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RESEARCH ARTICLE

STUDY ON THE FOAMABILITY, MORPHOLOGICAL STRUCTURE AND PRESSURE-RELIEF PERFORMANCE OF AMMONIA-FREE NATURAL RUBBER LATEX FOAM FOR SHOE INSOLES APPLICATIONS

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Abstract. Ammonia-free natural rubber (AFNR) latex is a newly developed commercial grade of natural rubber (NR) latex concentrate aimed to address environmental and health concerns related to ammonia in the NR latex industry. This study investigates the viability of using AFNR latex to produce latex foam shoe insoles and compares it with that of traditional high-ammonia NR (HANR) latex. The Dunlop foaming process was used to produce both types of latex foams, each with a wet foam density of 0.2 gcm^{-3}. The study assessed the effect addition of varying levels (0, 0.5, 1.0 and 2.0 phr) of potassium oleate, a foaming agent, on the foamability of the latex. The findings indicate that AFNR latex exhibits lower foamability compared to HANR latex, although increasing the potassium oleate level enhances the foamability of AFNR latex. Additionally, the results show that AFNR latex foam requires a longer time to get a gel during the gelling process compared to HANR latex. A cross-sectional view using FESEM reveals that both AFNR and HANR latex foams exhibit open-cell structures. FESEM images also show smoother, thicker skin at the bottom compared to the upper part of both samples, with AFNR having 60 µm at the bottom and 38 µm at the upper and HANR having 52 µm at the bottom and 37 µm at the upper. The difference in skin structure contributes to the variation in Shore F hardness values but does not show a significant difference in pressure-relief performance. Despite differences in foamability and skin structure, AFNR latex shows comparable pressure-relief performance to HANR latex, indicating its potential for application in latex foam shoe insoles.

Keywords: Latex foam shoe insoles, morphological structure, physical properties.

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1. INTRODUCTION

Shoe insoles is the supporting layer that serves as the point of contact between the foot and the shoes as well as the ground [1,2]. It provides immense comfort and pain relief, particularly in the heel and plantar areas of the foot. Previous studies [3,4] have indicated that prolonged standing (physical loading) can cause tissue deformation at the cellular level, which hinders arterial vessels from supplying oxygen to the tissues. This can lead to ischemia (disruptions in blood circulation), resulting in foot pain and ulcers, especially for individuals with diabetes. Thus, it is essential to use shoe insoles that offer effective pressure relief performance. Additionally, shoe insoles offer superior support and shock absorption, enhancing functionality and performance and preventing injuries during daily work and sports activities [5,6].

It was reported that the shoe insoles market was valued at approximately USD 5,836.8 million in 2023 and is expected to raise to USD 6,122.8 million by 2024 [7]. The same report also projects that by 2034, the market will reach USD 9,508.5 million, with a compound annual growth rate (CAGR) of 4.5 %. Ethylene vinyl acetate, polyurethane foam, cork, thermoplastic polyurethane, textile fabrics and rubber are among the common materials used to produce shoe insoles [8,9]. Each material used for shoe insoles has its own advantages and disadvantages.

Natural rubber (NR) latex is a natural and sustainable material obtained from tapping rubber trees, known as *Hevea brasiliensis*. There are two types of NR latex: high-ammonia NR (HANR) latex and low-ammonia NR (LATZ) latex, both of which are used in the production of latex foam products, including shoe insoles, due to their exceptional elasticity, durability and resilience [10,11]. The NR latex foam shoe insoles offer extraordinary comfort, excellent rebound resilience and remarkable support across various body weights, making them a favored choice for a wide range of footwear, especially sports and casual shoes [12].

However, both HANR latex and LATZ latex contain ammonia as a preservative to prevent coagulation of the latex during storage and processing [13]. The drawback of ammonia in the latex system is that it can destabilize the foam cell structure during latex foam manufacturing, resulting in production issues such as foam collapse, surface defects and rat holes [14]. Approximately 5 to 10 percent of the production is found to have defects and is therefore discarded or sold as lower-grade products [15]. This percentage represents a significant loss and indicates challenges that must be addressed to reduce waste and enhance overall product quality. Moreover, the use of ammonia has also been associated with environmental and health concerns, including skin irritation for workers, unpleasant odors, challenges in effluent treatment and building corrosion [13].

Recently, a new grade of NR latex known as ammonia-free NR (AFNR) latex has been developed. The availability of AFNR latex provides a safer manufacturing environment and an opportunity to produce latex-based products with improved quality. Therefore, the study investigates the viability of using AFNR latex as an alternative type of NR latex for producing latex foam shoe insoles. One of the objectives of the study is to evaluate the foamability of AFNR latex, along with the effect of adding potassium oleate on its foaming behavior. Potassium oleate is an unsaturated fatty acid serves as both a stabilizer and a foaming agent in the latex foam manufacturing process [10]. Therefore, in this study, the effect of different levels of potassium oleate loading (0, 0.5, 1.0 and 2.0 phr) in AFNR latex foam is evaluated to optimize foamability and manufacturing process. Additionally, the relationship between the morphological structure, hardness and pressure-relief performance of the manufactured shoe insoles is a key aspect of the study. In this study, the AFNR latex foam will be compared with the conventional HANR latex, emphasizing the differences and similarities between the two materials.

2. MATERIALS AND METHODS

2.1 Materials

In this study, all chemicals involved in the production of latex foam shoe insoles were purchased from Tiarco Chemical (M) Sdn. Bhd., Perak, Malaysia and were used as received. These chemicals include a 20 % solution of potassium oleate, a 60 % dispersion of sulfur, a 50 % dispersion of zinc 2-mercaptobenzothiazole, a 50 % dispersion of zinc diethyl dithiocarbamate, a 50 % dispersion of antioxidant, a 40 % dispersion of diphenyl guanidine, a 60 % dispersion of zinc oxide and a 50 % dispersion of sodium silicofluoride. On the other hand, commercial-grade AFNR latex and HANR latex were purchased from Getahindus Sdn. Bhd., Selangor, Malaysia. Table 1 shows the physicochemical properties of the latex. AFNR latex and HANR latex exhibit slightly different properties. HANR latex has a total solid content (TSC) of 61.78 % and meets the dry rubber content (DRC) minimum at 60.15 %, while AFNR latex has a TSC of 60.64 % and a DRC of 58.55 %, both below minimum standards. AFNR latex offers better mechanical stability (MST), with a stability time of 2390 seconds compared to 1620 seconds for HANR latex. Both types of latex meet the standard requirements for volatile fatty acid numbers and viscosity values.

Table 1: Comparison of physicochemical properties of AFNR and HANR latex

Properties	ISO 2004:2017	AFNR latex	HANR latex
Total solid content (%)	61.00 (minimum)	60.64	61.78
Dry rubber content (%)	60.00 (minimum)	58.55	60.15
Mechanical stability time (sec)	650 (minimum)	2390	1620
Volatile fatty acid number	0.060 (maximum)	0.022	0.024
Viscosity (cP)	150 (maximum)	65	58

2.2 Preparation of Latex Foam Shoe Insoles

The production of both AFNR latex foam and HANR latex foam shoe insoles are in accordance with the conventional Dunlop batch foaming process [4], as depicted in Figure 1. The process encompasses compounding, foaming, gelling, molding, vulcanizing, de-molding, washing, rinsing and drying. The formulations for compounding and gelling are detailed in Tables 2 and 3, respectively. The latex was first sieved through an 80-mesh stainless steel to eliminate any coagulated latex. The latex was then stirred at 110 rpm and 1.0 phr of potassium oleate was added as a stabilizer. Following this, the premix vulcanizing agents were incorporated and stirring continued for one hour.

Once the latex foam reached the desired density level (0.20 gcm^{-3}) the gelling ingredients listed in Table 3 were added to initiate the gelling process and stabilize the foam cell structure. It is important to note that, while sodium silicofluoride is typically used at 50 % TSC, this study utilized a 20 % TSC dilution to prevent sudden gelation of the latex foam. Once the gelling ingredients were incorporated, the latex foam was promptly poured into the shoe insole molds.

The latex foam was then allowed to gel at room temperature for 10 minutes. Then, the mold was covered with its lid. Following this, the latex foam shoe insoles were vulcanized in a hot air oven at 100 °C for one hour. After vulcanization, the insoles were removed from the mold. The insoles were then washed by soaking them in hot water at 60 °C for five minutes, rinsing with tap water and finally squeezing out the water using squeeze rollers. Finally, the latex foam shoe insoles were dried using a tumbler. All samples were kept dry at room temperature for 16 hours before being subjected to testing.

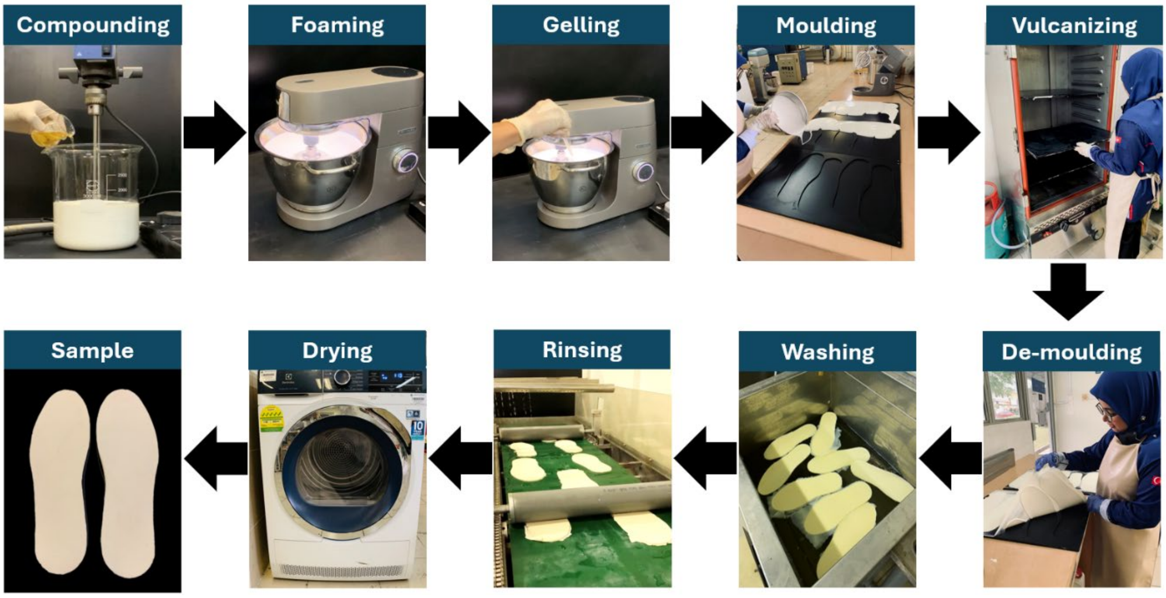


Figure 1: Stages involved in the production of latex foam shoe insoles

Table 2: Compounding formulation

Ingredient	TSC (%)	Dry weight (phr)	Wet weight(g)
NR latex	60	100	1000
Potassium oleate	20	1.0	30
Sulphur dispersion	60	2.5	25
Zinc diethyl dithiocarbamate dispersion	50	1.0	12
Zinc 2-mercaptobenzothiazole dispersion	50	1.0	12
Antioxidant dispersion	50	1.0	12

phr = parts per hundred rubbers

Table 3: Gelling formulation

Ingredient	TSC (%)	Dry weight (phr)	Wet weight (g)
NR latex	60	100	1000
Potassium oleate	20	0,0.5,1.0,2.0	0,15,30,60
Diphenyl guanidine dispersion	40	1.0	15
Zinc oxide dispersion	60	5.0	50
Sodium silicofluoride dispersion	50	2.0	24

phr = parts per hundred rubbers

Then, the stirrer speed was reduced to 50 rpm and the latex was left to mature at room temperature for 16 hours. After maturation, the compounded latex was sieved again using an 80-mesh stainless steel sieve to remove any coagulated latex and unreacted residual chemicals. The second step in producing latex foam shoe insoles involves foaming the latex to achieve a targeted wet latex foam density of 0.20 gcm^{-3}[16]. For this process, a Kenwood mixer model KM800 was used. Approximately 1000 g of latex was poured into the bowl mixer and the latex was whipped at speed number 4 (170 rpm) until the foam reached the 5 L mark on the side of the bowl mixer. This study evaluated the effect of varying levels of potassium oleate (0, 0.5, 1.0 and 2.0 phr), a foaming agent, on the foamability of the latex. A 5-minute timeframe was set for the latex to foam to the 5 L volume. The time taken for the foam to reach the targeted volume was recorded. If the latex foam did not reach the 5 L volume within 5 minutes, the volume of the foam at the end of the 5-minute period was also recorded.

2.3 Morphological Visualization

Scanning electron microscopy (SEM) (JEOL JSM-7610F Plus) was used to visualize the morphological structure of the latex foam samples. A test portion of 3 mm × 3 mm × 3 mm (L × W × H) was cut from the samples and attached to a specimen stub using carbon double-sided tape. The SEM was operated at an accelerating voltage of 1.0 kV and at a variable working distance, utilizing a secondary electron image (SEI) secondary detector. The SEM images were captured at 70× magnification. Three areas of the latex foam samples were visualized: the cross-section, the upper part and the bottom part. The resulting images were then analyzed using ImageJ software.

2.4 Hardness Test

A Shore F Durometer hardness tester was used to evaluate the hardness of the latex foam shoe insoles in accordance with ASTM D 2240 [17]. A minimum sample size of 100 mm × 100 mm × 20 mm (width, length, thickness) was utilized for the measurements. Hardness readings were taken at five distinct locations on each specimen, ensuring that each measurement point was spaced at least 6.0 mm apart. The highest and lowest readings from these measurements were excluded and the final hardness value was determined by calculating the average of the three remaining readings.

2.5 Pressure-relief Performance

A pressure sensor is a clinical technology used to map the pressure-relief performance of support materials, including shoe insoles [18]. The study utilized the F-Scan™ pressure sensor system from Tekscan, USA, to visualize pressure distribution through color scattering. A color map was generated to depict the pressure distribution pattern, surface contact area and peak pressure values, with maximum pressure indicated in red and minimum pressure in dark blue. Prior to data collection, the pressure sensor was calibrated in accordance with the manufacturer's guidelines. For the test, the sensor was positioned on top of latex foam shoe insoles and a volunteer applied weight by stepping onto it.

3. RESULTS AND DISCUSSION

3.1 Foamability Study

Table 4 shows the effect of potassium oleate levels on foamability of AFNR latex and HANR latex. Without potassium oleate as the foaming agent, both AFNR latex and HANR latex did not reach the targeted 5 L of latex foam within 5 minutes time frame. Addition of potassium oleate at 0.5, 1.0, 2.0 phr of potassium oleate improved the foamability of both types of latex. However, the results show that AFNR latex exhibits lower foamability than HANR latex. Although increasing the potassium oleate level improved the foamability of AFNR latex, the volume of latex foam generated by AFNR latex remains lower than HANR latex at every evaluated potassium oleate level. The study observed that within the 5-minute time frame of the foaming process, the addition of 1.0 phr of potassium oleate resulted in 5 L of latex foam for HANR latex in 4.5 minutes, whereas AFNR latex produced only 4.5 L of latex foam over the entire 5 minutes. A similar trend was observed when the potassium oleate level increased to 2.0 phr, where HANR latex generated 5 L of latex foam in 4 minutes, whereas AFNR latex took 4.75 minutes to produce the same volume of latex foam. The differences in formation between AFNR latex and HANR latex, even with increased potassium oleate levels, may be due to several factors, such as viscosity, total solid content and chemical compositions. According to a previous study [14], foam formation involves the incorporation of air into the latex through mechanical agitation. When latex has high viscosity, it is denser and flows less easily, which limits its ability to trap air effectively, thereby reducing its ability to generate foam. However, as shown in Table 1, the difference in viscosity between AFNR latex and HANR latex is minimal, suggesting that their foamability should be similar. Another possible explanation is that AFNR latex may have a different chemical composition, specifically the preservative used to replace ammonia, which could influence its foamability. As a result, the interaction between potassium oleate, the foaming agent and AFNR latex may be less effective than the interaction of potassium oleate with HANR latex. Further study in these parts could provide valuable understandings into the differences in the latex foam formation between the two types of latex, thus becoming a focus for future research.

Table 4: Effect of potassium oleate levels on foamability of AFNR latex and HANR latex

Level of potassium oleate	AFNR latex		HANR latex	
	Time (min)	Volume (L)	Time (min)	Volume (L)
0 phr	5.0	3.2	5.0	4.0
0.5 phr	5.0	3.8	5.0	4.5
1.0 phr	5.0	4.5	4.30	5.0
2.0 phr	4.75	5.0	4.0	5.0

Aside from differences in foaming behavior, the study observed that AFNR latex foam requires longer gelling time compared to HANR latex. In this study, latex foam prepared with 2.0 phr potassium oleate was examined as both AFNR latex foam and HANR latex foam reached the targeted 5 L within 5 minutes timeframe. The study revealed that AFNR latex foam requires 6 minutes to achieve complete gelation at room temperature, while HANR latex completely gels within 4 minutes. As mentioned earlier, AFNR latex incorporates a new chemical composition that substitutes ammonia as a preservative. This composition may also account for the exceptionally high mechanical stability time (MST) of AFNR latex, as shown in Table 1, which indicates the stability of the latex. Therefore, it is presumed that the new chemical composition used in AFNR latex could also contribute to a longer gelling time in the production of AFNR latex foam. Changes in chemical composition can affect reaction rates and properties during processing, potentially leading to variations in gelling time [10].

3.2 AFNR Latex Foam Shoe Insoles

Figure 2 demonstrates the appearance of HANR latex foam shoe insoles and AFNR latex foam shoe insoles produced in this study. Both HANR and AFNR latex foam shoe insoles appeared off-white, which is a typical color for NR latex foam products.

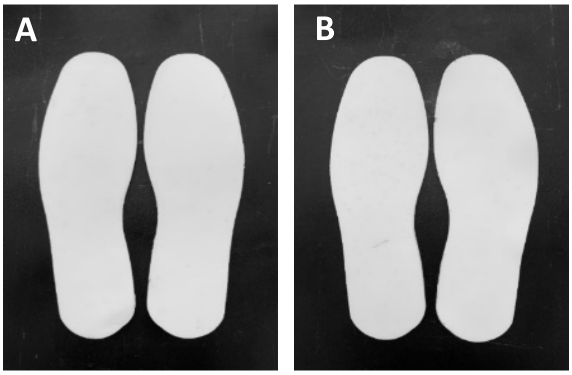


Figure 2: The appearance of latex foam shoe insoles of (a) HANR latex foam and (b) AFNR latex foam

This implies that the changes in the preservative system used for AFNR latex did not alter the basic aesthetic qualities of the latex foam insoles. This is an important aspect, as the color and overall aesthetics of footwear components can significantly influence consumer perception and marketability.

3.3 Morphological Structures

Figure 3 shows the morphological structure of AFNR latex and HANR latex. A cross-sectional view using FESEM reveals that both AFNR and HANR latex foams exhibit open-cell structures. An open-cell structure refers to a foam configuration in which the cells (or bubbles) are interconnected.

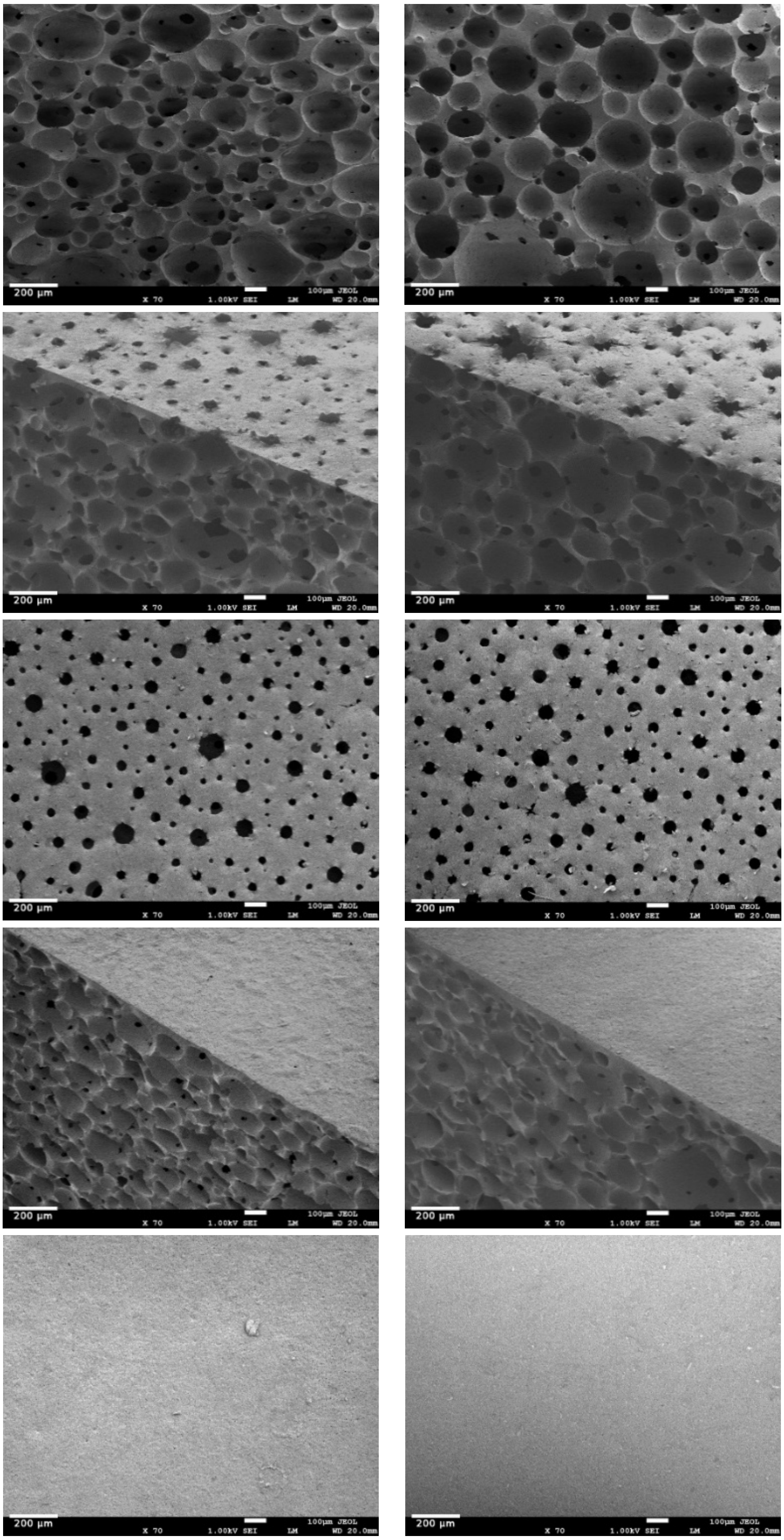


Figure 3: Morphological structure of latex foam shoe insoles visualized at ×70 magnification, 20 mm working distance, 1.0 kV

This arrangement results in high porosity, contributing to the lightweight characteristics of the foams. Additionally, open-cell foams are typically more flexible and compressible, allowing for air circulation (breathability), which makes them ideal for cushioning applications where comfort and temperature regulation are key concerns. In the context of shoe insole applications, the breathability of the latex foam helps prevent heat buildup, enhancing the overall comfort of the material while in use. The side view of the latex foam shoe insole samples reveals that both samples have a thicker, smoother skin on the lower part compared to the upper part. The lower part is where the foam faces the mold, while the upper part is exposed to the air. Although the mold is closed with a lid during the manufacturing process, the upper part does not contact or attach to the lid because the latex foam slightly deforms during gelling at room temperature. As a result, when the lid is placed, it does not touch the foam as the bottom part does. Consequently, the lower part has smoother skin compared to the upper part. The smoother skin on the bottom may also be attributed to the drainage of the latex foam. During the gelling process, some of the latex foam tends to liquefy (changing from the foam phase to the liquid phase) due to gravitational forces [14]. This could also be the reason the lower skin is thicker than the upper skin.

Figure 4 shows the average foam cell diameter and average skin thickness for AFNR and HANR latex foam obtained using ImageJ software analysis. The diameter of foam cells in HANR latex foam is slightly larger than that in AFNR latex foam. Moreover, the results indicate that the standard deviation for HANR latex foam is narrower than that for AFNR latex foam, suggesting that the foam has a more uniform size. The average skin thickness at the lower part of AFNR latex foam is slightly higher than that of HANR latex foam, although there is not much difference observed at the upper part. This can be attributed to the gelling process described earlier, where a slower gelling time may lead to latex drainage before gelation occurs, resulting in increased skin thickness at the lower part of the latex foam.

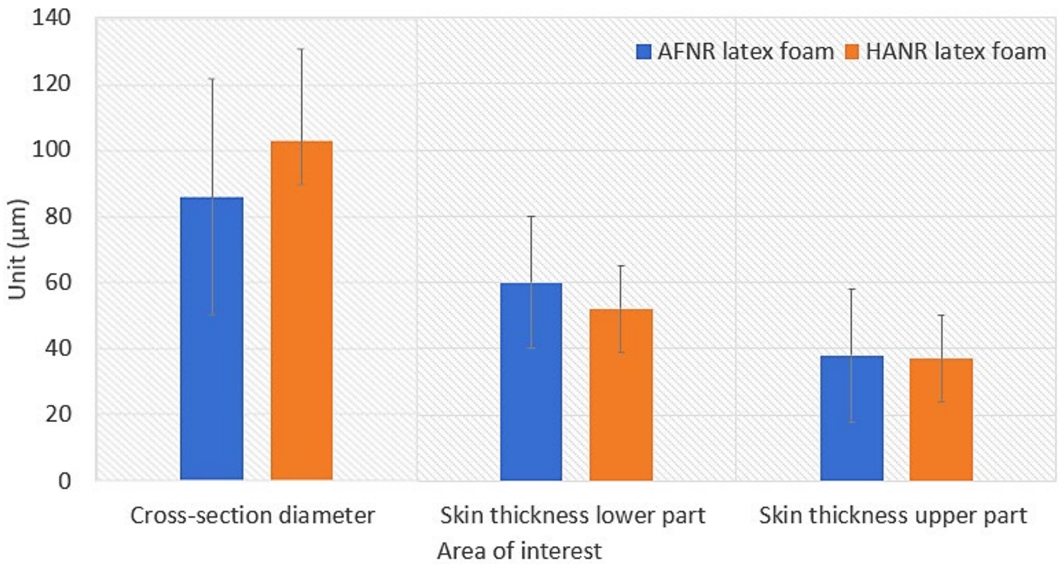


Figure 4: Comparison on foam cell diameter and skin thickness between AFNR latex foam and HANR latex foam

3.4 Shore F Hardness and Pressure-relief Performance

Table 5 shows the Shore F hardness values for the upper and lower parts of the latex foam samples, comparing AFNR latex foam and HANR latex foam. The findings reveal that the Shore F hardness values for the upper part of both types of latex foam are consistently lower than those for the lower part. This indicates that the upper part is softer while the lower part is firmer. A softer surface may offer better pressure-relieve performance. On the other hand, the firmer surface is significant for applications where durability is essential. In examining the differences between the two types of latex foam, the results show that there is minimal variation in Shore F hardness values in the upper part for both AFNR and HANR latex foam. This similarity suggests that both materials perform comparably in terms of softness as well as possibly pressure-relief performance. However, in the lower part, AFNR latex foam exhibits slightly higher Shore F hardness compared to HANR latex foam. This difference may imply that the morphological structure contributes to enhanced firmness of AFNR latex foam shoe insoles. The higher Shore F hardness values in the lower part, compared to the upper part, as well as between AFNR and HANR latex foams, can be attributed to the increased skin thickness, as illustrated in Figure 3 and Figure 4.

As indicated in a previous study [19], a thicker skin layer contributes to the foam's ability to maintain its shape and support under load, thereby resulting in higher Shore F hardness values. Thicker skin layers enhance the overall structural integrity of the latex foam, making it more resistant to deformation when subjected to heavy duty applications. This characteristic is crucial for applications such as shoe insoles, where both comfort and support are necessary for prolonged wear.

Table 5: Shore F hardness values for the upper and lower parts of the latex foams

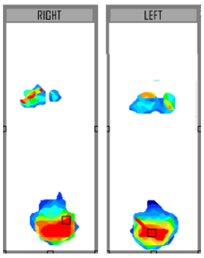
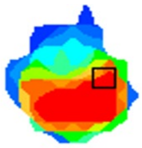
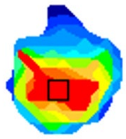
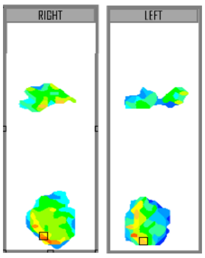
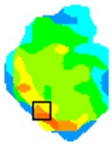
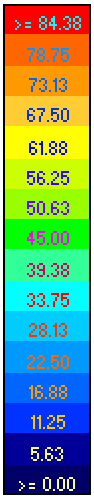
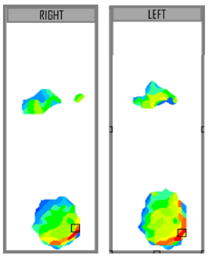
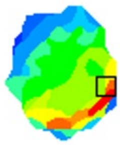
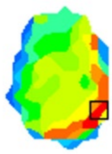
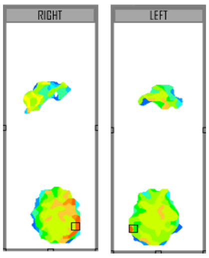
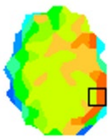
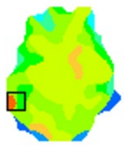
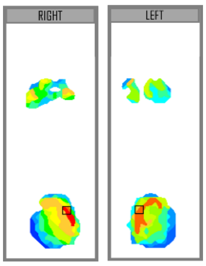
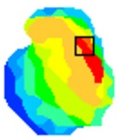
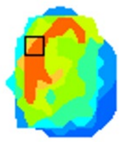
Type of material	Upper part	Lower part
HANR latex foam	71.56 ± 4.3	83. 5 ± 4.0
AFNR latex foam	71.6 ± 7.1	88.2 ± 1.2

Table 6 presents snapshots from real-time pressure mapping, showing pressure distribution patterns and average peak pressure values for both HANR latex foam shoe insoles and AFNR latex foam shoe insoles when a volunteer stepped on them. For comparative analysis, pressure mapping evaluation was conducted on both sides of the shoe insoles (the lower and upper parts) to investigate whether the skin thickness and hardness of the latex foam affect the pressure-relief performance of the materials. The study observed irregular color intensities and mapped contours, indicating the interface pressure between the latex foam shoe insoles and the volunteer's foot, corresponding to the volunteer's body weight. However, in each test, the pressure distribution was concentrated in two primary areas: the metatarsal and heel. This implies that these two areas have the greatest surface contact between the shoe insoles and the foot during standing. Additionally, the peak pressure value (indicated as a black square box) was observed at the heel area, signifying the highest pressure point of the foot.

The high pressure at the heel may be attributed to the bony structure of that area [20,21]. As the heel is a bony prominence, it exerts more pressure on the insoles than the softer, fleshy areas of the foot. The morphological analysis presented in Figure 3 shows that the lower part of the shoe insoles has a greater skin thickness compared to the upper part. This increased thickness is associated with a higher Shore F hardness, as indicated in Table 6. Despite this greater hardness in the lower part, the pressure mapping study indicated that the peak pressure values between the lower and upper parts of the insoles did not differ significantly.

Furthermore, the study found that the pressure distribution patterns and peak pressure values between the shoe insoles made from AFNR latex foam and those made from HANR latex foam are quite similar. This similarity indicates that both types of latex foam perform comparably in terms of pressure distribution and peak pressure, despite any differences in material properties. This finding suggests that, although the skin thickness at the lower part is thicker, it does not translate to a notable difference in pressure experienced by the foot. This could imply that factors other than skin thickness such as the overall design of the insoles or the materials used play a crucial role in pressure distribution [20,22]. Understanding these dynamics is essential for optimizing shoe insole design to enhance comfort and reduce pressure points, especially in critical areas like the heel and metatarsal areas.

Table 6: Pressure-relief performance of latex foam shoe insoles

Type of material	Pressure distribution pattern	Average peak pressure value (kPa) - Right	Average peak pressure value (kPa) - Left	Pressure scale
Without latex foam shoe insoles		 167.22	 159.75	
HANR latex foam shoe insoles – upper part		 98.87	 97.97	
HANR latex foam shoe insoles – lower part		 110.87	 101.97	
AFNR latex foam shoe insoles – upper part		 100.55	 98.45	
AFNR latex foam shoe insoles – upper part		 113.15	 109.76	

*Black square box located the average peak pressure.

4. CONCLUSION

AFNR latex foam and HANR latex foam shoe insoles were produced using the Dunlop batch foaming process. Even though AFNR latex demonstrates lower foamability than HANR latex, increasing the potassium oleate level in the foaming formulation improves its foamability. Cross-sectional analysis using FESEM of both types of latex foam samples reveals that both are open-cell foams. The FESEM images also show that both samples have smoother, thicker skin on the bottom part compared to the upper part. This difference in skin structure contributes to higher Shore F hardness values, but no significant difference in pressure-relief performance was observed. Despite differences in foamability and skin structure, AFNR latex shows comparable pressure-relief performance to HANR latex, indicating its potential for application in latex foam shoe insoles. Nevertheless, further research is needed to evaluate the mechanical properties (e.g., compression strength, aging) of the latex foam. Such studies would provide valuable insights into its durability and comfort in footwear applications, particularly as shoe insoles.

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Author Contributions

Conceptualisation; [RR, AM, RCA, SM]; Methodology: [RR, RCA, SM] Formal analysis and investigation: [RR, RCA, SM]; Writing - original draft preparation: [RR, AM, RCA, SM]; Writing - review and editing: [RR, AM]; Funding acquisition: [RR, AM]; Resources: [RR, AM, RCA, SM]; Supervision: [AM]. All authors reviewed the results and approved the final version of the manuscript.

Disclosure of Conflict of Interest

The authors declare that there is no conflict of interest with respect to the publication of this article.

Compliance with Ethical Standards

The work is compliant with ethical standards.

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