillow mould, the mould lid was closed and the NR latex foam was subjected to the vulcanization process in a hot air oven at 100 °C for 60 minutes. After the vulcanization process, the NR latex foam pillow was peeled out of the mould and subjected to washing and drying processes.

Table 2. Description of gel formulation

Ingredient	TSC (%)	Dry weight (phr)		
		HD	MD	LD
NR Latex	60	100	100	100
Diphenyl guanidine (DPG) dispersion	40	0.3	0.3	0.3
Zinc oxide (ZnO) dispersion	60	5	5	5
Sodium silicofluoride (SSF) dispersion	50	0.5[a]/0.8[b]	0.7[a]/1.0[b]	0.8[a]/1.2[b]

[a]LATZ latex; [b]DPNR latex

Determination of Wet and Dry Foam Density

For wet foam density, the latex foam sample was taken out from the bowl mixer during foaming process and poured into a 50 mL beaker to determine the wet density of the latex foam, to ensure each batch of latex foam achieved the targeted latex foam wet density. The wet density was calculated in accordance with Equation 1.

$$\text{Wet density} = \frac{\text{Mass of the sample (g)}}{\text{Volume of the beaker (cm}^3\text{)}} \quad (1)$$

On the other hand, for dry foam density, after the latex foam achieved the targeted latex foam wet density, approximately 250 mL of latex foam was poured into a 250 mL square container. Then, the latex foam sample was subjected to a similar fabrication process. The dry density of the latex foam was determined in accordance with Equation 2.

$$\textbf{Dry density} = \frac{\textbf{Mass of the specimen (g)}}{\textbf{Volume of the specimen (cm}^3\textbf{)}} \quad (\mathbf{2})$$

Pillow Shape and Structure

For the specific purpose of this study, two different pillow shapes were investigated: a standard pillow, and a cervical-shaped pillow. For the cervical-shaped pillow, a novel dual density cervical-shaped DPNR latex foam pillow prototype was produced, where the pillow has a lower density at the upper part and a higher density at the lower part. In this work, the dual density pillow was produced by first pouring the LD latex foam until marked level in the pillow mould, followed by pouring MD latex foam. It should be noted that, the pillow mould is upside down, thereby, a dual density pillow with LD at upper part (curvy-shaped) and MD at bottom part was obtained. The dimensions of the cervical-shaped pillow are shown in Figure 2. In addition, two commercially available pillows, one NR latex foam (CNRL) and one a memory foam (CMF), were purchased from G-Foam Industries Sdn. Bhd. for comparison.

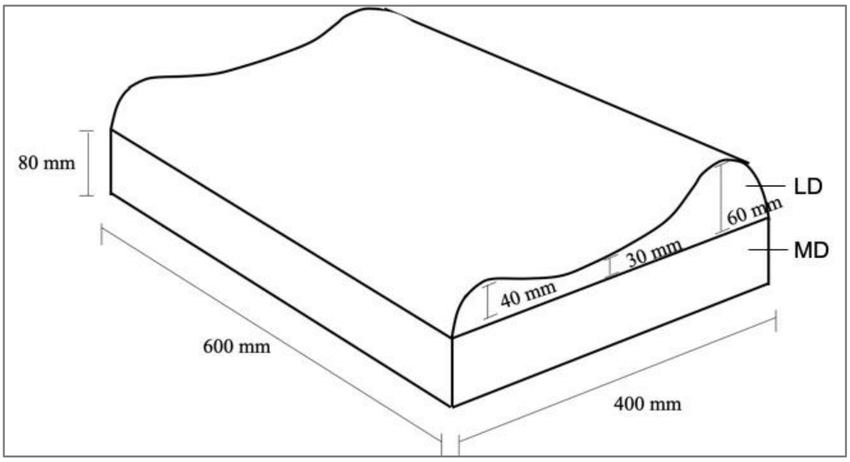


Figure 2: The shape and dimensions of the cervical-shaped dual density NR latex foam pillow, showing the regions of low density (LD) and medium density (MD)

Chemical Analysis

Extractable proteins content

Exposure to extractable rubber proteins (ERP) can caused *Type I* immediate hypersensitivity reactions such as urticaria, facial swelling and asthma[21,22]. Therefore, the ERP content of DPNR, LATZ and CNRL latex foam samples was investigated. The testing procedure was done according to American Society for Testing Materials (ASTM) D5712 standard method. Approximately 5g of latex foam was cut from the sample. The test specimen was extracted in phosphate-buffered saline (0.025 M) (1 g/5 ml) at 25 ºC for 2 hours. Three replicates of each latex foam samples were used to determine level of ERP.

Extractable residual chemicals

Exposure to extractable residual chemicals (ERC) can caused *Type IV* delayed hypersensitivity reactions (48-96 hours after contact) such as vesiculation, erythema, swelling, cracking and itching of the skin at the site of contact[26,36]. Therefore, the ERC content of DPNR, LATZ and CNRL latex foam samples was determined. For the specific purpose of this study, similar compounding ingredients were used to produce both the DPNR and the LATZ latex foams (Table I). The compounding ingredients for the CNRL foam are unknown as this is a commercial product. Three chemicals that were used as vulcanizing agents and are also roots of *Type IV* allergy were examined: zinc diethyl dithiocarbamate (ZDEC); zinc dibutyl dithiocarbamate (ZDBC); and zinc 2-mercaptobenzothiazole (ZMBT). To determine the extractable residual chemicals, 3-5 g of the test specimen were extracted and boiled for one hour in 100 mL of deionized water. The water was further extracted via liquid-liquid extraction by using methylene chloride. The concentrations of the extracted chemicals were individually determined using high performance liquid chromatography (HPLC) by comparing them to reference materials. The limits of detection were 2 µg/g for ZDEC and ZDBC, and 10 µg/g for ZMBT. Three replicates of each latex foam samples were used to determine level of ERC.

Volatile organic compounds

In recent years, concern has been raised about the smell of rubber and its possible health implications due to the emission of volatile organic compounds (VOCs) from foam materials used for bedding products[12,13]. Additionally, odour-less foam materials with reduced emissions of VOCs have become desirable in the bedding industries[37,38]. Unlike ERP and ERC, where allergic reactions can occur when the substances are in contact with skin, VOCs are chemicals that move to the gaseous phase, particularly at higher temperatures. Exposure to VOCs at high concentrations in a closed space may cause health hazards[10–13,39,40]. Although there is no regulatory standard for VOCs, the levels of VOC emission are typically classified into four levels: low, acceptable, marginal and high[41]. Low VOC concentration levels are considered to be less than 300 µg/m^3; acceptable levels of VOC range from 300 to 500 µg/m^3; from 500 to 1000 µg/m^3 of VOC is considered to be marginal; and above 1000 µg/m^3 is classed as high. The VOCs of DPNR, LATZ, CNRL latex foams and of CMF foam were tested in accordance to MS300-55 (2018-06)[42]. Thermal desorption gas chromatography with mass spectrometric detection (TDS-GC/MS) and high-performance liquid chromatography with a diode-array detector (HPLC-DAD) were used to determine VOCs. Each sample was heated at 65 °C for two hours. 3 L sampling bag was used for sampling condition.

Physical Properties

The physical properties of the produced LATZ and DPNR latex foams were measured in accordance to the Malaysian Standard MS 679[43]. This includes density, compression set, indentation hardness, accelerated aging, pounding and elongation at break.

Compression set

Compression set is a static fatigue measurement which measures the degree of irreversible deformation under specific load[18]. Compression set is also often correlated with the degree of vulcanization of NR latex foam[44]. When the compression set value of NR latex foam is greater than 6 %, the NR latex foam is considered under-vulcanized, and thus does not meet the NR latex foam product specifications[43,44]. The compression set value was determined using a compression device consisting of two flat steel plates between which the latex foam specimen was compressed. Each test specimen was compressed to 50% of its original thickness. The compression was maintained for 72 hours at room temperature (approximately 23 °C). At the end of the test period, the test specimen was removed, and the thickness was measured after 30 minutes of rest at room temperature. The compression set was then calculated as

$$\text{Compression set} = \frac{\Delta h}{h_0} \qquad \text{Equation 3}$$

where h_0 is the initial thickness and Δh is the change in thickness.

Pounding effect

The pounding test machine consists of a pounding plate connected to two push rods, one at each side, and held in a horizontal position locking the nuts. The initial thickness (t_0) of the sample was determined. The sample was placed with the cavities face resting on the perforated plate. The pounding machine was adjusted such that the stroke of the compression plate is equal to 20% of the measured thickness of the sample and that, at the top of its stroke, the plate compresses the sample by 40% (and therefore by 60% at the bottom of the stroke). After pounding the sample for 60 seconds at a rate of 4 Hz (240 poundings) the initial indentation hardness index is recorded (H_0). The sample was returned to the pounding machine so that the sample received a total of 250,000 poundings. On completion of the required pounding, the sample is removed and left to recover in an unloaded state for 30 minutes. After this time, the sample is again measured for its thickness (t_1) and indentation hardness index (H_1).

$$\text{Change in hardness index (CHI)} = \frac{H_0 - H_1}{H_0} \qquad \text{Equation 4}$$

$$\text{Change in thickness (CIT)} = \frac{t_0 - t_1}{t_0} \qquad \text{Equation 5}$$

Elongation at break

The elongation at break value is a measure of the physical strength of the latex foam. A higher elongation at break value indicates that a latex foam can be stretched more before breakage occurs. If the elongation at break value of the latex foam does not meet the requirements, this indicates that the material is easy to tear and thus extra care is needed during product handling. The elongation at break of the latex foam was determined using an Instron universal machine. Parallel-sided test samples having cross-sectional dimensions of 10 mm x 12.7 mm and of 150 mm in length were pulled at a constant rate of 500 mm/min until failure of the test specimen. The elongation of the gauge length to the ultimate breaking point of test sample was recorded, and the elongation at break was determined from

$$\text{Elongation at break} = \frac{L_1 - L_0}{L_0} \qquad \text{Equation 6}$$

where L_0 is the undeformed gauge length and L_1 is the gauge length at break.

Indentation hardness

Individual perception of hardness of foam products such as pillows can be very subjective. Therefore, MS679[43] was developed to classify NR latex foam into three different categories which are soft, middle firm, and firm based on an indention hardness (IH) value. The hardness of the latex foam was determined using this indentation test. The indentor foot was brought into contact with the top surface of the test specimen, and the test sample was then indented to 40% of its initial thickness. The corresponding force in Newtons was recorded as the indentation hardness index.

Accelerated ageing

In NR rubber product industries, an ageing test is used to estimate the durability or life span of the NR latex foam products. This is generally achieved by accelerated ageing followed by a measurement of IH value of the NR latex foam after the ageing process. According to MS679[43], the effect of ageing on the IH of NR latex foam should be ±20%. In this test, the test specimen was placed in a heated air oven at a temperature of 70 °C for seven days. After the exposure period, the test specimen was allowed to cool to room temperature and rest for at least 16 hours. The test specimen was then subjected to an indentation test following a similar procedure. The age hardening was then calculated according to the following formulation.

$$\text{Age hardening} = \frac{H_0 - H_\mathrm{a}}{H_0} \qquad \text{Equation 7}$$

where, H_0 is the initial hardness index and H_a is the hardness index after ageing.

Morphological Structure

Scanning electron microscopy (SEM) was used to visualize the morphological structures of the foam samples. A test portion of 5 mm × 5 mm × 5 mm (L × W × H) was cut from the samples and attached onto a specimen stub using a carbon double-sided tape. The specimen was coated with an ultra-thin layer of platinum under a high vacuum evaporation process before visualization under SEM operated at 15kV. The SEM images were captured at 70× magnification. Images obtained from SEM were than analysed using ImageJ software to quantify pore size distribution in the latex foam.

Pressure-relief Performance Evaluation

Pressure mapping technology is a valuable clinical tool that has been used to measure pressure-relief performance of a material as a support material. In this study, a mannequin head and a CONFORMat™ pressure sensor system from Tekscan, USA, were used to investigate the pressure-relief performance of the pillows. The pressure sensor mat was placed first on top of the pillow. Then, a 4.5 kg mannequin head was used to simulate the human head in a reproducible way. The weight of the mannequin head was matched to the average weight of an adult human head[39]. The mat system was calibrated before data collection based on the manufacturer's guideline. The pressure distribution pattern and surface contact area were visualized via a colour map where the maximum pressure appears as a red colour, and the minimum as a dark blue colour.

RESULTS AND DISCUSSION

Chemical Analysis

Extractable proteins content

Figure 3 reports the ERP content for LATZ, DPNR and CNRL foams, and shows that DPNR latex foam exhibits the lowest ERP content followed by CNRL foam and LATZ latex foam. This implies that DPNR latex is a suitable material to produce hypoallergenic pillows without changing the existing NR pillow's production technology. Overall, the ERP contents detected are low. The intensive washing process, where the latex foams underwent hot water washing treatment during production process may have removed most of the water-soluble substances including ERPs.

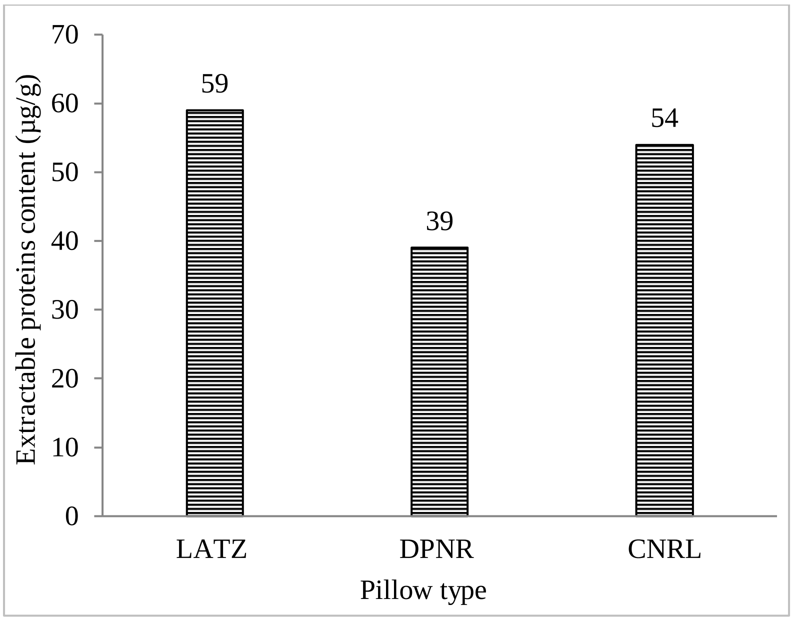


Figure 3: Extractable protein content of latex foam pillow materials

Extractable residual chemicals

Figure 4 reports the ERC for LATZ, DPNR and CNRL foams. ERC of ZDEC and ZDBC content of both DPNR and LATZ latex foams are very low, and almost invisible in the bar chart. CNRL latex foam exhibits a higher extractable ZDEC and ZMBT content compared to DPNR and LATZ latex foam, but no extractable ZDBC was detected.

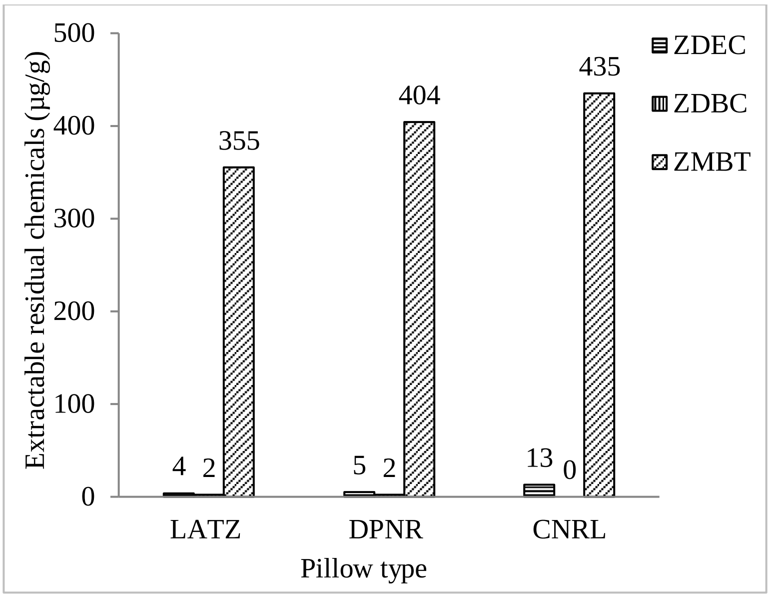


Figure 4: Extractable residual chemicals of NR latex foam pillows

For ZMBT, DPNR latex foam exhibits slightly higher extractable ZMBT residue compared to LATZ latex foam but slightly lower than CNRL latex foam. It should be noted that, there is no standard specification of ERP for NR latex foam products, thus the result shown is for research purpose especially when *Type IV* allergies issue has raised awareness among users of the potential risks of using pillows made from NR latex. The study revealed that, although the washing process may have removed most of the water-soluble substances, there are still remaining quantities of chemicals within the NR latex foam. Nevertheless, the test was conducted in boiling water to force extract as much water-soluble residues as possible, which may not be extracted in real practice. Further to that, pillows are usually covered by two layers of fabric to prevent direct contact between user and pillow. Therefore, the likelihood of the material causing *Type IV* allergies is considered low.

Volatile organic compounds

Figure 5 shows the concentration of VOCs in each sample examined in this study. It is clear from the figure that CMF produces the highest VOC emission, which is classified as high. This could be due to degradation of CMF material that is made from polyurethane. This is in agreement with previous study[45] whereas beddings product made from polyurethane foam is a significant source of VOC emission. Although VOC emission also was observed in all NR latex foam samples, the detected amount is below than 500 µg/m^3 thereby free from health risk[46]. It is well-known that, NR is a natural material obtained from rubber trees. Other than that, the rubber latex contains proteins, higher fatty acids, phospholipids and a very small portion of surfactant which could all contribute to small-volume of VOC, the slightly high-volume of VOC detected on NR latex foam examined in this study could be due to the use of chemicals during the NR latex preparation process, as well as during the NR latex foam production process. Additionally, the use of chemical additives, for example β-sodium naphthalene sulphonate formaldehyde during the preparation of the vulcanizing ingredients (i.e., to change the sulphur from powder form into dispersions) might also contribute to VOCs because during sampling condition, latex foam sample was heated up to 65 °C for two hours. Therefore, further investigation should be conducted to clarify the source of VOCs in NR latex foam samples in future study.

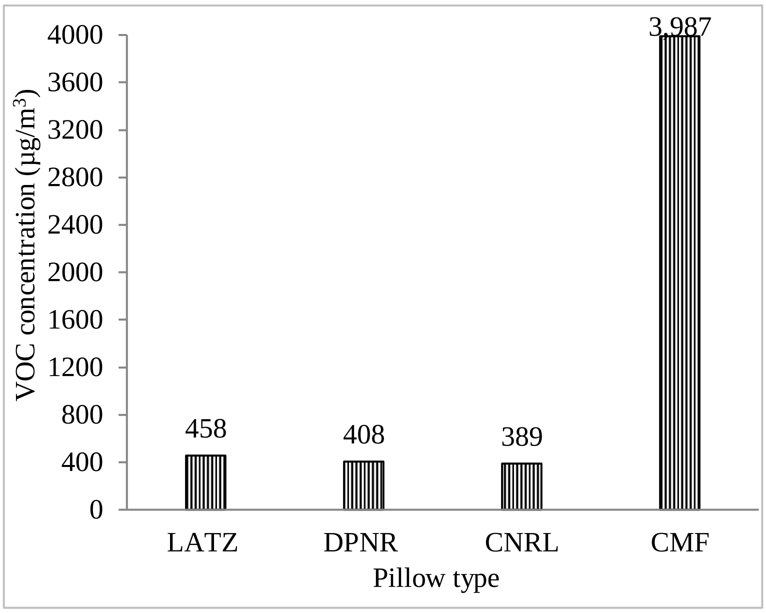


Figure 5: VOC concentrations of NR latex foam and memory foam pillows

Physical Properties

Table 3 shows a summary of the physical properties of all foam materials examined in this study, including samples from the commercial CNRL and CMF pillows. The results shows that the dry density of both DPNR and LATZ latex foams can be manipulated by controlling the wet density of the latex foam during foaming process. Both LATZ and DPNR latex foams obtained a similar density value at each targeted density level. Table 3 also shows that the dry density of both CNRL and CMF was 0.08 g/cm^3, similar to the MD of LATZ and DPNR latex foams produced in this study. The compression set value of all samples examined complied with the requirements stipulated in the standard specifications. Although DPNR (MD) is higher than HD and LD, the difference is not considered significant. Also, the recovery of CMF was observed 100 %. For elongation at break, no significant differences between the elongation at break values of CNRL, LATZ and DPNR latex foam samples was observed, indicating that DPNR latex foam is comparable to CNRL and LATZ latex foam in these terms. The elongation at break of CMF, however, did not meet the standard requirement. This could be due to the soft nature of the material, leading to faster damage of the material compared with NR latex foam[47–49].

Table 3: Physical properties of foam samples examined in this study

Properties	MS679 specifications	LATZ			DPNR			CNRL	CMF
		HD	MD	LD	HD	MD	LD		
Wet density (g/cm^3)		0.16	0.12	0.09	0.16	0.12	0.09	N/A	N/A
Dry density (g/cm^3)	-	0.10	0.08	0.06	0.10	0.08	0.06	0.08	0.08
Compression set (%)	6 (max)	2	2	1	2	3.1	2	2.9	0
Elongation at break (%)	Min 150	270	250	230	260	270	230	270	130
Indentation hardness (N)	<100 (Soft) 101-170 (Middle firm) >170 (Firm)	203	105	89	186	91	70	97	39
Accelerated aging (%)	±20	5	11	16.5	2	-8.5	-13	6.2	50.6
Pounding Change in thickness (%)	5 (max)	0	2.5	2.3	0	2.3	3.7	0	3.3
Pounding Change in hardness index (%)	±20 (max)	-1.2	-3.1	-1.8	-1.5	6.3	6.6	0.2	10.2

N/A = not available

Table 3 clearly shows that decreasing the density levels of both DPNR and LATZ latex foams decreases the IH value. For LATZ latex foam, HD, MD and LD are as categorized firm, middle firm and soft respectively. For DPNR latex foam, HD, MD and LD are categorized firm, soft and soft respectively. IH of DPNR latex foam is lower compared to LATZ latex foam. The reason behind this is unclear but possibly due to removal of proteins that act as non-rubber reinforcing material in NR[50–52]. On the other hand, both CNRL latex foam and CMF foam are categorized as soft foam materials. Table 3 also shows a significant ageing effect on CMF, where the IH value of the CMF increased by more than 50%. This result is in agreement with a previous study[49] where CMF materials were observed to become harder after prolonged use leading to less cushioning support. On the other hand, the pounding test is one of the testing methods to measure the lifespan of NR latex foam products, which is estimated to be equivalent to 10 years of daily (8 hours per day) usage. The comparative study between LATZ and DPNR latex foams found that, at equivalent density levels, LATZ latex foam exhibits slightly better pounding resistance compared to DPNR latex foam, whereas CIT and CHI of LATZ are lower than DPNR latex foam. Although the CHI is observed to change from -3.1 to -1.2 to 1.8 as the foam density goes from HD to MD to LD, these values are all considered to be small and within the experimental uncertainty.

Generally, this study observed that all NR latex foam samples complied to the standard requirements of the accelerated ageing test. However, it is interesting to observe the opposite effect of aging between LATZ and DPNR latex foams, whereas after ageing LATZ shows an increase in hardness whilst DPNR shows a decrease in hardness, although their compounding formulation is the same. The effect of thermal aging of NR vulcanizate has been studied by many researchers[53–59], but the effect of thermal aging on NR latex foam has not been explored. Special methods are needed to investigate this phenomenon which is beyond the scope of this paper. Nevertheless, it is possible to suggest some reasons for this unusual behaviour based on literature. According to Gui Yang and Koening[58], the thermal oxidation will either cause hardening or softening, depending on the microstructure of the diene elastomer. However, the process is very complex and involves several intermediates and side reactions. Azura[57] and Samsuri[60] suggested that the effect of thermal oxidation on physical properties of NR vulcanizate is governed by two competitive factors. One, oxidative and post vulcanization crosslinking reactions which increases hardness. Second, oxidative chain scission reactions and reversion which decreases hardness. Previous studies[54,58,59] stated thermal oxidation of NR leads to main-chain modifications of various types of crosslinks (e.g. poly-sulphidic, di-sulphidic, mono-sulphidic, cyclic sulphides, conjugated dienes and trienes, etc.) Samsuri[60] stated, sulphur-vulcanized NR vulcanizates may hardens before chain scissions take place. This hardening is due to crosslinking associated with oxidative reactions of sulphur species in the molecular network. Hardening is much due to the formation of new crosslinks, thus an overall increases in crosslink density would be expected thereby, would increases the IH of the foam material. Additionally, the increased of IH of the foam material also could be contributed by the presence of non-rubber components (proteins, phospholipids, etc.) which act as filler, as well as residual chemicals (soap, vulcanizing agents, etc.) possibly act as catalysts under aging conditions (70 °C for seven days) used in this work that harden the latex foam material[53,56,60].

On the other hand, the reduction of IH observed on the DPNR foams may well be associated with oxidative scission during ageing. It is well-understood[58,60], that, NR is an unsaturated rubber, subjected to degradation due to the attack of heat, ozone, oxygen and ultraviolet light. It should be noted that, for NR latex foam the scission reactions could occur throughout the foam material for a longer time because NR latex foam contains an additional source of oxygen, the air within the foam-cell structures. Further to that, Lucille[53] stated that proteins play an important role in heat resistance and aging properties of DPNR vulcanizate. Removal of proteins may result in the reduction of thermal-oxidative stability, thus inducing degradation on the physical properties of the DPNR vulcanizate[53,55]. Additionally, Surakit et al.[56] stated that the deproteinization process leads to a reduction of naturally occurring antioxidants (i.e. phospholipids), thus enhancing the deterioration process, which, under the influence of heat, could accelerate the oxidation of rubber chains, thus rendering the material softer. The study also observed a correlation between density levels and ageing effects, where decreasing the density levels increases the accelerated aging value. Therefore, it is suggested that increasing the density levels would improve the durability of the latex foam material.

Morphological Study

The effect of density levels on physical properties of DPNR latex foam was further investigated through scanning electron microscopy (SEM). Figure 6 illustrates that CMF foam exhibits a visibly different foam-cell structure compared to CNRL, DPNR and LATZ latex foam. CMF appears to have elongated pores which are less circular than those observed in the other CNRL, DPNR and LATZ latex foams. CNRL latex foam exhibits a more uniform pore size compared to DPNR and LATZ latex foam. This could be due to different foaming processes used between CNRL latex foam and DPNR and LATZ latex foams. The CNRL latex foam was produced using a commercial continuous foaming process in which air is metered under controlled pressure into an Oakes foaming head. On the other hand, DPNR and LATZ latex foams were produced using a batch foaming process where air is introduced through whipping the latex in a Hobart mixer. For DPNR and LATZ latex foams, it can be observed that the pore size of HD latex foam is smaller compared to that of MD and LD.

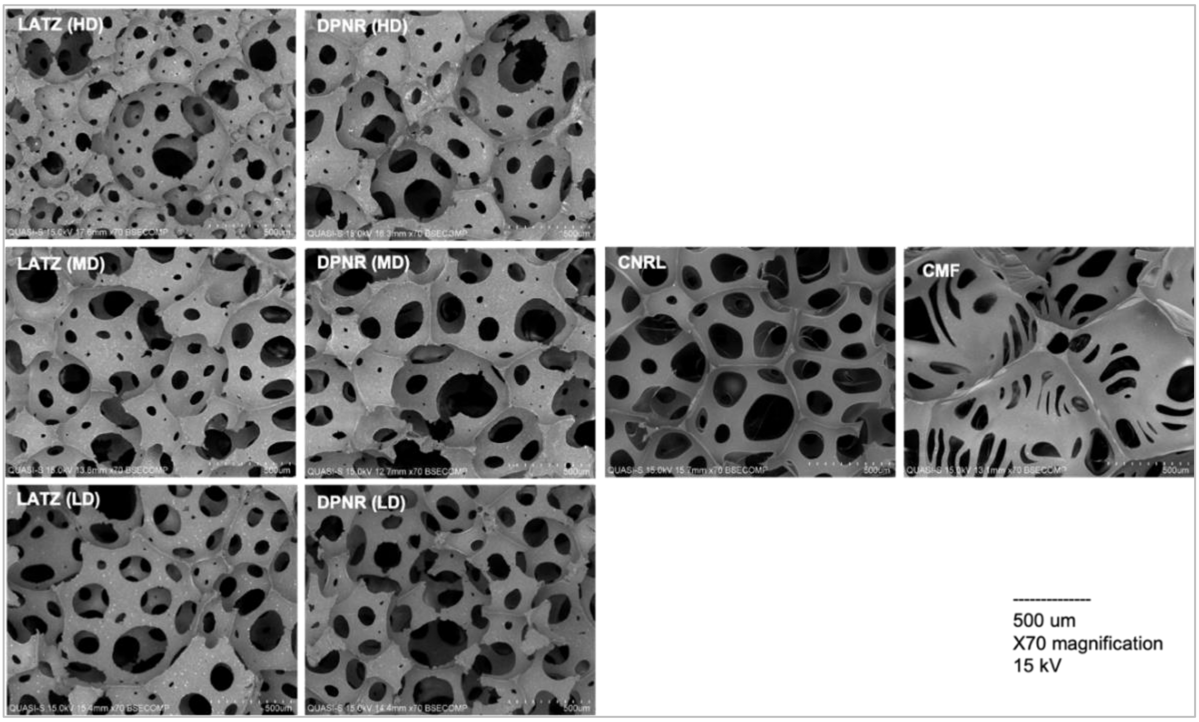


Figure 6: Morphological structures of LATZ, DPNR, CNRL latex foam and CMF foam

To further investigate the effect of density on pores size of the latex foams, ImageJ software was used to quantify the pore size distribution from the images. Figure 7 shows the distribution of pores size of LATZ, DPNR, CNRL latex foams and CMF foam.

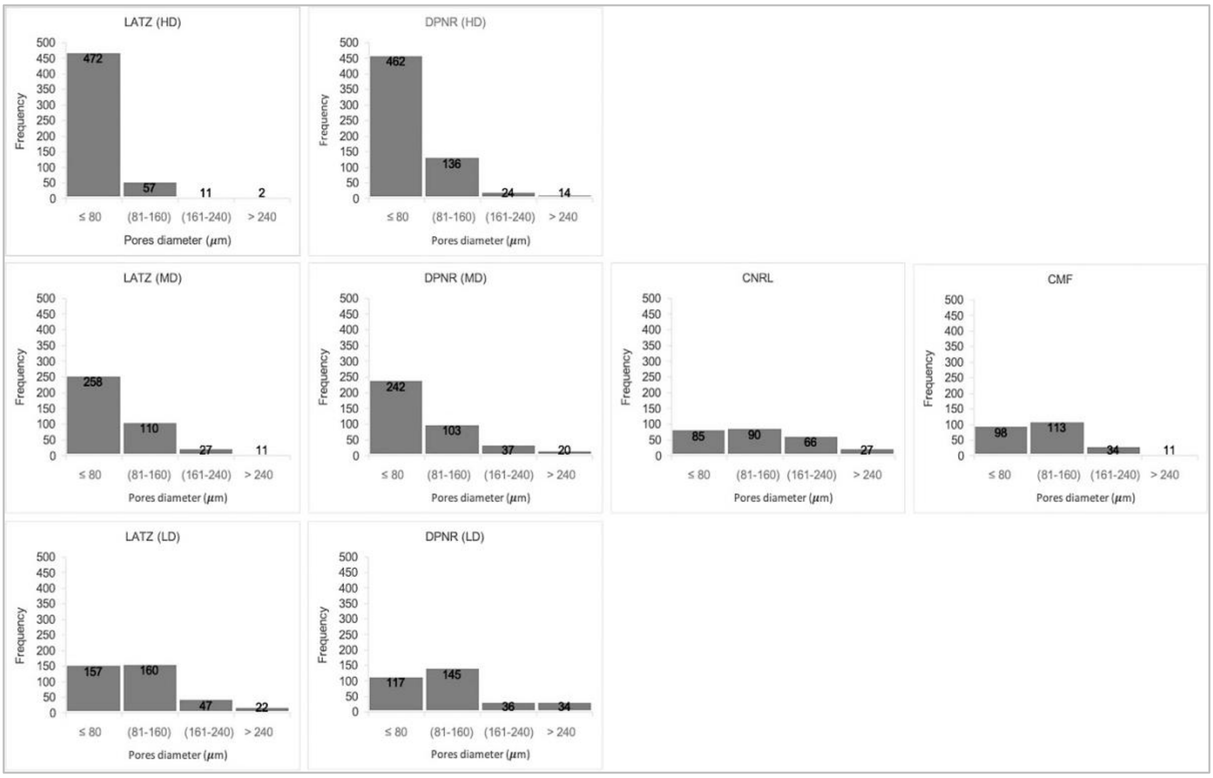


Figure 7: Distribution of pore size of the foam materials

It is clear that, the dominant pore size is 80 µm and below in the HD foam of both LATZ and DPNR latex foam. Decreasing the density from HD to MD and to LD has the effect of shifting the distribution to larger pore sizes. For CNRL, the fraction of pore size of 80 µm and below is similar to that of pore size of 80-160 µm and of pore size of more than 160 µm. For CMF, the fraction of pore size of 80 µm and below is slightly lower than that of pore size of more than 80 µm.

Figure 8 shows the mean pore size of of LATZ, DPNR, CNRL latex foams and of CMF foam. LATZ (HD) demonstrates the smallest mean pore size compared to the other foam type examined in this study. Figure 8 also shows that perhaps unsurprisingly, decreasing the density levels of both LATZ and DPNR latex foam from HD to MD and to LD leads to an increase in the mean pores size of the latex foams. The study also observed that although CMF foam, CNRL, LATZ (MD) and DPNR (MD) all have a similar density level (0.08 g/cm^3), they exhibit different mean pore sizes. This could be due to different processes of production between each foam. Technically CMF foam is a different material compared to the other foams, and is produced by chemical reactions between polyols and isocyanates. Thus the morphological characteristics, including the pore size of CMF, are expected to be different to the other foams. On the other hand, CNRL, LATZ (MD) and DPNR (MD) latex foam are basically NR latex, but the foaming mechanism between CNRL latex foam and LATZ (MD) and DPNR (MD) latex foam is different. The effects of the foaming mechanism on morphological characteristics of NR latex foam have been described in previous studies[35,47,61].

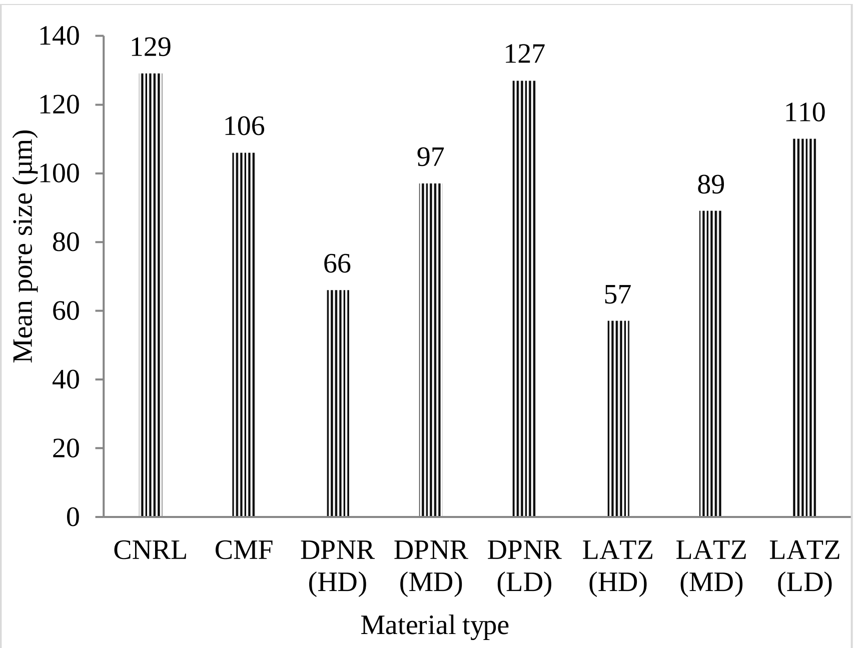
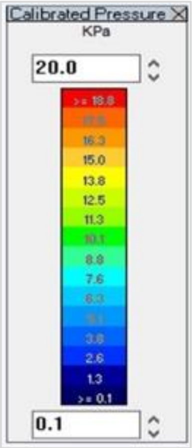
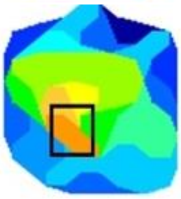


Figure 8: Mean pore size of foam materials examined in this study

Pressure-relief Performance

Table 4 shows snapshots from real-time measurements of pressure distribution, surface contact area and average peak pressure values of LATZ and DPNR latex foam pillows when a mannequin head was placed on the top of the pillow.

Table 4: Snapshots of pressure distribution, average peak pressure value and surface contact area arising from a 4.5 kg mannequin head load, comparing LATZ and DPNR foams

Type	Pressure distribution*	Surface contact area (mm^2)	Average peak pressure value (kPa)	Pressure scale
LATZ (HD)		5209	15.6	
LATZ (MD)		6511	14.2	
LATZ (LD)		7464	11.0	
DPNR (HD)		5860	15.4	
DPNR (MD)		6729	13.8	
DPNR (LD)		7596	10.9	

*Black square box located the average peak pressure

It should be noted that, peak pressure is the peak interface pressure or physical loading between mannequin head and the foam material. Previous studies[62,63] stated that, such physical loading caused tissue deformation at cellular level and thus prevents arterial vessels from resupplying tissues with oxygen leading to ischemia (disrupts of blood circulation). Therefore, it is important to optimize the interface pressure distributions. In this work, irregular colour intensities and contours of the mannequin head in each type of pillow, shown as a red box in Figure 9 were observed. The results indicate dissimilar physical responses of the pillows to the loading of the mannequin head. This implies that the physical properties of the pillows affect the pressure distribution pattern. Table 4 also demonstrates that decreasing the density levels from high-density (HD) to medium-density (MD) and low-density (LD) increases the surface contact area value. Comparison between LATZ and DPNR latex foam pillows indicates that, at similar density levels, DPNR latex foam pillows produce a larger surface contact area than LATZ latex foam pillows.

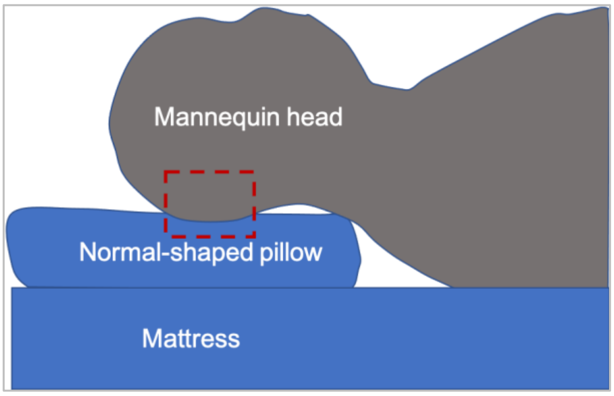


Figure 9: Modelling pillow conforming area under the head

The results also indicated there is an inter-relationship between surface contact area and average peak pressure value, illustrated in Figure 10. It can be seen in the graph that increasing the surface contact area decreases the average peak pressure value. Empirically, there is evidence linking material with higher surface contact area able to optimize interface pressure distribution, hence reduce the average peak pressure value. It is clear that, LD pillows of both DPNR and LATZ latex foam produce the highest surface contact area followed by MD and HD pillows, and thus LD pillows exhibit the lowest average peak pressure value followed by MD and HD pillows. This is directly proportional to hardness of the material whereas, LD pillows exhibit low IH value. Therefore, it can be concluded that density levels and hardness of both LATZ and DPNR latex foam play the important role in the capability of the material to improve pressure-relief performance of the pillows.

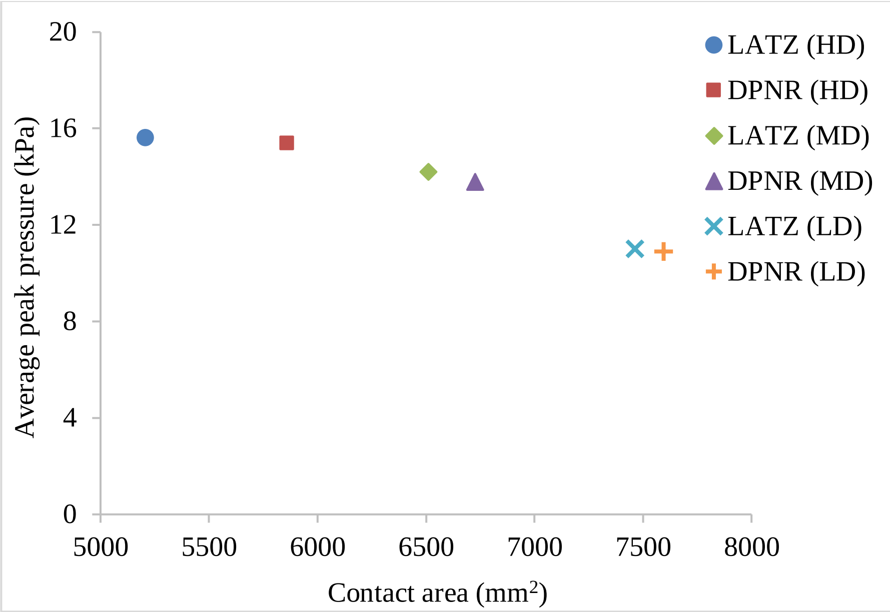


Figure 10: Relationship between surface contact area and average peak pressure value

The dependence of pressure-relief performance on surface contact area and average peak pressure of a pillow was investigated. Table 5 shows pressure-relief performance of normal-shaped and cervical-shaped pillows. The cervical-shaped dual-density DPNR latex foam pillow exhibits the highest surface contact area followed by the cervical-shaped CMF foam pillow, cervical-shaped DPNR (MD+LD) latex foam pillow, cervical-shaped CNRL latex foam pillow, cervical-shaped DPNR (MD) latex foam pillow and normal-shaped DPNR latex foam pillow. A clear increase of surface contact area at the region under the neck, shown as a red box in Figure 11, was observed. The results indicate that an increase in surface contact area at the region under the neck leads to a decrease in the average peak pressure value. This finding suggests that cervical-shaped pillows could provide a better neck support and pressure-relief performance compared to normal-shaped pillows.

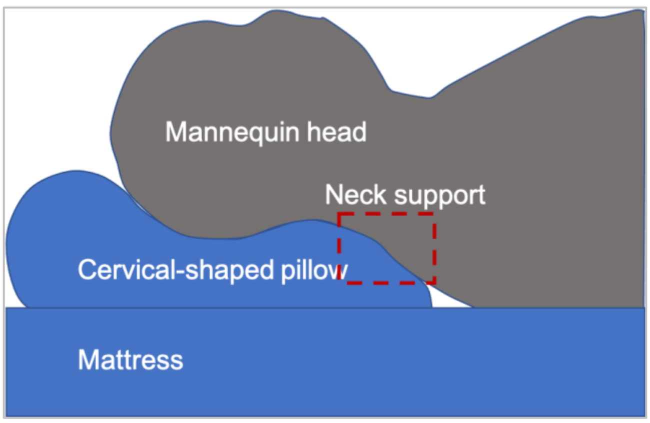
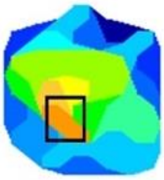
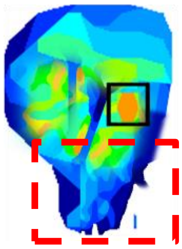
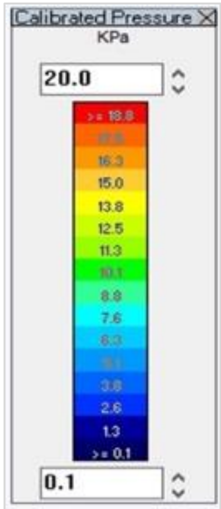


Figure 11: Modelling pillow conforming area under the neck

Table 5: Snapshots of pressure distribution, surface contact area and average peak pressure values arising from a 4.5 kg mannequin head load, comparing different pillow shapes and materials examined in this study

Type	Pressure distribution*	Surface contact area (mm^2)	Average peak pressure value (kPa)	Pressure scale
Normal-shaped DPNR latex foam pillow (MD)		6729	13.8	
Cervical-shaped DPNR latex foam pillow (MD)		7901	10.4	
Cervical-shaped DPNR latex foam pillow (MD+LD)		8681	9.2	
Cervical-shaped CNRL latex foam pillow (MD)		8064	10.2	
Cervical-shaped CMF foam pillow (MD)		8549	9.3	

*Black square box located the average peak pressure; the red rectangular box indicates the region under the neck

This is in agreement with previous studies[2,63,64] where the ability of cervical-shaped pillows to contour the neck's natural curve have been shown to offer a better support to the cervical spine and restore cervical lordosis, and therefore, provide relief to neck and shoulder pain. Comparisons between the cervical-shaped dual-density DPNR pillow with other pillows, indicate that the average peak pressure value of the dual density cervical-shaped DPNR latex foam pillow is similar to that of the cervical-shaped CMF pillow. The excellent pressure-relief performance of CMF foam pillows is expected due to its low hardness and has been observed elsewhere[2,6–8]. This study has shown a cervical-shaped dual-density DPNR pillow also capable to offer excellent pressure-relief performance as CMF foam pillow do. The LD foam part at the upper part of the pillow is expected to provide extra comfort to users, and it is encouraging that the pillow exhibits the lowest average peak pressure value. On the other hand, the MD foam at the lower part of the pillow is expected to provide a firmer feeling and superior neck support. It should be noted that LD foam is soft, and thus, when the pillow is overloaded, the foam will be flattened (bottoming out), and thus will no longer be able to provide its important pressure distribution feature, something that has been observed before in some CMF foam pillows[47–49]. The bottoming out issue not only affects pressure-relief performance but also reduces its durability over time. Therefore, the combination of LD and MD foam, namely dual-density DPNR foam, is designed not only to offer a good pressure-relief performance but also address the bottoming out issue, as well as to be more durable than single density (LD) pillow. This dual density/multiple density foam combination approach has already been applied in mattresses[65–67] to provide uniform and enhanced body support and extra comfort to users, but to the authors' knowledge has not been applied to pillows. This present study attempts to provide some initial information, but due to the limitations of time and materials, there is no conclusive data to support the view that dual density DPNR latex foam pillows could enhance the pressure-relief performance as well as increased durability, something which should be the focal point of a future study.

CONCLUSION

This study has shown that DPNR latex can be used to produce hypoallergenic latex foam pillows in terms of low ERP content compared to normal NR latex, without changing the existing NR pillow's production technology. It was found that DPNR latex foam exhibits open-cell foam structures, and decreasing the density levels from HD to MD and to LD leads to an increase in the mean pore size of the DPNR latex foam. Physical properties of DPNR latex foam were shown to be influenced by the density of the latex foam, where the lower the density the softer the latex foam. Also, there is a correlation between density levels and ageing effects, where decreasing the density levels increases the effect of accelerated aging. This study also found that lower density foams exhibit lower average peak pressure values when subjected to the loading of a mannequin head. There is an inter-relationship between surface contact area and average peak pressure value where high surface contact area leads to lower average peak pressure values. It was also shown that the surface contact area of a cervical-shaped pillow is higher than that of a normal-shaped pillow. Thus, a cervical-shaped pillow is expected to offer better pressure-relief performance compared to a normal-shaped pillow.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest regarding the publication of this paper.

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