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Abstract

The accumulation of used engine oil poses severe ecological threats to soil and groundwater systems, particularly in developing industrial regions where proper waste disposal systems are lacking. Simultaneously, the manufacturing of flexible polyurethane foams remains heavily dependent on volatile, petroleum-derived polyols, contributing to a high carbon footprint. This study presents a circular economy approach by chemically converting hazardous used engine oil into a polyhydroxylated organic extender (oil polyol) to partially replace commercial polyether polyols in flexible polyurethane foam formulations. The non-polar hydrocarbon chains of used engine oil were functionalized via an acid-catalyzed hydration reaction, converting thermally cracked alkenes into reactive secondary hydroxyl groups capable of forming urethane linkages. To investigate structural preservation and prevent cellular collapse, two composite foam series were synthesized using a one-shot free-rise foaming method: one containing 1.0 wt% natural starch as an organic biodegradable filler, and another containing 1.0 wt% natural zeolite as an inorganic nucleating agent and

chemical scavenger. The substitution level of polyhydroxylated oil was varied systematically from 0 to 5.0 wt%. Physical and mechanical characterization showed that the starch-filled foams suffered a severe bulk density drop from 40.8 to 15.1 kg/m^3 and catastrophic mechanical failure at 5.0 wt% oil substitution due to thermodynamic phase separation and cell wall thinning. Conversely, the natural zeolite-filled foams maintained highly stable bulk densities (ranging between 41.3 and 43.1 kg/m^3) across all substitution levels, with compressibility rising to 3.19×10^{-8} m^3/Pa at 5.0 wt% oil. Optical microscopy confirmed that natural zeolite acts as an exceptional cell stabilizer, facilitating uniform heterogeneous bubble nucleation and adsorbing residual heavy metals and acidic impurities from the waste oil. These results demonstrate that natural zeolite provides a robust physical and chemical framework that permits high-ratio waste-oil valorization without compromising the mechanical performance of flexible polyurethane foams.

Keywords: Polyurethane, Used engine oil, Oil polyol, Starch, Natural zeolite

Introduction

Flexible polyurethane foams (FPUFs) represent a highly versatile class of porous polymeric materials, widely utilized in automotive seating, domestic furniture, thermal insulation, packaging, and acoustic dampening due to their lightweight nature, high strength-to-weight ratio, and energy-absorbing viscoelastic behavior. Traditionally, the three-dimensional network of FPUFs is formed through a highly exothermic, simultaneous gelation and blowing reaction between polyfunctional isocyanates and polyether polyols. However, the industrial reliance on petroleum-derived polyols presents critical environmental and economic challenges, including greenhouse gas emissions, depleting fossil reserves, and fluctuating global crude oil prices. This has prompted intense scientific interest in developing sustainable, bio-based, or waste-derived polyol alternatives [1].

Concurrently, the rapid growth of the transportation and industrial sectors has led to the generation of millions of metric tons of used engine oil (UEO) globally [2,3]. Raw engine oil consists of a base lubricating mineral oil blended with additives (such as detergents, anti-wear agents, and viscosity index improvers) [4]. During engine operation, high-temperature thermal stress and mechanical shear degrade these additives, leading to the accumulation of hazardous components, including polycyclic aromatic hydrocarbons (PAHs), heavy metals (such as calcium, zinc, lead, and iron), and highly acidic oxidation products. Due to its fluidity, improperly discarded UEO easily migrates through ecosystems, contaminating soil and groundwater, destroying microbial communities, and showing severe phytotoxic effects, such as reducing plant growth and causing chromosomal abnormalities in flora. In developing nations, such as Yemen, where waste management infrastructure is constrained, UEO is frequently dumped into open gutters, soil, and waterways [5]. Conventional re-refining and combustion processes are energy-intensive and can release airborne pollutants. Reclaiming and chemically functionalizing UEO as a polymer precursor therefore represents a valuable waste-to-wealth pathway within a circular economy framework.

The primary obstacle to utilizing UEO in polyurethane chemistry is its chemical structure, which consists of long-chain aliphatic and aromatic hydrocarbons that lack the active hydroxyl (−OH) functional groups

required to form urethane linkages. However, the thermal and mechanical stress experienced by lubricating oil in internal combustion engines results in localized cracking, generating unsaturated carbon-carbon double bonds (alkenes) along the hydrocarbon chains [6]. By utilizing an acid-catalyzed hydration reaction, these alkenes can be chemically modified to introduce secondary hydroxyl groups, converting the non-polar hydrophobic waste oil into a polar, reactive polyhydroxylated organic extender.

Despite successful chemical functionalization, replacing commercial polyols with waste-derived extenders often compromises the foaming kinetics, cell morphology, and ultimate mechanical performance of FPUFs. To address these structural deficiencies, auxiliary particulate fillers are commonly introduced into the foam matrix [7]. Natural starch has been widely investigated as a biodegradable organic filler because of its abundant hydroxyl groups, which can promote partial biodegradability and alter physical properties. Alternatively, natural zeolites—porous crystalline aluminosilicates characterized by highly ordered channels and cavities—have gained traction as inorganic functional fillers [8]. Zeolites can act as highly efficient heterogeneous nucleating agents, regulating cell size distribution and density [9]. Moreover, zeolites possess strong ion-exchange and adsorption capacities, enabling them to capture residual heavy metals and acidic impurities from the waste oil, preventing catalyst deactivation during polymerization [4].

This study systematically investigates the physical, mechanical, and morphological characteristics of modified flexible polyurethane foams prepared by partially replacing commercial polyether polyols with polyhydroxylated UEO (0 to 5.0 wt%). By comparing a 1.0 wt% starch-filled composite series with a 1.0 wt% natural zeolite-filled series, this work evaluates the limits of waste-oil substitution and details the physical and chemical mechanisms by which natural zeolite stabilizes the porous cellular framework of polyurethane.

Theoretical and Chemical Framework

The synthesis of flexible polyurethane foams is governed by two competing reaction pathways: the gelling (polyaddition) reaction and the blowing (gas-producing) reaction [10].

The Gelling Reaction

The gelling reaction involves the nucleophilic attack of the hydroxyl groups (−OH) of the polyol and the polyhydroxylated UEO on the electrophilic carbon of the isocyanate groups (−NCO) in toluene diisocyanate (TDI). This step leads to the formation of urethane linkages that build the covalent polymer network [10]:

$$\text{R-NCO} + \text{R'-OH} \rightarrow \text{R-NH-CO-O-R'}$$

The Blowing Reaction

Concurrently, the blowing reaction occurs when TDI reacts with water (the chemical blowing agent) to yield an unstable carbamic acid intermediate. This intermediate spontaneously decomposes into carbon dioxide (CO_2) gas and a primary amine. The generated CO_2gas diffuses into nucleated micro-bubbles, expanding the polymerizing liquid mixture into a cellular foam [11]:

$$\text{R-NCO} + H_2O \rightarrow\rightarrow \text{R-NH}_2 + CO_2 \uparrow$$

The primary amine then rapidly reacts with an adjacent isocyanate group to form symmetric urea linkages [11]:

$$\text{R-NH}_2 + \text{R-NCO} \rightarrow \text{R-NH-CO-NH-R}$$

These rigid urea segments hydrogen-bond with one another, self-assembling into hard domains that provide structural stiffness and elastic resilience to the flexible foam matrix.

Acid-Catalyzed Hydration of Alkenes

To enable UEO to participate in the gelling reaction, its thermally cracked unsaturated hydrocarbons must undergo acid-catalyzed hydration. The reaction mechanism proceeds through three distinct steps [12]:

1- **Protonation**: The strong acid catalyst transfers a proton to the carbon-carbon double bond of the alkene, creating a highly reactive carbocation intermediate:

$$\text{R-CH=CH-R'} + H^+ \rightarrow \text{R-CH}^+ - CH_2 - \text{R'}$$

2- **Nucleophilic Attack**: Water, acting as a weak nucleophile, attacks the carbocation, forming a protonated alcohol intermediate [12]:

$$\text{R-CH}^+ - CH_2 - \text{R'} + H_2O \rightarrow \text{R-CH}(OH_2^+)\text{-}CH_2 - \text{R'}$$

3- Deprotonation: A water molecule deprotonates the oxonium ion, regenerating the acid catalyst and leaving a stable secondary hydroxyl group covalently bound to the hydrocarbon chain [12]:

$$R\text{-}CH(OH_2^+)\text{-}CH_2 - R' + H_2O \rightarrow R\text{-}CH(OH)\text{-}CH_2 - R' + H_3O^+$$

Through this mechanism, non-reactive UEO hydrocarbon chains are converted into active polyhydroxylated organic extenders, which are chemically incorporated into the polyurethane network.

Experimental

Materials & Chemicals

Used engine oils (UEO) were collected from several auto oil change shops in Taiz, Yemen. Raw materials used in the production of polyurethane foams, such as TDI, Polyol, Amine catalyst, and Tin catalyst, were supplied by the National Company for Sponge and Plastic Industries, Ltd. in Taiz, Yemen. Natural zeolite was obtained from Yemen Company for Ghee and Soap Industries, Ltd. Resorcinol antioxidant and Ethyl acetate were obtained from Al-Saeed University in Taiz, Yemen. Concentrated sulfuric acid and sodium hydroxide were supplied by Taiz University in Taiz, Yemen.

Pre-treatment and Functionalization of Used Engine Oil

The conversion of hazardous UEO into a polyhydroxylated organic extender was performed through a sequential mechanical and chemical process (Figure 1):

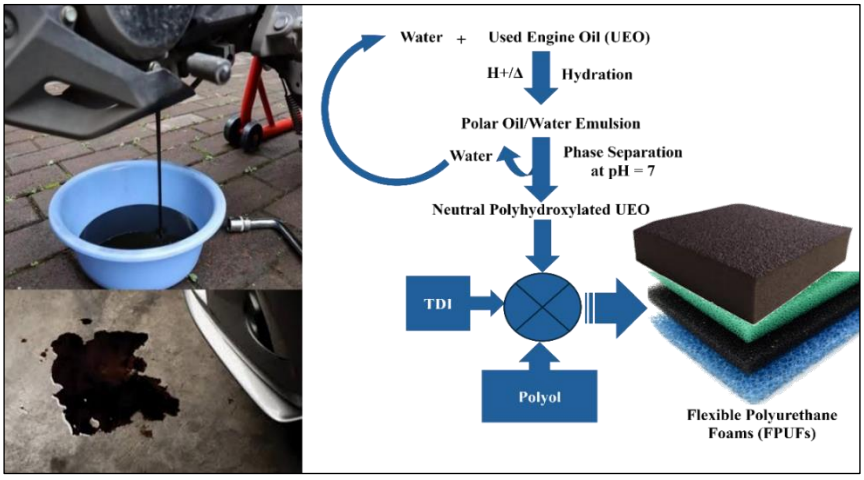

Figure 1: Discharging UEO to the ground (Left), and fabricating polyhydroxylated UEO into the production of FPUFs (Right).

1- **Mechanical Filtration**: The raw UEO was filtered and centrifuged to remove carbonaceous sludge, soot, and macroscopic metal wear debris.
2- **Dehydration**: The filtered oil was heated to 110°C for 60 minutes under continuous stirring to eliminate residual water. This step is critical, as residual moisture would react uncontrollably with TDI during foam synthesis, releasing excess CO_2 gas and causing structural defects.
3- **Acid-Catalyzed Hydration**: The dehydrated UEO was heated to 150°C in an acidic medium. Under these conditions, the acid catalyst protonates the cracked unsaturated alkenes, initiating the nucleophilic addition of water to yield secondary hydroxyl groups along the hydrocarbon backbone. This reaction converts the hydrophobic oil into a polar oil-in-water emulsion.
4- **Neutralization and Phase Separation**: The polar emulsion was neutralized to pH 7 using a dilute sodium hydroxide solution to halt the reaction. This neutralization induced distinct phase separation. The aqueous phase, containing dissolved salts and catalytic acid residues, was separated, leaving the purified, neutral polyhydroxylated UEO (often termed "oil polyol").

Determination of hydroxyl value (OHV) of Used Engine Oil

The hydroxyl value (OHV) expressed in mg KOH/g of untreated and treated UEO samples was determined by the acetylation method according to ASTM D1957. All reagents used were of analytical grade.

Polyurethane Foam Formulation and Polymerization

The foam synthesis was executed using the one-shot free-rise method. To ensure proper stoichiometry and reaction kinetics, the components were divided into two streams: the binder-rich A-side and the polyol-rich B-side.

In industrial polyurethane manufacturing, the A-side typically consists of the diisocyanate component and associated plasticizers, while the B-side contains the polyol blend, catalysts, blowing agents, surfactants, and fillers [1]. This study maintains standard industrial nomenclature, as detailed in the experimental formulations of Table 1 and Table 2.

Table 1.

Formulation of modified flexible polyurethane foams containing 1.0 wt% starch filler.

A- Side Composition		B- Side Composition	
Component	**wt%**	**Component**	**wt%**
TDI	**35.0**	**Polyol blend**	**60.0 - *x***
Ethyl acetate (Plasticizer)	**0.5**	**Water**	**2.2**
		Polyhydroxylated UEO	**x**
		Amine catalyst	**0.2**
		Tin catalyst	**0.2**
		Diethanolamine (DEA)	**0.5**
		Phenolic antioxidant	**0.4**
		Starch	**1.0**
	35.5%		**64.5%**
Total = 100%			

Table 2.

Formulation of modified flexible polyurethane foams containing 1.0 wt% zeolite filler.

A- Side Composition		B- Side Composition	
Component	**wt%**	**Component**	**wt%**
TDI	**35.0**	**Polyol blend**	**60.0 - *x***
Ethyl acetate (Plasticizer)	**0.5**	**Water**	**2.2**
		Polyhydroxylated UEO	**x**
		Amine catalyst	**0.2**
		Tin catalyst	**0.2**
		Diethanolamine (DEA)	**0.5**
		Phenolic antioxidant	**0.4**
		Zeolite	**1.0**
	35.5%		**64.5%**
Total = 100%			

The synthesis procedure was performed as follows:

1- **B-Side Preparation**: The commercial polyether polyol blend, water, polyhydroxylated UEO (*x* wt%), catalysts (amine and organotin), surfactant (silicone oil), diethanolamine crosslinker, and the respective filler (1.0 wt% starch or 1.0 wt% natural zeolite) were mixed at high speed (2000 rpm for 60 seconds) to form a homogeneous B-side emulsion.

2- **Polymerization**: A stoichiometrically determined amount of TDI (pre-blended with the ethyl acetate plasticizer to form the A-side) was rapidly added to the B-side emulsion and stirred vigorously for 5 to 10 seconds.
3- **Molding and Curing**: The reacting mixture was quickly poured into an open mold. The kinetic milestones, including cream time, rise time, and tack-free time, were monitored as the rising foam expanded freely under ambient conditions. The cured foams were stored at room temperature for 24 hours to complete polymerization prior to physical, mechanical, and morphological testing.

Effect of acid- catalyzed hydration of UEO

It has been found that the OHV of UEO before treatment is equal to 11.22±16 mg KOH/g and after the treatment, it reaches 88.77±23 mg KOH/ g. This eight-time increase of OHV by the acid-catalyzed hydration is clear evidence for the stronger incorporation of resulting polyhydroxylated UEO in the FPUF polymerization.

Physical and Mechanical Characterization

The bulk density of the cured composite foams was determined by measuring the ratio of the sample mass to the bulk volume of equal-sized specimens (6.0 cm × 6.0 cm × 6.0 cm):

$$d_{\text{bulk}} = \frac{Wt}{V_f}$$

where $d_{\text{bulk}}$is the bulk density (kg/m^3), $Wt$is the weight of the foam sample (kg), and $V_f$is the bulk volume (m^3).

The mechanical compressibility was defined as the ratio of the change in foam volume (ΔV_f) to the applied axial dynamic pressure (P_{ax}):

$$C = \frac{\Delta V_f}{P_{\text{ax}}}$$

where $C$is the compressibility (m^3/Pa). To determine this value experimentally, a calibrated metal plate was placed on the surface of the foam specimen, and a constant static mass (m_t, ranging from 3.0 to 5.0 kg) was applied to the center. The resulting vertical displacement or thickness change (ΔTh, in cm) was measured. The change in foam volume (ΔV_f, in m^3) was calculated as:

$$\Delta V_f = \Delta Th \times A \times 10^{-6}$$

where A is the surface area of the foam sample (cm^2). The applied axial dynamic pressure (P_{ax}, in Pa) was calculated using the following equation:

$$P_{ax} = \frac{\text{Force applied}}{A} = \frac{m_t \times g}{A \times 10^{-4}}$$

where m_t is the total applied mass (kg), and g is the gravitational acceleration (9.81 m/s^2).

Optical microscopic images (OMIs) were acquired using an optical microscope (Model 6331, Nauborn-Wetzlar) equipped with a 100X magnification objective lens, and were also treated using an ImageJ software, version 1.54g (https://imagej.org) to analyze cell morphology, open-cell ratios, and cell size distribution.

Results and Discussion

Starch-Filled Foam Series (1.0 wt% Starch)

The physical and mechanical properties of the modified flexible polyurethane foams containing 1.0 wt% natural starch and varying levels of polyhydroxylated UEO (0 to 5.0 wt%) are compiled in Table 3 and presented in Figure 2.

Table 3.

Bulk density and compressibility of modified FPUFs containing 1.0 wt% starch filler with x wt% addition of Polyhydroxylated UEO.

Polyhydroxylated UEO Content, x (wt%)	Bulk Density, d_{bulk} (kg/m^3)	Compressibility, C (m^3/Pa)
0	40.8	2.16×10^{-8}
1.0	41.0	2.25×10^{-8}
2.0	41.2	2.11×10^{-8}
3.0	30.2	2.44×10^{-8}
5.0	15.1	4.89×10^{-9}

The starch-filled composite foams maintained highly stable bulk densities (around 41.0 kg/m^3) and consistent mechanical compressibility (2.11×10^{-8} to 2.25×10^{-8} m^3/Pa) at low oil substitution levels ($x \leq 2$ wt%). Within this low concentration range, the reactive secondary hydroxyl groups of the polyhydroxylated UEO reacted successfully with TDI, integrating the oil into the polyurethane network without disrupting the foam structure [13,14].

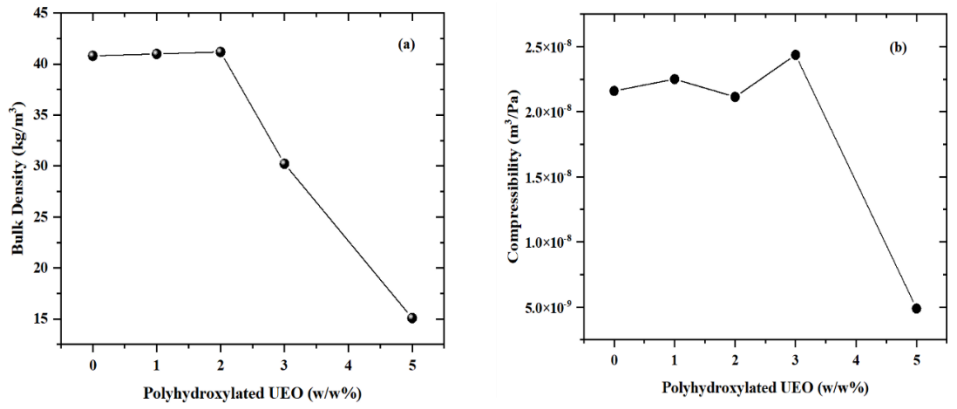

Figure 2: Variation of (a) bulk density and (b) compressibility of modified FPUFs containing 1.0 wt% starch filler as a function of *x* wt% addition of Polyhydroxylated UEO.

However, when the oil substitution level was increased to 3.0 wt%, the bulk density decreased to 30.2 kg/m^3, while the compressibility rose to 2.44×10^{-8} m^3/Pa. At 5 wt% substitution, the bulk density fell sharply to 15.1 kg/m^3, and the compressibility dropped to 4.89×10^{-9} m^3/Pa, indicating physical and mechanical collapse.

This severe structural degradation at higher oil concentrations is due to the chemical and thermodynamic incompatibility between the natural starch filler and the hydrophobic segments of the modified oil [14]. Starch is a highly polar, hydrophilic biopolymer containing numerous primary and secondary hydroxyl groups [15]. In contrast, functionalized UEO, despite carrying secondary hydroxyl groups, consists primarily of long, highly non-polar aliphatic hydrocarbon chains. At a high oil content (5.0 wt%), the interfacial tension between the hydrophilic starch particles and the hydrophobic oil-modified polymer phase increases, leading to localized phase separation in the rising foam [16].

Additionally, the steric hindrance of the large starch granules disrupts the alignment of the polymerizing molecular chains, thinning the bubble walls and struts. Because the secondary hydroxyl groups on the UEO are less reactive than the primary hydroxyl groups on commercial polyether polyols, the gelation reaction is slowed [15,17]. Consequently, the polymer struts do not gain sufficient strength to support the expanding CO_2 gas, leading to cell wall rupture, bubble coalescence, and macroscopic collapse of the cellular

network. This yields an ultra-lightweight, collapsed material with very low compressibility that "bottoms out" under load.

Zeolite-Filled Foam Series (1.0 wt% Natural Zeolite)

The physical and mechanical properties of the flexible polyurethane foams containing 1.0 wt% natural zeolite and varying levels of polyhydroxylated UEO (0 to 5.0 wt%) are presented in Table 4 and Figure 3 as well.

Table 4.

Bulk density and compressibility of modified FPUFs containing 1.0 wt% natural zeolite filler with x wt% addition of Polyhydroxylated UEO.

Polyhydroxylated UEO Content, x (wt%)	Bulk Density, d_{bulk} (kg/m^3)	Compressibility, C (m^3/Pa)
0	40.8	2.16×10^{-8}
1.0	42.3	1.25×10^{-8}
2.0	43.1	1.01×10^{-8}
3.0	41.3	2.34×10^{-8}
5.0	42.4	3.19×10^{-8}

The natural zeolite-filled foams exhibited remarkable physical stability, with bulk densities remaining highly stable between 41.3 and 43.1 kg/m^3 across all oil substitution levels. The mechanical compressibility trend showed a two-stage profile: it initially decreased at 1.0 and 2.0 wt% oil (reaching a minimum of 1.01×10^{-8} m^3/Pa), indicating structural stiffening, and then rose to 3.19×10^{-8} m^3/Pa at 5.0 wt% oil, reflecting restored flexibility and elasticity.

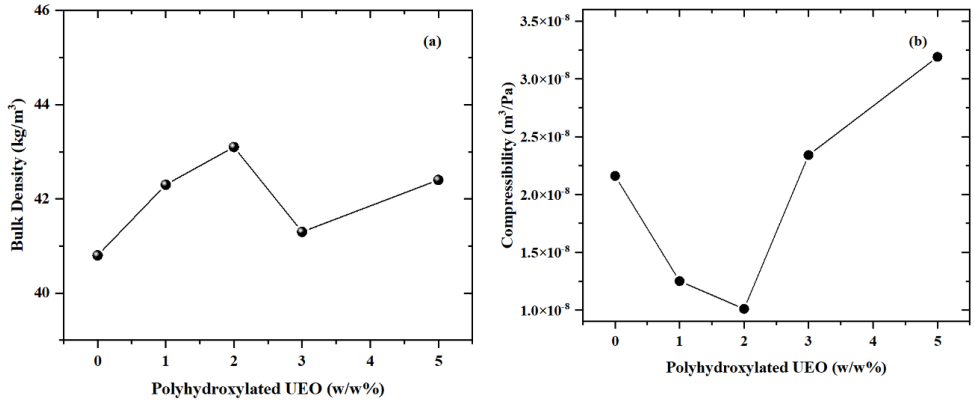

Figure 3: Variation of (a) bulk density and (b) compressibility of modified FPUFs containing 1.0 wt% natural zeolite filler as a function of *x* wt% addition of Polyhydroxylated UEO.

The stabilization provided by natural zeolite can be attributed to its role as a physical nucleating agent and a chemical scavenger. Zeolites possess a rigid, three-dimensional aluminosilicate framework with a high specific surface area and regular micro-cavities. During the one-shot foaming process, the highly porous surfaces of the zeolite particles act as heterogeneous nucleation sites, lowering the free energy barrier for bubble formation. As CO_2 gas is generated, it nucleates uniformly at these sites, producing a high density of small, uniform bubbles and preventing the bubble coalescence that typically leads to cell collapse [18].

Furthermore, natural zeolites are outstanding cation exchangers and adsorbents, capable of capturing residual wear metals (such as calcium, zinc, and iron) and acidic degradation products (such as carbonic acids) present in the functionalized UEO [3]. In the starch-filled series, these free metallic and acidic impurities can coordinate with and deactivate the amine and organotin catalysts, disrupting the balance between the gelling and blowing reactions. By chemically immobilizing these contaminants, natural zeolite maintains optimal catalyst activity and ensures complete, stable polymerization of the polyurethane struts [4].

At low oil concentrations (1.0 to 2.0 wt%), the rigid, crystalline zeolite particles restrict the mobility of the polyurethane chains, increasing stiffness and reducing compressibility. At higher oil concentrations (3.0 to 5.0 wt%), the plasticizing effect of the flexible hydrocarbon segments in the polyhydroxylated UEO becomes dominant, overcoming the steric constraints of the zeolite particles to yield a flexible, resilient foam with high compressibility and a stable bulk density.

Morphological Analysis and Cell Structure Evolution

Optical microscopic imaging at 100X magnification provided structural evidence supporting the observed physical and mechanical trends.

Cell Morphology in the Starch Series

Figure 4 shows OM images at 100X magnification for modified FPUFs containing 1.0 wt% starch filler with the addition of polyhydroxylated UEO. The cell structure of the starch-filled foams was highly dependent on the oil concentration:

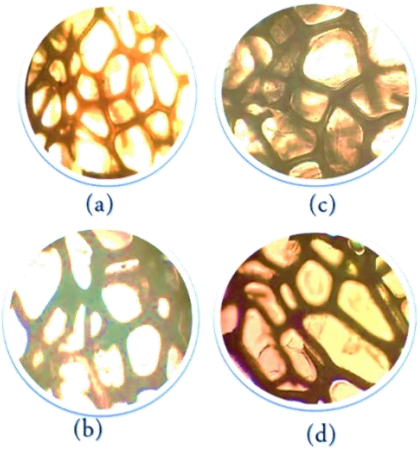

Figure 4: OM images at 100X magnification for modified FPUFs containing 1.0 wt% starch filler with the addition of polyhydroxylated UEO; (a) *x*=1.0 wt%; (b) *x*=2.0 wt%; (c) *x*=3.0 wt%; (d) *x*=5.0 wt%.

- At 1.0 wt% and 2.0 wt% oil, the foam displayed a uniform, closed-to-open cell morphology with well-defined polyhedral cell structures and intact struts.
- At 3.0 wt% oil, the cell size distribution broadened, and the cell walls showed visible thinning and minor localized rupture.
- At 5.0 wt% oil, the morphology revealed severe cellular collapse. The cell walls were heavily ruptured, and the struts were fragmented, leading to massive bubble coalescence and large, irregular voids. This structural collapse explains the dramatic drop in density and compressibility observed in physical testing.

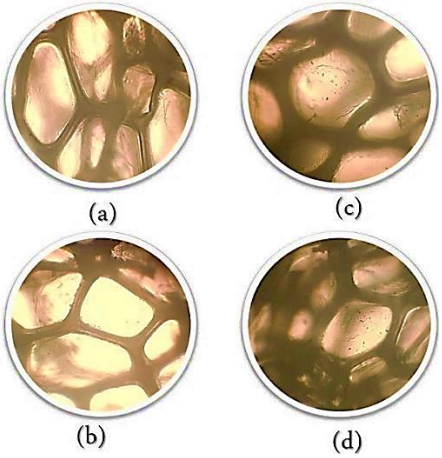

Figure 5: OM images at 100X magnification for modified FPUFs containing 1.0 wt% natural zeolite filler with the addition of polyhydroxylated UEO; (a) *x*=1.0 wt%; (b) *x*=2.0 wt%; (c) *x*=3.0 wt%; (d) *x*=5.0 wt%.

Cell Morphology in the Zeolite Series

In contrast, the natural zeolite-filled foams maintained excellent structural integrity across all formulations. OM images at 100X magnification for modified FPUFs containing 1.0 wt% natural zeolite filler with the addition of polyhydroxylated UEO are illustrated in Figure 5:

- At 1.0 wt%and 2.0 wt%oil, the foam exhibited a fine, uniform cell structure with thick struts, corresponding to the observed structural stiffening and lower compressibility.
- At 3.0 wt% and 5.0 wt% oil, the cell structure remained highly uniform and intact, with no evidence of cell collapse or void formation. The zeolite particles were well dispersed within the polymer struts, providing physical reinforcement and maintaining cell wall stability during gas expansion [14]. This uniform open-cell structure preserves the density and mechanical resilience of the foam, demonstrating that natural zeolite enables successful polyol substitution up to 5.0 wt%.

Conclusions

This study demonstrates a successful circular economy approach by converting toxic, hazardous used engine oil into a reactive polyhydroxylated organic extender to partially replace petroleum-derived polyols in flexible polyurethane foams. Through acid-catalyzed hydration, reactive secondary hydroxyl groups were introduced into the cracked hydrocarbon chains of the used engine oil, enabling its chemical incorporation into the polyurethane network.

The choice of filler played a decisive role in the foam's physical and mechanical performance:

- In the starch-filled series (1.0 wt% starch), thermodynamic phase separation and cell wall thinning led to cell rupture and structural collapse at 5.0 wt% oil substitution, limiting the practical substitution threshold to $\leq$ 2 wt%.
- In the natural zeolite-filled series (1.0 wt% zeolite), the foam maintained highly stable bulk densities and excellent mechanical flexibility up to the maximum tested substitution of 5.0 wt%. Natural zeolite acts as an active physical nucleating agent and a chemical scavenger, facilitating uniform heterogeneous bubble nucleation and adsorbing residual impurities from the waste oil.

These findings indicate that natural zeolite enables higher levels of waste-derived oil substitution, providing a viable pathway for the clean, cost-effective manufacture of flexible polyurethane foams while reducing the environmental hazards associated with used engine oil.

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References

[1] Tomon, T. R. B., Omisol, C. J. M., Aguinid, B. J. M., Sabulbero, K. X. L., Alguno, A. C., Malaluan, R. M., & Lubguban, A. A. (2024). A novel naturally superoleophilic coconut oil-based foam with inherent hydrophobic properties for oil and grease sorption. *Scientific reports*, 14(1), 14223. https://doi.org/10.1038/s41598-024-64178-2.

[2] Armioni, D. M, Ratiur, S. A., Ionel, I., & Vasilescu, M. (2025). The Potential of Using Biomaterials for Sustainable Treatment of Used Engine Oil. *Annals of Faculty Engineering Hunedoara – International Journal of Engineering*, February, 2025. https://annals.fih.upt.ro/pdf-full/2025/ANNALS-2025-1-14.pdf

[3] Alattabi. I. A., Almudhafar, S. M., Jafar, I., Al-Naffakh, J., & Almayahi, B. A. (2025). Environmental and geo-life assessment to produce alternative fuel from plastic waste and used motor oil: a sustainable energy approach for Iraq. *International Journal of Aquatic Research and Environmental Studies*, 5(S1), 95–109. https://doi.org/10.70102/IJARES/V5S1/5-S1-11.

[4] Elamin, E.O. (2019). Recycling of Used Engine Oil. M.Sc. Thesis, *International University of Africa*. https://scispace.com/pdf/recycling-of-used-engine-oil-12xn2zod1c.pdf.

[5] Dhanya, V. (2020). Removal of Used Engine Oil by a Novel Lab Scale Bioreactor, *J. Pure Applied Microbiology*, 14(1), 509–516. https://doi.org/10.22207/JPAM.14.1.53.

[6] Ahmad, I., Khan, R., Ishaq, M., Khan, H., Ismail, M., Gul, K., & Ahmad, W. (2016). Catalytic Pyrolysis of Used Engine Oil over Coal Ash into Fuel-like Products. *Energy Fuels*, 30(1), 204–218. https://doi.org/10.1021/acs.energyfuels.5b02316.

[7] Nguyen, T. H., Huynh, D. N. L., Tran, T. H. C. X. T., Le, H. C., & Pham, N. D. K. (2026). Rice straw and rice husk-based absorbents for oil spill response. *Journal of King Saud University – Science*, 38,17672025. https://doi.org/10.25259/JKSUS_1767_2025.

[8] JP4606789B2. Method for producing flexible slab polyurethane foam. *Google Patents*, https://patents.google.com/patent/JP4606789B2/en.

[9] Fierascu, R. C., Lungulescu, E.-M., Fierascu, I., Stan, M. S., Voinea, I. C., & Dumitrescu, S. I. (2023). Metal and Metal Oxide Nanoparticle Incorporation in Polyurethane Foams: A Solution for Future Antimicrobial Materials? *Polymers*, 15(23), 4570. https://doi.org/10.3390/polym15234570.

[10] Latif, U., & Dickert, F. L. (2011). Conductometric Sensors for Monitoring Degradation of Automotive Engine Oil. *Sensors*, 11(9), 8611–8625. https://doi.org/10.3390/s110908611.

[11] US20070110984A1. Process for producing wood/plastic composites and foams using foaming agents containing zeolite, and wood/plastic composites and foams produced thereby. *Google Patents*, https://patents.google.com/patent/US20070110984A1/en.

[12] Wade, L. G., & Simek, J. W. (2017). *Organic Chemistry* (9th ed., pp. 359–406). Pearson.

[13] Lim, C. C. N. D., Ong, M. J. H., Wu, M., Lee, C.-L. K., & Choong, P. S. (2024). Emerging Trends in Nonisocyanate Polyurethane Foams: A Review. *ACS Engineering Au*, 4(6), 493–518. https://doi.org/10.1021/acsengineeringau.4c00026.

[14] Al-Areqi, N. A. S., Al-Alas, A., Al- Absi, A. A. A. M., Al-Shameri, H. A. A. H., & Al-Hashedi, Hudhaifa H. A. A. (2021). Study on Development and Quality Improvement of Flexible Polyurethane Foams by Substitution of Polyol with Zeolite and/or Unrefined Oils. *International Journal of Scientific & Engineering Research*, 12(4), 115–120.

[15] Mcclurg, R. (2004). Design criteria for ideal foam nucleating agents. *Chemical Engineering Science*, 59, 5779–5786. https://doi.org/10.1016/j.ces.2004.06.025.

[16] Jin, F.-L., Zhao, M., Park, M., & Park, S.-J. (2019). Recent Trends of Foaming in Polymer Processing: A Review. *Polymers*, 11(6), 953. https://doi.org/10.3390/polym11060953.

[17] Gama, N. V., Ferreira, A., & Barros-Timmons, A. (2018). Polyurethane Foams: Past, Present, and Future. *Materials (Basel, Switzerland)*, 11(10), 1841. https://doi.org/10.3390/ma11101841.

[18] Meimei, H., Xiaolong, L., Yanhui, Z., Bei, Z., & Wang Fuming. (2021). Experimental Study of Polyurethane Grout Diffusion in a Water-Bearing Fracture. *Journal of Materials in Civil Engineering*, 33(3), 04020485. https://doi.org/10.1061/(ASCE)MT.1943-5533.0003612.