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# Clove Powder and Oil Effects on Polyurethane Foam Density, Crystallinity, and Antimicrobial Activity

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## ABSTRACT

Polyurethane (PU) foam is widely used as an insulation material; however, its porous structure increases susceptibility to microbial growth. This study compares the effects of clove leaf powder and clove oil (*Syzygium aromaticum*) as natural additives on the density, crystallinity, and antimicrobial activity of PU foam. PU foam was synthesized via a one-shot method using a polyol-to-isocyanate ratio of 1:1.6, followed by the incorporation of clove leaf powder (0–5 wt%) and clove oil (0–15 wt%). Density was measured, crystallinity was analyzed using Differential Scanning Calorimetry (DSC), and antimicrobial activity was evaluated against *Escherichia coli*. The addition of clove leaf powder decreased density due to non-uniform pore formation, whereas clove oil increased density, reaching approximately 0.067 g/cm³ at 15 wt%. Crystallinity increased with both additives; however, clove oil provided a more consistent enhancement, while clove leaf powder increased crystallinity up to ~15%. Antimicrobial activity was observed in all modified samples, with enhanced antimicrobial effectiveness for clove oil. Overall, clove oil outperforms clove leaf powder in improving the structural and antimicrobial properties of PU foam.

**Keywords**: Polyurethane foam, clove oil, clove leaf powder, crystallinity, antimicrobial activity

## INTRODUCTION

Polymer-based materials, particularly polyurethane (PU), continue to play an important role in industrial applications due to their versatility and tunable properties. The properties of polyurethane foam are not only governed by formulation but also by processing parameters, which play a critical role in determining cell morphology and overall performance (Węgrzyk et al., 2023a). Among various polyurethane products, rigid polyurethane foam accounts for a significant portion of total polyurethane consumption, especially in insulation applications (Członka et al., 2020). This is due to its low thermal conductivity, typically ranging from 0.018 to 0.025 $W \cdot m^{-1} \cdot K^{-1}$, making it highly efficient as an insulating material (Oktariani et al., 2024).

Despite these advantages, polyurethane foam has limitations related to microbial contamination. Its porous structure and exposure to humid environments facilitate the growth of microorganisms such as *Escherichia coli* (Czel et al., 2021). This may lead to degradation of material performance and potential health risks. Therefore, the modification of polyurethane foam with antimicrobial agents has become an important research focus (Członka et al., 2020). Previous studies have demonstrated that polyurethane-based composites can be engineered to exhibit antibacterial properties through the incorporation of functional additives, which influence microbial interaction and surface activity (Kasi et al., 2022).

The development of antifungal polyurethane foam is focused on preventing fungal infections and inhibiting their growth. Since the proliferation of these microorganisms can cause health issues in humans during use, the incorporation of additives is necessary to suppress their growth and prevent their accumulation (Kośny & Yarbrough, 2022). The use of synthetic antimicrobial additives has raised environmental and health concerns, leading to increased interest in natural alternatives. Clove (*Syzygium aromaticum*) is one of the most promising natural additives due to its high content of phenolic compounds such as eugenol, which exhibit strong antimicrobial, antioxidant, and antifungal activities (Chaieb et al., 2007; Członka et al., 2020). As reported by Oktariani et al. (2024), clove leaf contains functional groups similar to clove flower, including hydroxyl (–OH), aromatic C=C, and phenolic groups, which are responsible for antimicrobial activity (Oktariani et al., 2024).

Previous studies have demonstrated that the incorporation of clove-based fillers into polyurethane foam can enhance antibacterial properties. For instance, the addition of clove filler significantly improved bacterial inhibition against *E. coli* and *Staphylococcus aureus* (Członka et al., 2020). However, the effectiveness of clove additives is influenced by their form, concentration, and interaction with the polymer matrix. This is consistent with previous studies reporting that the physical form and compatibility of additives play a crucial role in determining the dispersion and resulting properties of polymer composites (Sienkiewicz & Członka, 2022).

In addition, natural fillers can also affect the thermal and structural properties of polyurethane foam. The incorporation of fillers in composite systems has been widely reported to influence structural and surface properties, depending on the dispersion and interaction within the matrix (Mauliana et al., 2023). Previous studies have shown that the incorporation of inorganic fillers such as natural zeolite can significantly improve the thermal stability and crystallinity of polyurethane foam. The addition of zeolite increases the degree of crystallinity and enhances thermal resistance due to its high thermal stability and porous structure(Oktariani & Sari, 2021). These findings highlight the important role of filler type in modifying the structural properties of polyurethane materials. The study by Oktariani et al. (2024) showed that the addition of clove leaf powder increased crystallinity and glass transition temperature (Tg), indicating stronger interactions between filler and polymer chains (Oktariani et al., 2024).

However, a comparative understanding between different physical forms of clove additives, such as solid (leaf powder) and liquid (essential oil), is still limited. This distinction is important because the physical state of additives can significantly influence dispersion, compatibility, and overall performance of the composite material.

Therefore, this study aims to analyze the effect of clove leaf powder and clove oil on the density, crystallinity, and antimicrobial properties of polyurethane foam, as well as to explain the underlying mechanisms governing these effects.

## MATERIAL AND METHOD

Polyurethane foam was prepared using polyol and methylene diphenyl diisocyanate (MDI) with a ratio of 1:1.6 through a one-shot method. Clove leaf powder was used as a solid filler with concentrations of 0%, 1%, 3%, and 5%

wt, while clove oil was used as a liquid additive with concentrations of 0%, 5%, 10%, and 15% wt.

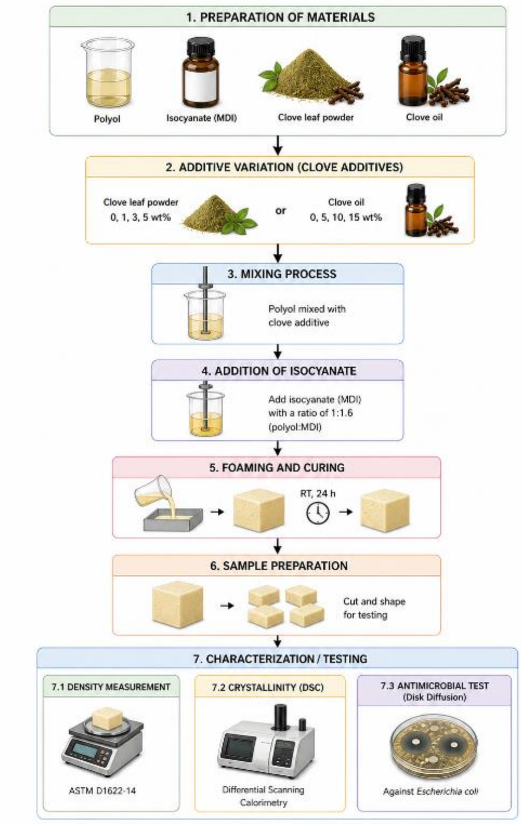

Figure 1. Flow diagram of polyurethane foam preparation and characterization

Fig. 1 illustrates the experimental workflow for preparing and characterizing polyurethane (PU) foam modified with clove-based additives. The process begins with the preparation of raw materials, including polyol, isocyanate (MDI), clove leaf powder, and clove oil, followed by the incorporation of additives at specific concentrations (0–5 wt% for leaf powder and 0–15 wt% for clove oil). The mixture is then homogenized and reacted with isocyanate at a polyol-to-isocyanate ratio of 1:1.6 using the one-shot method, leading to foam formation and curing at room temperature for 24 hours. The mixture was stirred using a mechanical mixer and poured into an open mold, followed by curing at room temperature for 24 hours. This procedure is consistent with previous studies on polyurethane composite preparation (Ludwick et al., 2008; Oktariani et al., 2024).

To ensure reproducibility and enable statistical analysis, each formulation was prepared in triplicate. The cured foams were cut into appropriate dimensions for testing. Density was measured according to ASTM D1622, crystallinity was analyzed by differential scanning calorimetry (DSC), and antimicrobial activity was evaluated against *Escherichia coli* using the disk diffusion method. The results were presented as mean ± standard deviation and analyzed statistically to determine the effect of clove additives on the properties of PU foam. The resulting foam is subsequently cut into appropriate dimensions for testing. Finally, the samples are characterized through density measurement (ASTM D1622), crystallinity analysis using Differential Scanning Calorimetry (DSC), and antimicrobial testing against *Escherichia coli* to evaluate the effect of clove additives on the structural and functional properties of PU foam.

## RESULT AND DISCUSSION

Fig. 2 shows the effect of clove additive type and concentration on the density of polyurethane foam. The density of polyurethane (PU) foam was influenced by both the type and concentration of clove-based additives. Density was measured on three specimens for each formulation, and the results were reported as the average value. For clove leaf powder, the density exhibited a non-monotonic behavior rather than a simple decreasing trend. The density decreased from 55.58 kg/m³ at 0 wt% to 51.05 kg/m³ at 1 wt%, then slightly increased at 3 wt%, before decreasing again to 47.71 kg/m³ at 5 wt%. This fluctuation indicates that the effect of clove leaf powder on foam density was governed by competing mechanisms during foaming. At low loading, solid particles can disturb bubble growth and reduce cell stability, which lowers density. At moderate loading, some particles may act as nucleating sites and temporarily promote a more compact cell structure, resulting in a slight increase in density. However, at higher loading, particle agglomeration and poorer dispersion become more pronounced, producing local structural heterogeneity, irregular cell growth, and greater void formation, which finally reduces density.

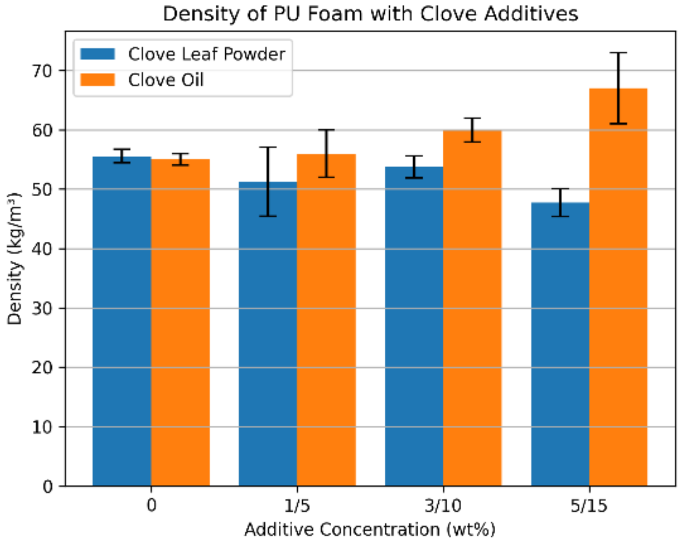

Figure 2. Effect of clove additive type and concentration on the density of polyurethane foam

This behavior is consistent with previous studies reporting that particulate materials can affect foam structure by altering pore formation and introducing heterogeneity within the polymer matrix. Solid fillers may provide active sites for nucleation, but when their dispersion is not uniform, they can also disrupt the formation of stable and homogeneous cells (Dukarska et al., 2022; Oktaviansyah & Piestie Hawa, 2025). In the present study, the irregular response of clove leaf powder suggests that its interaction with the PU matrix was strongly dependent on concentration. At low and intermediate concentrations, the particles may have been partially accommodated within the foam network, whereas at higher concentrations their accumulation likely reduced the uniformity of the expanding cells. As a result, the density of the foam did not change linearly with additive content.

In contrast, the addition of clove oil exhibited a more consistent increase in density with increasing concentration. The density rose from 55.58 kg/m³ at 0 wt% to 66.97 kg/m³ at 15 wt%, corresponding to an increase of approximately 21.8%. This increase can be attributed to the plasticizing and viscosity-modifying effect of clove oil, which slows down foam expansion during the foaming process and promotes the formation of smaller, more compact, and more uniform cells. A higher viscosity system generally restricts excessive gas expansion, thereby producing a denser foam structure. Similar behavior has been reported in previous studies, where processing conditions and matrix viscosity strongly influenced pore formation, cell uniformity, and the final density of polyurethane foam (Węgrzyk et al., 2023b).

Compared with clove leaf powder, clove oil demonstrated a more stable and effective role in increasing foam density. This difference suggests that the physical form of the additive plays an important role in determining the final morphology of PU foam. Liquid additives are generally more compatible with the polymer matrix and can distribute more evenly during mixing, whereas solid fillers may introduce dispersion-related heterogeneity. Therefore, clove oil provided a more controlled effect on foam expansion and structural compactness, while clove leaf powder produced a more variable response due to particle dispersion and agglomeration effects.

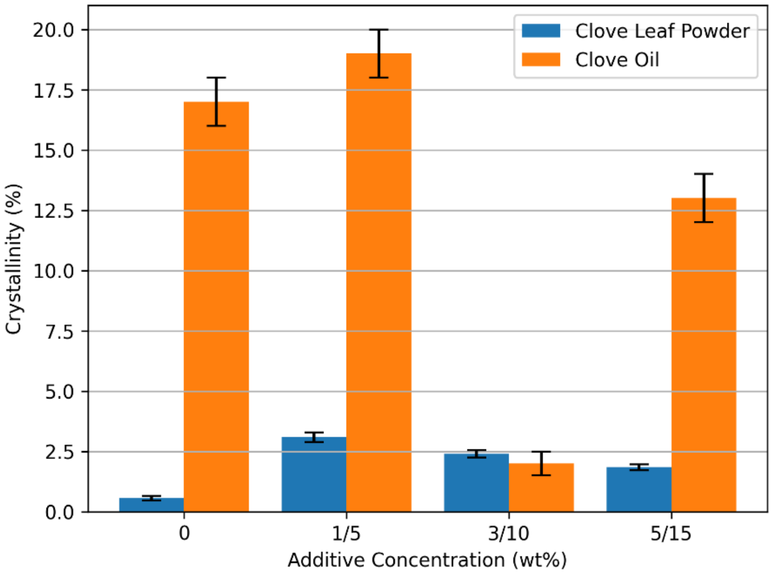

Figure 3. Effect of clove additive type and concentration on the crystallinity of polyurethane foam

Figure 3 shows the effect of clove additive type and concentration on the crystallinity of polyurethane (PU) foam. The crystallinity of PU foam was evaluated using Differential Scanning Calorimetry (DSC), where the degree of crystallinity was calculated based on the melting enthalpy relative to a fully crystalline reference. The results indicate that both clove leaf powder and clove oil influence the crystalline structure of PU foam, with distinct behaviors depending on additive form and concentration.

For clove leaf powder, the crystallinity increased from 0.56% in neat PU foam to 3.09% at 1 wt%, followed by a gradual decrease to 2.39% at 3 wt% and 1.84% at 5 wt%. This trend suggests that low filler loading promotes heterogeneous nucleation, facilitating the organization of polymer chains into more ordered domains. At higher concentrations, however, particle agglomeration and reduced dispersion quality likely restrict chain mobility and disrupt regular packing, resulting in decreased crystallinity. This behavior is consistent with the typical response of particulate fillers in polymer systems, where nucleation enhancement at low loading is offset by structural disruption at higher concentrations.

In contrast, clove oil exhibits a non-monotonic crystallinity profile. The crystallinity increases from 17% in the control sample to 19% at 5 wt%, decreases sharply to 2% at 10 wt%, and then rises again to 13% at 15 wt%. The initial increase at 5 wt% may be attributed to improved molecular mobility and enhanced interaction between clove oil and the PU matrix, which facilitates chain rearrangement and ordering. The significant drop at 10 wt% suggests that excessive plasticization or interference with hard-segment formation disrupts the microphase-separated structure of PU. At 15 wt%, the partial recovery indicates that the system may undergo structural reorganization at higher additive content, allowing some degree of chain ordering to be restored. This behavior highlights the sensitivity of PU crystallization to the balance between chain mobility, phase separation, and additive concentration.

The higher crystallinity observed in clove oil-modified systems reflects a more uniform interaction within the polymer matrix, while the incorporation of clove leaf powder introduces structural heterogeneity that limits ordered chain packing.

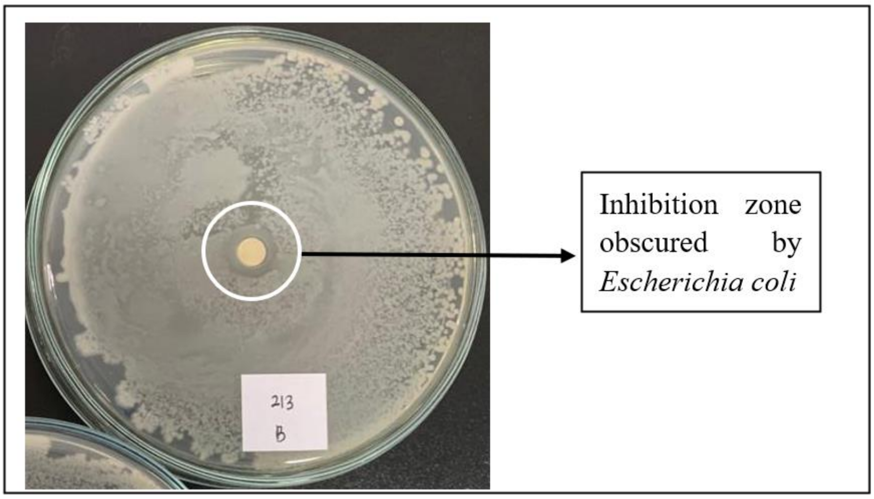

Figure 4. Representative agar plates showing inhibition zones of polyurethane foam modified with clove leaf powder and clove oil after 24 h incubation.

The antimicrobial activity of polyurethane (PU) foam modified with clove-based additives was evaluated using the disk diffusion method against Escherichia coli. The inhibition zone diameter obtained after 24 h of incubation is presented in Figure 4.

The incorporation of clove leaf powder resulted in a gradual increase in inhibition zone diameter, from 10 mm (0 wt%) to 12 mm (5 wt%). A slight plateau was observed at low concentrations (1 wt%), while a more pronounced increase occurred at higher loadings (3–5 wt%).

This trend, as shown in Fig. 5, indicates that increasing the concentration of clove leaf powder enhances antimicrobial effectiveness. The improvement can be attributed to the presence of phenolic compounds such as eugenol, which are known to disrupt bacterial cell membranes and increase cell permeability, ultimately leading to bacterial inhibition.

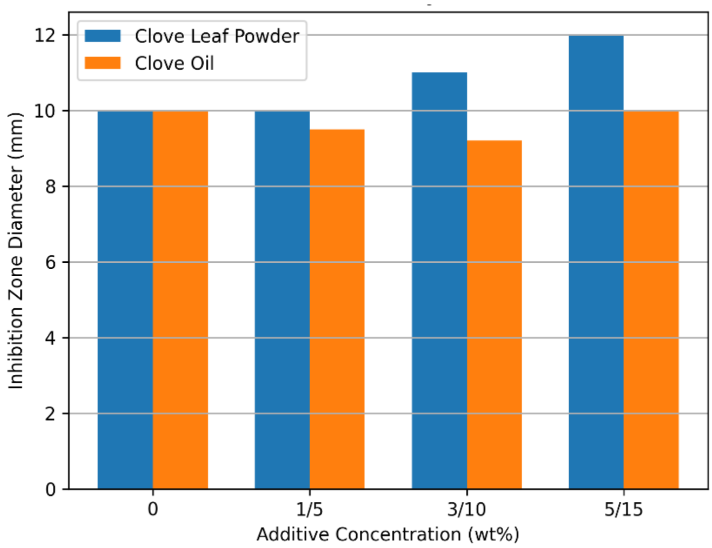

Figure 5. Effect of clove additive type and concentration on the antimicrobial activity of polyurethane foam against *E. coli*.

In contrast, the clove oil-modified samples exhibited relatively stable inhibition zone diameters, ranging from 9.2 to 10.0 mm across all concentrations. A slight decrease was observed at intermediate concentrations (5–10 wt%), followed by stabilization at 15 wt%. This non-linear behavior suggests that although clove oil contains active antimicrobial compounds, its effectiveness within the PU matrix may be influenced by diffusion limitations or interactions with the polymer network. At higher concentrations, the distribution of clove oil appears to become more uniform, resulting in more consistent antibacterial performance.

The classification of antimicrobial activity based on inhibition zone diameter indicates that all samples fall within the "partially active" category (9–12 mm). Despite this, clove leaf powder demonstrates a more pronounced concentration-dependent improvement, whereas clove oil shows a relatively constant behavior.

The agar plate images provide important qualitative support for the quantitative data. As shown in Figure Y, several samples exhibited technical artifacts such as disk displacement, uneven bacterial distribution, and partially obscured inhibition zones. For example, in the clove leaf powder series, certain replicates showed zero inhibition due to disk movement or incomplete zone formation, which may be attributed to airflow-induced vibration in the incubator or non-uniform sample dispersion. Similarly, in the clove oil series, unclear inhibition boundaries were observed in some plates, indicating potential variability in diffusion behavior. These observations highlight that experimental factors, including agar thickness, inoculum distribution, and disk positioning, play a significant role in determining the measured inhibition zone.

This finding is consistent with previous studies reporting that polyurethane-based composites can exhibit significant antibacterial properties depending on the type and distribution of active components within the polymer matrix. The incorporation of functional additives has been shown to enhance bacterial inhibition through improved interaction between the material surface and microbial cells (Kasi et al., 2022). This result suggests that the solid form may act as a more effective reservoir of active compounds within the polymer matrix. However, both additives demonstrate consistent antibacterial activity, confirming the potential of clove-based natural additives for enhancing the functional properties of polyurethane foam.

## CONCLUSION

The results show that the form of clove-based additives significantly affects the properties of polyurethane (PU) foam. Clove leaf powder reduced density due to non-uniform pore formation, while clove oil increased density by producing a more compact structure. Both additives enhanced crystallinity, with clove oil showing a more consistent effect and clove leaf powder exhibiting an optimum at low concentration. All samples demonstrated antibacterial activity against *Escherichia coli* (9–12 mm), categorized as partially active, where clove leaf powder showed a concentration-dependent increase, while clove oil remained relatively stable. These findings indicate that clove oil is more effective in improving structural properties, whereas clove leaf powder provides slightly better antimicrobial performance, highlighting the importance of additive form in polyurethane modification.

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