



Production of Bio-Polyurethane Foam from Pumpkin Seeds

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Abstract

Bio-based polyurethane (PU) foams were synthesized from pumpkin seed oil (PSO) polyol, extracted via the cold bulk method, converted to polyol, and blended with Arcol 1180 (petroleum-based polyol) at 30:70 and 50:50 ratios, alongside formulations containing 100% PSO polyol and 100% Arcol polyol. The physical properties of the foams, including compression set (CS), apparent density, tensile properties (such as yield strength, ultimate tensile strength, elongation at break, and Young's modulus), and porosity index, were thoroughly characterized. The surface properties and structure of the samples were studied using scanning electron microscopy (SEM), while the chemical structure was analyzed using Fourier Transform Infrared (FTIR) spectroscopy. The compression set results showed that increasing the PSO content improved compression strength, with the 50:50 PSO-PU blend exhibiting the lowest compression set (CS) value of 37.5%, followed by the 30:70 PSO-PU blend with a CS value of 44.23%, and the 100% Arcol-PU sample had the highest CS value of 49.9%. Apparent density values ranked as 30:70 PSO-PU (63.10 kg/m³) > 50:50 PSO-PU (62.0 kg/m³) > 100% Arcol-PU (27.10 kg/m³). Tensile testing showed 50:50 PSO-PU (1.80 N/mm² tensile strength; 1.36 N/mm² yield strength) outperforming 30:70 PSO-PU (1.70 N/mm²; 1.30 N/mm²), while 100% Arcol-PU exhibited the highest tensile and yield strengths (2.73 N/mm²; 2.27 N/mm²). Elongation at break was highest in 50:50 PSO-PU (5.51 mm) compared to 3.51 mm (30:70 PSO-PU) and 3.90 mm (100% Arcol-PU). Porosity indices were 43% (50:50 PSO-PU), 35% (30:70 PSO-PU), and 50% (100% Arcol-PU). SEM analysis revealed uniform, thin-walled cells in PSO-PU blends (30:70 and 50:50), but coarse, thick-walled cells in the pure PU samples (100% PSO-PU and 100% Arcol-PU). FTIR spectra confirmed the absence of free –OH groups, indicating complete conversion of hydroxyl functionalities into urethane linkages (NH–C(O)–O).

Key words: Polyurethane, Flexible, Foam, Polyol, Biobased, Pumpkin Seed, Synthetic

Introduction

Vegetable oil (VO) is a valuable renewable resource due to its universal availability, biodegradability, and low cost (Lucas et al., 2011). It consists of triglycerides and three long-chain fatty acids, with properties influenced by chain length and double bonds (Islam et al., 2014). VO is chemically modified to create polyols, essential for the synthesis of polymers like polyurethanes and polyesters (Shida et al., 2014). Pumpkins are North American natives (Paris, 2016), widely distributed in Nigeria and belong to the Cucurbitaceae family (Gourd family) of plants, which include a wide variety of popular fruits like cucumbers, squash, and melons (Altafi et al., 2025). The botanical name for pumpkin is *Cucurbita pepo* and are also known by their scientific name Pepita, which is the edible kernel of the seed, often roasted or dried for snacking or culinary use (Ranallo, 2018). Pumpkin seeds are obtained from pumpkins (*Cucurbita pepo*) and are excellent source of unsaturated fatty acids, especially linoleic acid, as well as essential amino acids, carbohydrates, fiber, and minerals (Vinayashree & Vasu, 2021). They are also a good source of antioxidants (Vinayashree et al., 2024). Pumpkin seeds are a nutrient-rich oilseed (Sharma et al., 2020) that can be used as a sustainable raw material for the production of polyurethane. The seeds contain a high percentage of oil which can be extracted and converted into polyols through transesterification or hydroxylation (Oluwafunmilayo et al., 2020). These Pumpkin seeds are a renewable resource and the production process requires less energy and generates fewer emissions than the traditional polyurethane production methods (Njoku et al., 2019). In addition, pumpkin cultivation requires less water and pesticides than many other oilseed

crops. Pumpkin seeds, being rich in oil and protein make them an attractive feedstock for producing bio-based materials (Sharma et al., 2020; Nimalaratne et al., 2018). Much attention is being paid to the preparation and application of bio-based polymers because of environmental concerns (Islam et al., 2014). Polyurethane (PU) is a polymer composed of isocyanate and polyol components. PU is typically produced from petroleum-based polyols, such as polyethylene glycol (PEG) or polypropylene glycol (PPG). Polyurethane (PU) is a highly versatile polymer widely utilized across numerous applications, particularly in the production of foams, coatings, adhesives, and elastomers. PU foams commonly utilized in cushioning, insulating and packaging are typically derived from petroleum-based polyols. Pumpkin seeds, like vegetable oils and biomass, are a renewable, biodegradable resource and their use as raw material for PU production offers a sustainable alternative to petroleum-based polyols and decreases reliance on fossil fuels (Njoku et al., 2019). The PU derived from pumpkin seed oil has demonstrated the ability to have similar properties to petroleum-based polyurethane, such as mechanical strength, thermal stability, and chemical resistance (Ranallo, 2018; Adewale & Oluwaseun, 2020).

Producing bio-based polyurethane (PU) foam from pumpkin seed oil presents a promising sustainable alternative to traditional PU production (Paris, 2016; Adewale & Oluwaseun, 2020). Polyurethanes (PUs) are versatile materials with excellent mechanical strength, wear resistance, and chemical resistance, widely used in various industries, including consumer goods, automotive, construction, and medical devices (Okoro & Nwabanne, 2019). Conventionally, PUs are synthesized by reacting polyols with hydroxyl groups with polyisocyanates (Borowicz et al., 2020). However, the reliance on finite petroleum resources and environmental concerns necessitate alternative pathways. Renewable seed oils, such as pumpkin seed oil, offer a reliable platform for synthesizing novel PUs (Piotr, 2012). These oils can be modified to produce polyols, which can then be reacted with diisocyanates to yield eco-friendly PUs (Maisonneuve et al., 2016; Ameh et al., 2024). The properties of PUs are closely linked to the characteristics of the contributing polyols (De Vasconcelos et al., 2013), and ongoing research focuses on developing strategies to transform seed oils into PUs with enhanced mechanical and thermal properties (Maisonneuve et al., 2016).

This research focuses on the production of polyurethane using a biobased polyol from pumpkin seed oil (100 % PSO), and a mixture of the biobased polyol and the petroleum based polyol in the ratios 30:70 and 50:50 and lastly 100% petroleum based polyol (Arcol 11080) polyurethane subsequently compare their properties with those of the polyurethane produced with the 100% petroleum based polyol (Arcol).

Foam formulation and preparation

The flexible foams were prepared by using well- established formulations (Table1.1) based upon parts per hundredth parts of the polyol. The amount of each component was based on the 100 parts by weight of total polyols. The amount of diphenylmethane diisocyanate (MDI) stoichiometrically balance NCO and reactive hydrogen species, i.e. isocyanate index= 80.

Table 1: Formulation for the foam sample

Ingredients	(30:70)% PSOPU (g)	(50:50)% PSOPU (g)	100% PSOPU (g)	100% ARCOLPU (Reference foam)	(g)
PSO polyol	45(30%)	75(50%)	42(100%)	-	
Arcol 1108	105(70%)	75(50%)	-	150(100%)	
Water	3.5	3.5	3.5	3.5	
Silicon oil	0.5	0.5	0.5	0.5	
Stannous octate	4	4	4	4	
Triethanol amine	0.5	0.5	0.5	0.5	
CaCO ₃	0.5	0.5	0.5	0.5	
Pigment (yellow)	0.5	0.5	0.5	0.5	
MDI	100	100	100	100	

Foams with an isocyanate index of 80 were prepared with 30% PSO Polyol, 50% PSO Polyol, and 100% PSO Polyol, respectively. A reference foam was also prepared using 100% petroleum polyols (Arcol 1180). The components of the PUR mixture were mixed with a wooden stirrer.

These materials listed in Table1 were weighed in beakers and labeled appropriately. Water, silicone oil, and triethanolamine (catalyst) were then poured into a steel container and stirred for 60 seconds. Polyol, CaCO_3 (filler), stannous octoate, and the yellow pigment were then added to the mixture and stirred vigorously for 10 seconds. The mixture was then poured into a mold, after which the isocyanate (methylene diisocyanate) was added and the mixture was stirred vigorously for 30 seconds using a wooden stirrer. The PU foam rose and allowed to cure in open air at room temperature for 24 hours. The foam was conditioned for no less than 72 hours under ambient conditions (25°C and 50% relative humidity), and foam cubes were eventually removed from further testing.

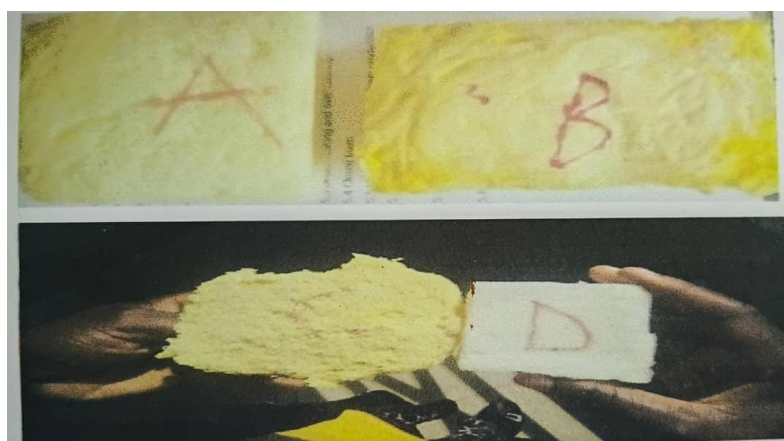


Figure 1: Foam samples for (50:50)%PSOPU(A), (30:70)%PSOPU(B), 100%PSOPU(C) and 100%ARCOLPU (D).

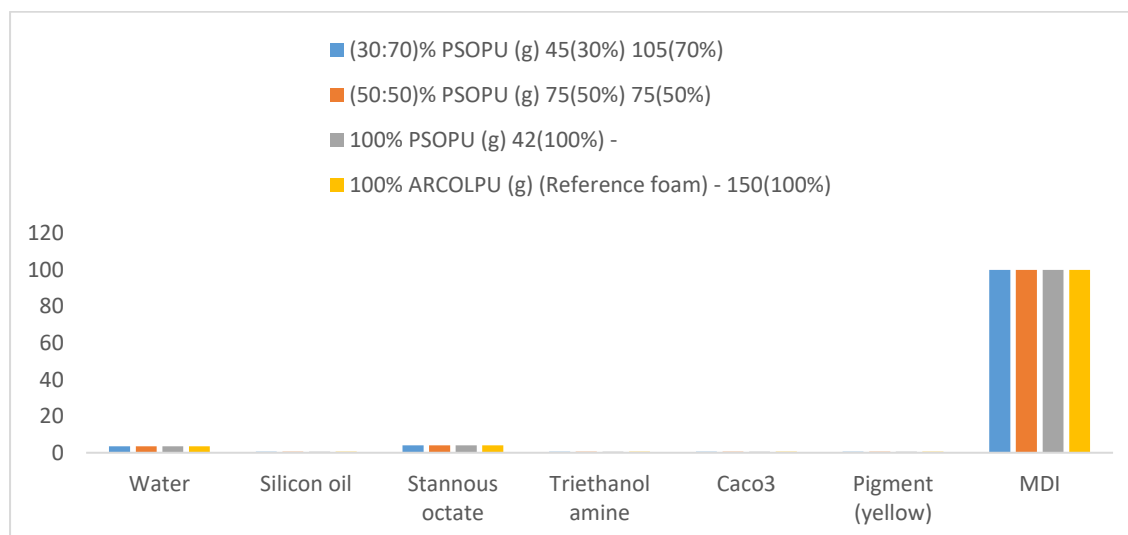


Fig. 2: Formulation for the foam samples.

Results

Properties of the PSO

Table 2: The physical characteristics of the fluted pumpkin seed oil (PSO)

Characteristics	PSO
Refractive index	1.46
Specific gravity	0.92
Colour	Deep yellow
Odour	Non- offensive
Viscosity(cSt)	60.4
Smoke point(°C)	170
Density(g/cm ³)	911
Flash point (°C)	290
Boiling point(°C)	60
PH	5.9

Discussion

The refractive index value obtained for PSO (1.460) was low compared to the value (1.476) obtained by Nwabanne, (2012) but higher than 1.453 reported by Okpashi et al., (2013). The refractive index for PSO aligns closely with the values of conventional oils, which range from 1.466 to 1.470 for soybean oil and from 1.449 to 1.451 for palm kernel oil. Refractive-index of oils is a measure of how much a light ray is bent when it passes from air into the oil and it usually depends on the density of the oil. In general, the refractive index and relative density/specific gravity values of edible vegetable oils are physical measures of adulteration of vegetable oils, since different oils have characteristic density/specific gravity and refractive index (Olutoye & Garba, 2008).

The PSO demonstrated a specific gravity of 0.921, though higher than the value reported for tomato seed oil by Sangeetha et al., (2023), but closely aligned with the specific gravity of 0.920 reported by Muibat et al., (2011) and the 0.926 noted by Nwabanne, (2012). The oil was observed to have a yellow hue and a pleasant smell, similar to the findings reported by Muibat et al., (2011), which also noted a light yellow color and non-offensive odor. The viscosity measurement of (60.40 cSt) indicates the oil's resistance to shear stress. The viscosity of the PSO was comparable to the value (60.20 cSt) found by Muibat et al., (2011) and was greater than that of groundnut and various other common oils like soybean (31 cSt), cottonseed (36 cSt), and sunflower (43 cSt) (Muibat et al., 2011). The smoke point of PSO was lower (170 °C) when compared to the 245 °C reported by Muibat et al., (2011), and the free fatty acids content for the PSO was also low. This aligns with the findings that a lower free fatty acid content correlates with a higher smoke point (FAO & WHO, 1993), indicating that the seed oil may be appropriate for deep frying.

The density of the PSO obtained was 0.911 g/cm³, a value comparable with 0.920, 0.921 and 0.934 obtained by Muibat et al., (2011); Okpashi et al., (2013) respectively. A flash point of 290°C was recorded for the PSO, which surpasses the 170°C found for *T. occidentalis* by Nwabanne, (2012). The flash point is a crucial parameter for evaluating the stability of oils, especially vegetable oils used in cooking. It defines the temperature at which oil vapors ignite when exposed to heat. A higher flash point indicates superior heat stability. The boiling point of pumpkin seed oil (PSO) was established at 60°C, consistent with the findings of Nwabanne (2012). Additionally, physical characterization revealed a slightly acidic pH of 5.9, closely matching the value of 6.03 reported by Olutoye and Garba (2008). This acidity can likely be attributed to the presence of unsaturated fatty acids in the oil.

Table 3: Chemical properties of the seed oil

Characteristics	PSO
Oil content (%)	50
Iodine value (gl ₂ /100g)	114.1
Acid value (MgKOH/g)	2.8
Free fatty acid (%)	1.92
Peroxide value (MeqKOH/g)	6.16
Saponification value (MgKOH/g)	174.05

Table 3: presents the chemical characteristics of pumpkin seed oil (PSO). The calculated oil yield is 50%, which is higher than the 46.20% reported by Okpashi et al., (2013). The iodine value indicates the degree of unsaturation in oil and can be used to assess the quantity of double bonds in the oil, reflecting its tendency to be oxidized. The high iodine value signifies a considerable proportion of unsaturated fatty acids in the seed oil. With an iodine value slightly over 100, it can be categorized as a semi-drying oil. The PSO's iodine value of 114.10 is comparable to the values of 115.60 and 123.88 reported by Muibat et al., (2011) and Nwabanne, (2012), respectively. Additionally, this iodine value is akin to those found in white and red *Sesamum indicum* seeds, which are 103 and 116, respectively (Mohammed & Hamza, 2008).

The acid value of the PSO is 2.8, which is lower than the values (4.77, 3.51, and 3.48) obtained by Nwabanne, (2012), and Muibat et al., (2011), respectively. However, it is higher than the value (0.43) obtained by Okpashi, (2013). The acid value of the oil falls within the recommended range (0.00 to 3.00 mg KOH/g) for cooking applications (Oderinde et al., 2009), suggesting that the PSO (*T. occidentalis*) may be appropriate for culinary use. The free fatty acid content of the PSO stands at 1.92%, which is comparable to the 1.74% reported by Muibat et al., (2011). Additionally, the free fatty acid level of the PSO is within the acceptable limit of 5% for high-quality palm oil in Nigeria (Muibat et al., 2011). The peroxide value serves as an indicator of rancidity; therefore, a lower peroxide value suggests that the oil is less prone to peroxidation during storage. The peroxide value of the PSO exceeds the value for tomato seed oil (2.50) reported by Sangeetha et al., (2023) and the values (0.20 and 2.26) reported by Okpashi, (2013) and Muibat et al., (2011) respectively for the same seed. The peroxide value obtained for the PSO was 6.16 mEq/Kg. This value is lower than the value 6.8 reported by Agu et al., 2023 for the same seed, but it is higher than average when compared to the maximum permissible limit of 10 meq KOH/g established by the Codex Alimentarius Commission for groundnut seed oils (Kozłowska et al., 2025).

The PSO is therefore stable and would not easily go rancid. The high saponification value (174.05) recorded for the PSO indicates its potential use in industry and its application in soap production and other cosmetic products such as shampoo (Amoo et al., 2004). It is closely related to the values (158.40, 162.69, and 179.02) obtained by Akubugwo et al., (2008); Nwabanne, (2012), and Agatemor, (2006), respectively for the same seed.

Table 4: Fatty acid compositions of the pumpkin seed oil (PSO)

Fatty acids	Relative (%)
Palmitic acid (C16:0)	17.42
Oleic acid (C18:1)	13.74
Linoleic acid (C18:2)	65.43
Eicosapentaenoic(C20:5)	3.41

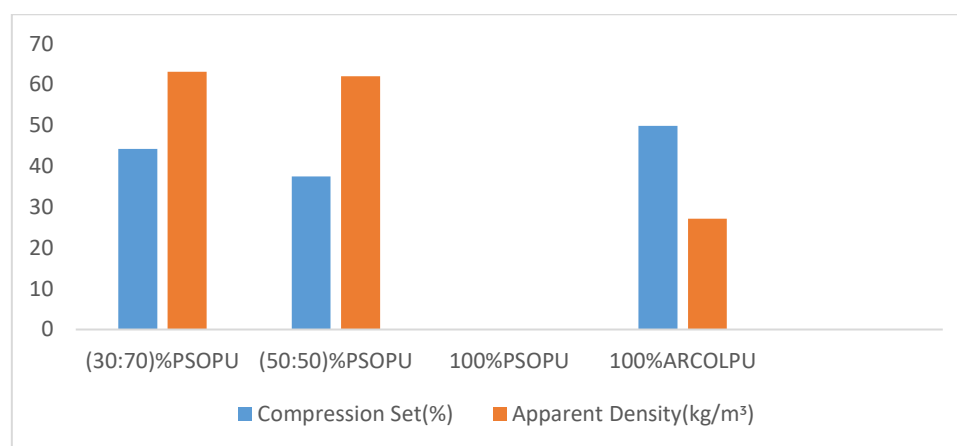
Table 4: presents the fatty acid composition of the seed oil. The only saturated fatty acid identified in *T. occidentalis* (PSO) was palmitic acid, which had a relative abundance of 17.42 %. Additionally, oleic acid accounted for 13.74 %, while linoleic acid made up 65.43 %, and Eicosapentaenoic acid was found at 3.41%. PSO can be categorized within the oleic-linoleic acid group. The fatty acid data obtained for the PSO was similar to the values (19.74 % palmitic acid, 13.42 % oleic acid, 64.41% linoleic acid and 2.43% Eicosapentaenoic acid) obtained by (Mubat et al., 2011).

Compressive set test (compression strength) and apparent density

The resulting foams PSOPU foams had similar apparent densities of about 62kg/m³ and 63.1kg/m³ for (50:50) % PSOPU and (30:70) %PSOPU respectively while 100%ARCOLPU had a density of 27kg/m³. The 100% PSOPU produced a flaky semi rigid solid which had no compression set and density. Table 5, shows the values for compression set and apparent density of the foam samples. The plots of compression set (strength) and apparent density versus foam formulations are shown in Figure 3.

Table 5: Compression Set and Apparent Density of Foam Samples

Foam samples	Compression Set (%)	Apparent Density(kg/m ³)
(30:70)%PSOPU	44.23	63.1
(50:50)%PSOPU	37.5	62
100%PSOPU	-	-
100%ARCOLPU	49.9	27.1

**Fig 3: Apparent Density and Compression Set of Polyurethane (PU) Foam**

The compression set of a material is the permanent deformation remaining when a force (that was applied to it) is removed. The compression set (strength) of the foam samples was measured in perpendicular directions (direction in which foam rose). From Figure 3 above: 100 % ARCOLPU foam sample offered the greatest compression set (49.9%), this is followed by (30:70) % PSOPU foam (44.23%). The (50:50) % PSOPU foam sample showed the least compression set (37.5%). The noteworthy point arising from the graphs is the decrease in the percentage of compression set of the PSOPU foams with an increase in PSO polyols content, which indicates an increase in compression strength. However, in the case of the reference foam (100% ARCOLPU), the opposite relationship can be noticed. This can be explained by different cross-linking densities of the obtained foams (Olivito et al., 2023)

Tensile Properties; The impact of foam formulations on their tensile properties, giving some indication of foam durability, is presented in Tables 6 and 7, where the values of force and extension, stress and strain, yield strength, ultimate strength, and elongation at break for the foam samples are represented.

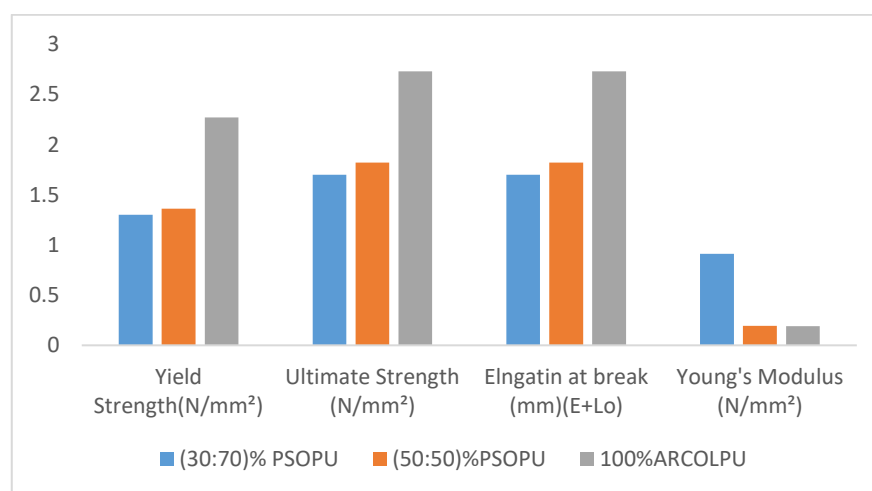
Table 6: Force against extension and stress against strain of foam samples

force (N)	Stress (N/mm)	Extension (E)(mm)			Strain (%)		
		(30:70)% PSOPU	(50:50)% PSOPU	100% ARCOLP U	(30:70)% PSOPU	(50:50)%P SOPU	100%ARC OLPU
0	0	0	0	0	0	0	0
50	0.45	0.2	0.1	0.1	0.7	2.1	2.9
100	0.91	0.3	0.2	0.15	10	4.2	4.3
150	1.36	0.4	0.3	0.25	13	6.3	7.1
200	1.82	0.1	0.45	0.3	20	9.4	8.6
250	2.27			0.38			11
300	2.73			0.5			14.3

Where Area=110mm²

Table 7: Yield strength, ultimate strength and elongation at break for the foam samples

Foam samples	Yield Strength(N /mm ²)	Ultimate Strength (N/mm ²)	Elongation break (mm)(E+Lo)	at Young's Modulus (N/mm ²)
(30:70)% PSOPU	1.3	1.7	1.7	0.91
(50:50)%PSOPU	1.36	1.82	1.82	0.194
100%ARCOLPU	2.27	2.73	2.73	0.191

**Fig 4: Yield strength, ultimate strength and elongation at break for the foam samples**

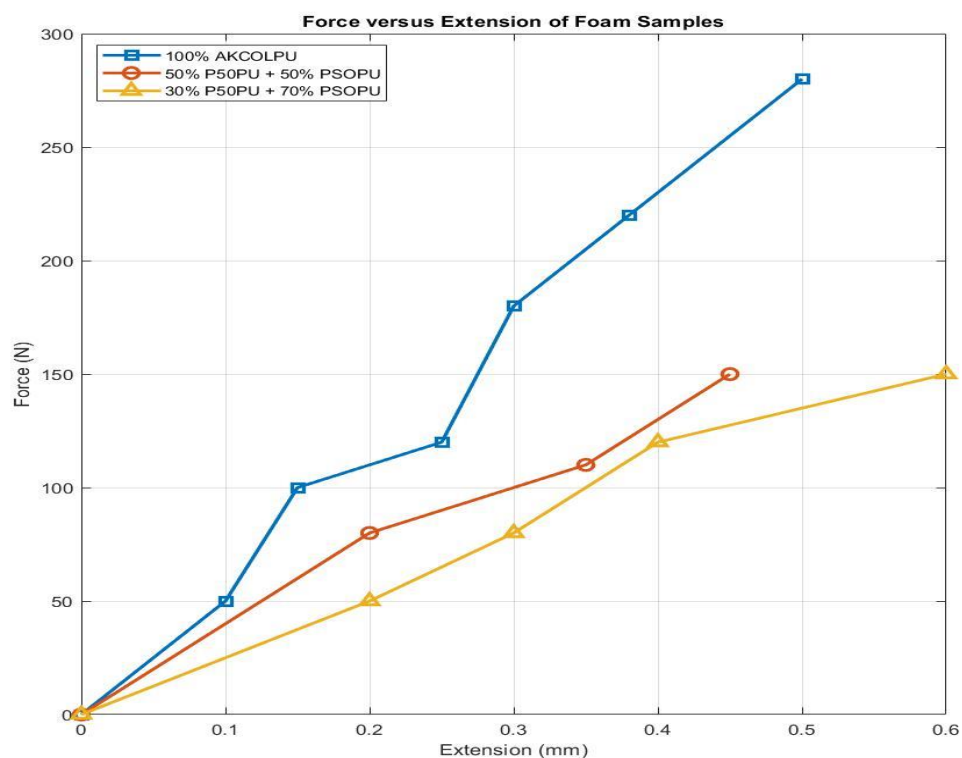


Fig 5: Force versus extension of foam samples

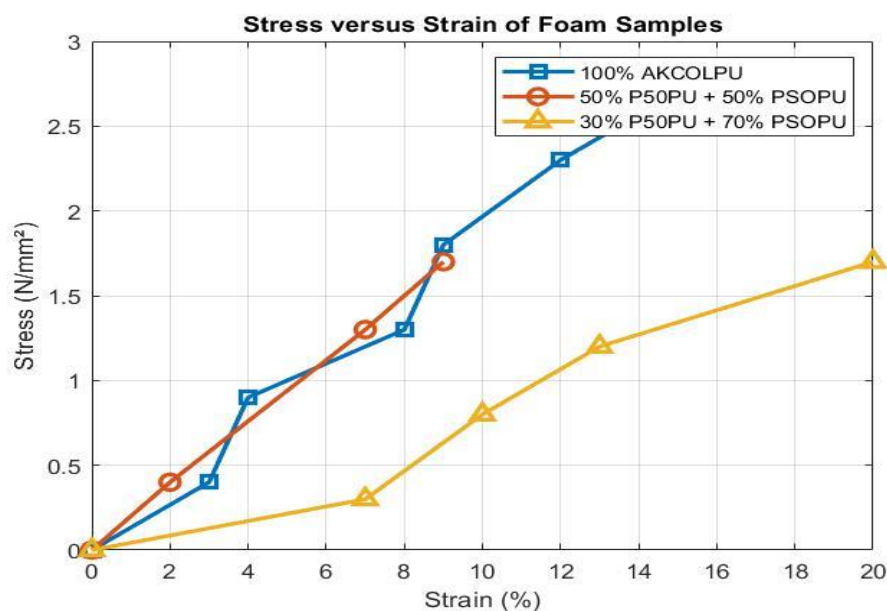


Fig 6: Stress versus Strain Samples

The ultimate tensile strength is the maximum tensile stress the foam samples sustain without fracture; it determines the resistance of the foam to tearing and shredding during its end-use application. The reference foam 100%

ARCOLPU gave the highest stress value of 2.73 N/mm² followed by (50:50)% PSOPU foam sample with 1.82 N/mm² and the (30:50)% PSOPU foam sample with 1.7 N/mm².

Yield strength refers to an indication of the maximum stress that can be developed in a material without causing plastic deformation. It is a stress at which a material exhibits a specified permanent deformation (Lligadas et al., 2013). The reference foam 100% ARCOLPU gave the highest value of 2.27 N/mm² followed by (50:50)% PSOPU foam sample with 1.36 N/mm² and the (30:50)% PSOPU foam sample with 1.3 N/mm².

The elongation at break is the amount of permanent extension of the foam sample that has been fractured in a tensile test. Both 100% ARCOLPU and (30:50)% PSOPU foam samples showed comparable elongation at 3.9 mm and 3.51 mm respectively, while (50:50)% PSOPU foam sample showed 5.1 mm elongation at break.

For the Young's modulus, which is the ratio of stress (below the proportional limit) to strain, i.e., the slope of the stress-strain curve and also considered the measure of rigidity or stiffness of the sample, showed that the (50:50)% PSOPU foam sample had the highest rigidity value of 0.194 N/mm², followed by 100% ARCOLPU foam sample with 0.191 N/mm², and then the (30:50)% PSOPU foam sample with 0.1368 N/mm².

Porosity Index; Porosity index indicates the degree of the cell openness of a material; it is the ability to absorb moisture into void spaces (Lligadas et al., 2013). From figure 4.7, the 100% ARCOLPU foam sample showed the highest porosity index of 50%, followed by the (50:50)% PSOPU foam sample with 43%, and the (30:50)% PSOPU foam sample with 43%. Finally, the 100% PSOPU foam sample had the lowest porosity index of 22%. It can be seen from figure 4, that an increase in PSO-Polyol content (i.e., from 30% to 50%) increases the porosity index of the foam and the rate of moisture absorption, although the 100% ARCOLPU foam sample showed higher cell openings.

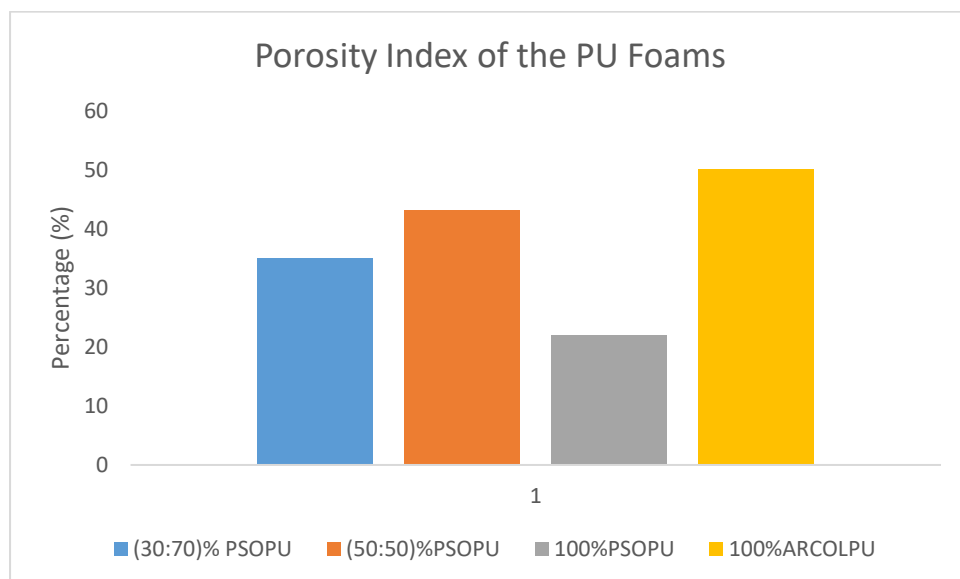


Fig 7: Porosity Index of the PU foams

SEM analysis; The polyurethane foams produced are flexible solids with an average void size of 1.3 mm (Skleničková et al., 2022). (30:70)% PU foams have small pores and are uniformly distributed with relatively thin walls, while (50:50)% PU foams also have a fine and uniform structure composed of larger elongated strip-like pores interconnected by larger pores of different sizes as shown in figure 8.

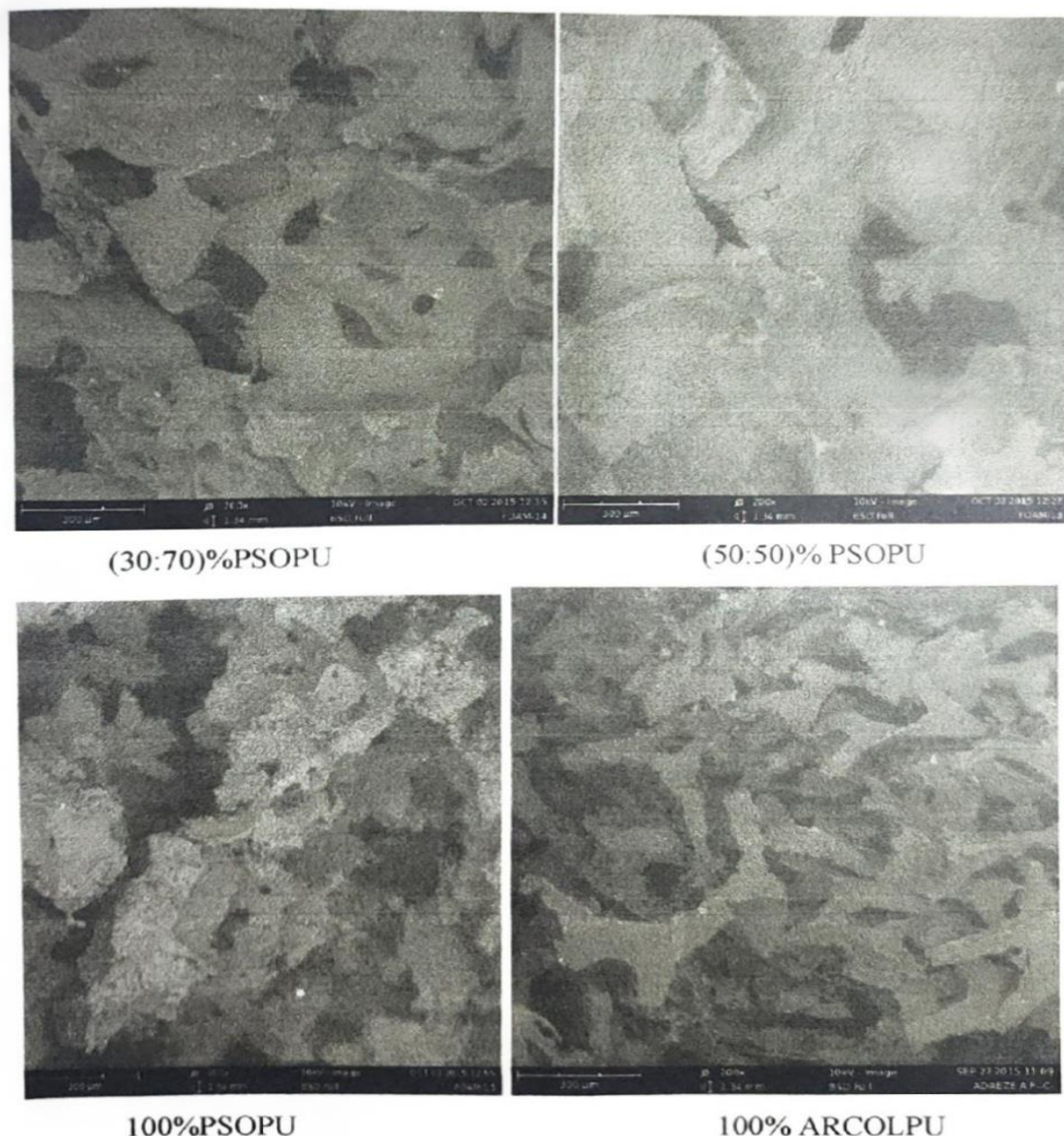


Fig 8: Scanning electron micrograms (SEM) of the PU foam samples (250 X magnification)

In the case of 100% ARCOLPU foams, the cells are very coarse, even when observed by the naked eye, and have the thickest walls, making them very porous. Combined with the lower closed-cell content, this resulted in the lowest compression strength. The 100% PSOPU foam also showed coarse cells and thick cell walls, which were lower in quality than those of 100% ARCOLPU.

The relatively poor quality of the 100% ARCOLPU foam microstructure might be attributed to its inadequate optimization of the catalyst and surfactant used in its formulation. It is known (Szycher, M. 1999) that the role of the surfactant is to stabilize the cell walls by lowering the surface tension between the cells and preventing their coalescence, resulting in smaller cells uniformly distributed over the network. Meanwhile, catalysts are added to accelerate reactions as required. The final cellular structure is a balance between the network formation and the blowing reaction.

FT-IR analysis; The PU foam samples were analyzed by FTIR spectroscopy, which showed the absence of free OH groups and indicated a complete conversion of both -OH groups of the PEA to the urethane moiety (NH-

C(O)-O). The typical FTIR spectrum of the PU foam samples is shown in figures 9a, 9b, and 9c. The characteristic -NH stretching vibration of the -NH₂- (amide) is located at 3405 cm⁻¹, overlapping with the OH peak as a broad band. Bands at 2932 cm⁻¹ and 2894 cm⁻¹ are the synchronous reflection of asymmetric and symmetric CH₂ bridges from the linkage of the urethane with the PEA. The band at 1650 cm⁻¹ corresponds to the overlapping of -N=C=O (urethane) and ester linkage of the PEA. The presence of bands at 1550, 1650, and 3350 cm⁻¹ indicates complete conversion to urethane.

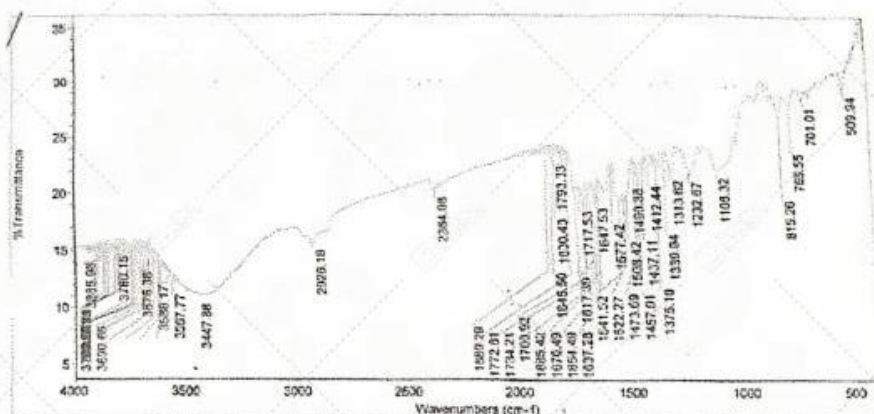


Fig 9a: FTIR spectrum of (30:70)%PSOPU

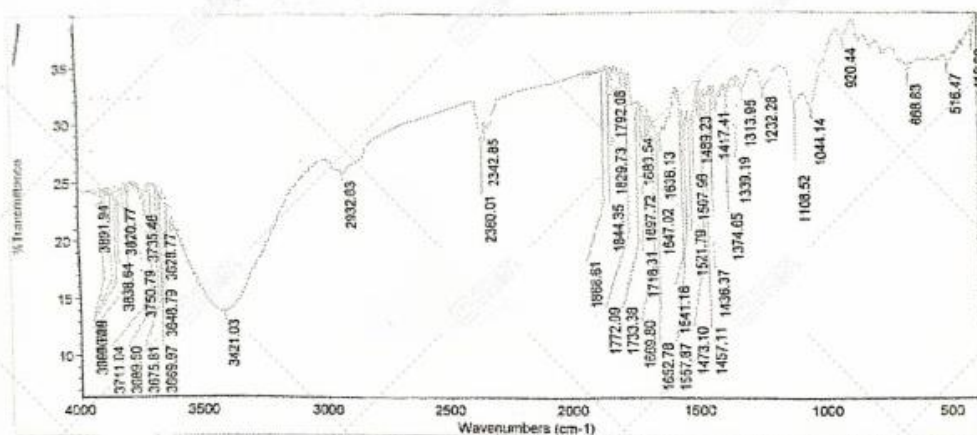


Fig 9b: FTIR spectrum of (50:50)% PSOPU

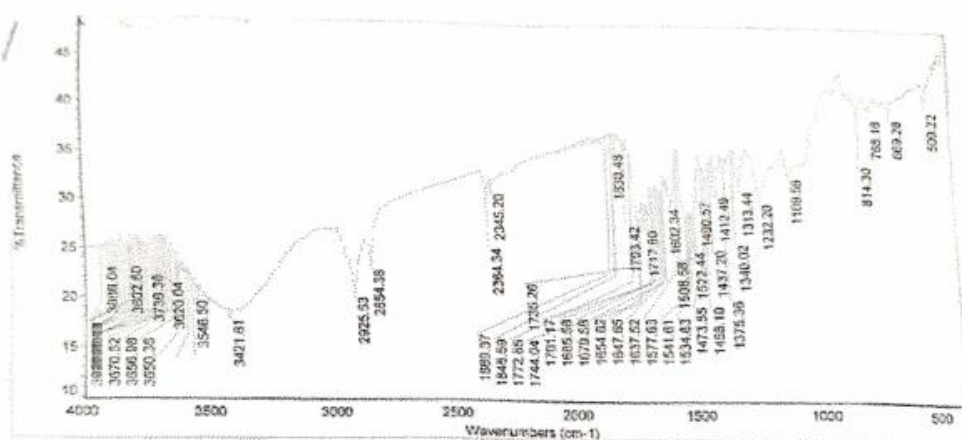


Fig 9c: FTIR spectrum of 100%PSOPU

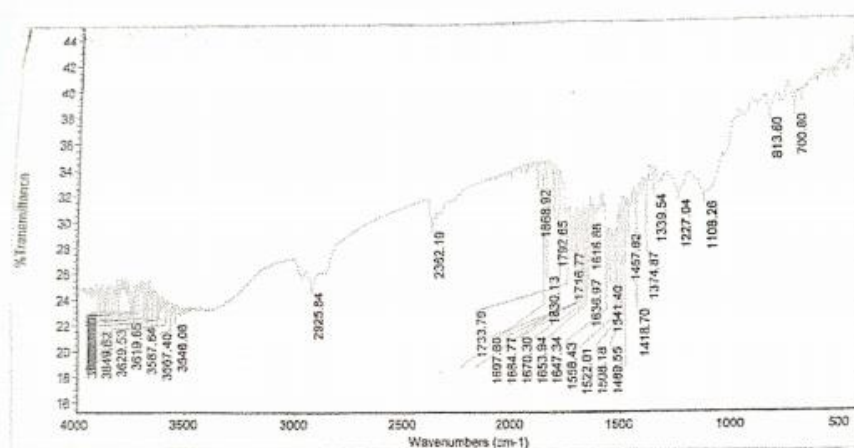


Fig 9d: FTIR spectrum of (30:40)%PSOPU, (50:50)%PSOPU, 100%PSOPU and the reference, 100%ARCOLPU

Conclusion

Currently, polyols derived from vegetable oils present a significant environmentally friendly opportunity. It is crucial to substitute petroleum-based polyols with those made from vegetable oils, such as pumpkin seed oil (PSO), which serves as an inexpensive and readily available renewable resource. Utilizing these green polyols lessens reliance on imported petroleum, reduces harmful impacts on human health and the environment, and enhances the use of renewable resources and agricultural products. Therefore, PSO-derived polyols are comparable to petrochemical-based polyols. The study also found that incorporating natural oil-based polyols into petroleum polyol-based foam formulations improves cell structure and compression strength. Despite this, observations revealed that foams with 50% PSO polyurethane and 30% PSO polyurethane demonstrated superior properties compared to 100% PSO polyurethane and 100% ARCOL polyurethane. This advancement is related to the cell structure, tensile strength, and elongation at break values. A key area for further exploration is the optimization of additives, catalysts, and surfactants for natural oil-based flexible foams.

Recommendations

The subsequent suggestions have been proposed:

1. PSO polyol is recommended for use in different formulations (20%, 40%, 60%, 70%, 80%, 90%) alongside petroleum polyol.
2. It is advised to combine PSO polyol with other seed oil polyols to evaluate the impact on foam characteristics.
3. PSO polyol should be employed in the manufacturing of rigid foam to assess its influence on the rigid foam.

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