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INVESTIGATION ON THE INFLUENCE OF WASTE-BASED FILLERS ON THE MECHANICAL AND THERMAL CHARACTERISTICS OF RIGID POLYURETHANE FOAMS

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Abstract

An investigation was conducted to analyze the impact of incorporating coal powder particles at different weight ratios (5, 10, 15, 25, and 35) on the mechanical properties and thermal conductivity coefficient of the polyurethane polymer. The thermal conductivity coefficient of the samples was calculated using Holmarc's Lee's Disc apparatus device. The mechanical properties like compressive, tensile, and bending strengths were measured using a universal machine. The results indicated that increasing the coal powder ratio leads to an improvement in the thermal insulation ability due to a decrease in the value of thermal conductivity. Also, the addition of these percentages led to a rise in the values of the mechanical qualities represented by the compressive strength, especially at the ratio of 25 wt. %, with a value equal to 2.79 MPa (MPa). The flexural resistance and tensile strength increase at a ratio of 35 wt. %, with values equal to 20.4 MPa and 2.86 MPa respectively. The results indicate that the addition of coal powder enhances the ability of thermal conductivity at the ratios (5 %, 10) wt. %, with values equal to 0.119 W/m °C and 0.114 W/m °C, respectively, by increasing the thermal conductivities of the samples. The aim of this study is, investigate the effect of filler used coal powder waste on the mechanical and thermal properties of PU. The filler materials show the advantages of recycling waste. Filler influences the morphology and strengthens the brittleness. Additionally, the technology of polyurethane materials conforms to the use of coal powder. The overall amount of energy used to produce PU composites is decreased when waste of filler is used to partially replace petrochemical components.

Keywords: polyurethane foam, fillers, coal powder (waste), mechanical properties, thermal conductivity, polymer.

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1. Introduction

By incorporating fillers into polymers, the characteristics of the underlying materials can be rapidly and cost-effectively modified. As a result, both the industry and research sectors are showing growing interest in particle-filled polymers. This enables the customization of several of qualities, including as strength, rigidity, electrical and thermal conductivity, hardness, and dimensional stability [1]. Fillers, also known as organic or inorganic are substances, added to the polymer for many reasons like increasing the volume of material plastic, reducing the cost, improving some mechanical properties, and increasing the flexibility for the final product [2–4]. Almost entirely in

the polymer industry, fillers are used to increase dimensional stability, mechanical, thermal, and electrical qualities. Polymer composites find application in diverse sectors such as packaging film, pipes, tubes, window frames, and, more recently, in the automotive industry. They are also utilized as thermal and sound insulation for decks [5–8]. Polymers can improve desirable properties in the final product via using many types of additives including antioxidants, fillers, anti-stabilizing agents, colouring agents, plasticizing agents, stabilizers, and others. Based on their chemical composition and origins, fillers are divided into two categories: inorganic fillers and organic fillers. Polyurethane (PU) is one of the crucial polymers, and it is used in many applications due to its enjoyment of many qualities, such as UV resistance, long-term durability, high resistance to acids and alkalis diluted in seawater. So, the researchers try to add some fillers to improve their properties [9]. Nevertheless, polyurethane (PU) is susceptible to combustion, hence heightening the potential for fire when it is ignited. It exhibits rapid burning and subsequently releases intense heat and noxious, irritating odors. Polyurethane-based fire retardants are used to reduce the risk. Foam fire retardants can be added by changing the chemical structures of the backbone or adding fillers [10]. The main properties of thermoplastic polyurethane with natural fiber, as water absorption, mechanical strength, microstructural and thermal properties were examined. The mechanical tests showed that the tensile strengths of the composite samples with 3 % and 6 % fibers were higher than those of pure polyurethane by 26.8 % and 19.7 %, respectively. The modulus was also higher by 6.6 % and 45.1 % [11]. The effect of adding fillers to cement concrete was 5 % and the results indicated for that the fillers work to reduce the amount of cement used and improve the compressive strength [12]. The study findings indicate that the optimal ratio for achieving the highest tensile strength (89 N/mm^2) and bending strength (148 N/mm^2) in modified polyurethane foam with fly ash is when the filling is 30 % of the foam's weight. The inclusion of fly ash fillers to enhance the mechanical and thermal characteristics of the polyurethane composites. Additionally, the incorporation of the fly ash resulted in a 10 % increase in the compressive strength of the composites, with a value of 220 kPa [13]. The work undertaken by [14] aimed to improve the mechanical characteristics of thermoplastic polyurethane (TPU) through the incorporation of sugar palm fibers as reinforcement. The investigation revealed that composites mixed with 30 % or higher concentrations of alkali-treated fibers exhibited reduced tensile strength compared to the other composites. Composite fibers treated with a ratio 2 wt. % showed a greater tensile modulus and strength of 440 MPa compared to ratio 6 % alkaline-treated fibers. The study by [15] demonstrates an improvement in the mechanical and thermal properties of composites. The composite formulation with a ratio of 40 % sugar palm fiber exhibited the highest tensile strength, flexural strength, and impact strength values, measuring 17.22 MPa, 13.96 MPa, and 15.47 kJ/m^2 respectively. The aim of this study is, investigate the effect of filler used coal powder waste on the mechanical and thermal properties of PU. The filler materials show case the advantages of recycling waste. Filler influences the morphology and strengthens the brittleness. Additionally, the technology of polyurethane materials conforms to the use of coal powder. The overall amount of energy used to produce PU composites is decreased when waste of filler is used to partially replace petrochemical components.

2. Materials and methods

2. 1. Preparation method

Polyurethane was prepared, using trimethylamine (TEA) as the reaction catalyst and in a ratio of 1:1 % by weight of isocyanate and (polyester-polyol) provided by Sigma-Aldrich company. Drops of water were also added to release carbon dioxide gas, which generated cellular voids within the mixture mass **Fig. 1**, shows the chemical composition of polyurethane. As filler, coal powder (coal waste) was used. Large coal pieces were initially collected and then cut into smaller pieces. Subsequently, the small coal pieces were pulverized into a fine powder using an electric grinder of French provenance. To produce fine coal powder with particle sizes of about (125) micrometers using a wire sieve equipped by the American company, Allen-Bradley Sonic Sifter Model L3P, and **Fig. 2** shows coal powder.

Samples of a polyurethane mixture with coal powder were prepared at laboratory temperature. Different weight percentages of powder of coal (5, 10, 15, 25, and 35) wt. %, were added to the polyurethane to form a mixture of polymer and fillers. After that, the final product of the mixture

was placed in a cylindrical, rectangular, and circular mold, and **Fig. 3** displays the measurements of the dimensions for each individual sample in the combination.

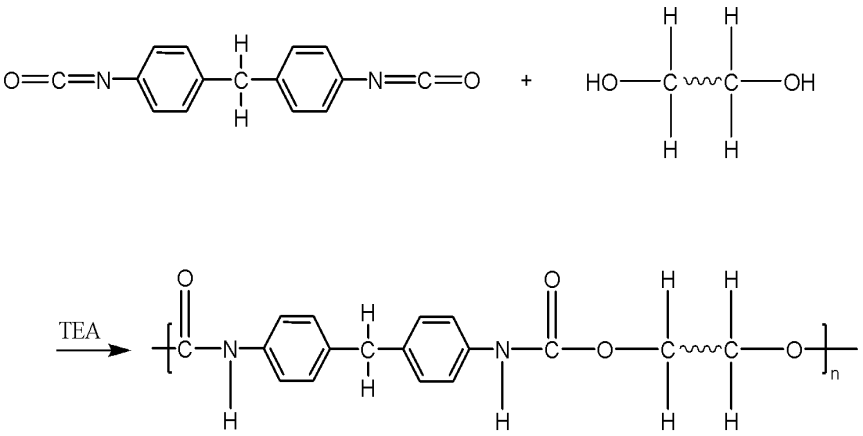
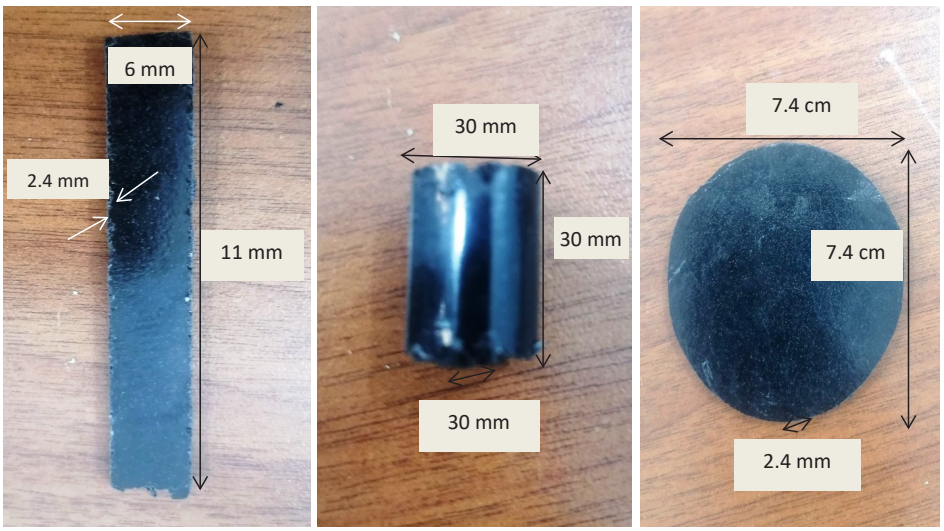


Fig. 1. Chemical composition of polyurethane

Fig. 2. Coal powder (coal powder waste)



a *b* *c*

Fig. 3. Compressive force, tensile strength and thermal conductivity test samples:
a – rectangular; *b* – cylindrical; *c* – circular

2. 2. Mechanical and thermal properties measuring devices

A tensile instrument (Zwick Rell) manufactured in Germany, which measures the mechanical properties (tensile strength, compression resistance, and bending resistance), was used to test the polymers models show a **Fig. 4, *a***. The mechanical properties measuring device (Tensile). Using equation (1) the tensile strength (Q) of the materials can be calculated:

$$Q = \frac{F(\mathrm{N})}{A(\mathrm{mm}^2)}, \tag{1}$$

where F – cutting strength (Newton), A – sample area (mm^2).

The relationship (elasticity modulus) Y = Stress/Strain (MPa) was used to compute the Young modulus.

Thermal conductivity coefficient (K) of the samples was calculated using a done by the Holmarc's Lee's Disc apparatus model HO-AE-LD18 (Kochi, India, Holmarc Opto-Mechatronics Ltd manufactures). **Fig. 4, *b***, shows the Hilmarc's Lee's Disc apparatus for thermal conductivity measurement. Equation (2) was used to calculated thermal conductivity (K) using Fourier's law:

$$K = -\frac{q\,x}{A \cdot T}, \tag{2}$$

where K – thermal conductivity (W/m·°C), q – heat energy (W), T – temperature (K), x – thickness of test specimen (m), and A – area of test specimen (m^2). At the steady state, the thermal conductivity K is calculated using equation (3):

$$K = \frac{MC\dfrac{dT}{dt}}{\pi r^2 (T_2 - T_3)} \times \frac{(r+2h)x}{2(r+h)}, \tag{3}$$

where M – mass of the metallic disc; C – specific heat capacity of the metallic disc; h – height of the metallic disc; r – radius of the metallic disc; dT/dt – rate of cooling of the metallic disc at T_3, (T_2–T_3) – temperature difference across the sample thickness (x); Diameter = 76.2 mm; Thickness = 4–6 mm for sample dimensions.

a *b*

Fig. 4. Devices are used to evaluate the specimens:
a – mechanical properties measuring device (Tensile); *b* – Hilmarc's Lee's Disc apparatus for thermal conductivity measurement

2. 3. Characterization

Using a Japanese- made instrument, the JASCO FTIR-4200, a spectrometric examination of polyurethane (PU) polymers and their composites with coal powder were achieved. Potassium bromide (KBr) tablet powder was used with samples prepared from the mixture to be tested and analyzed by a fourier transforms infrared Spectroscopy device with a wavenumber range from 400 cm^{-1} to 4000 cm^{-1}.

3. Results and Discussion

3. 1. Fourier transforms infrared Spectroscopy (FTIR):

Fig. 5 shows the FT-IR spectra of polyurethanes synthesized with NCO/OH molar ratios of 1:1. The existence of C=O groups is indicated by the presence of the absorption band at 1720 cm^{-1}, whereas the NH stretching vibration is assigned to the absorption band at 3362–3290 cm^{-1}. The presence of C=O and N–H groups suggests that the synthesized polyurethanes have urethane bonds. The signal at 2375 cm^{-1} corresponds to the isocyanate groups (NCO group) is absent in sample spectra, illustrating that the MDI has fully reacted with the hydroxyl groups (OH group) [16–19]. **Table 1** shows the most important absorption bands for polyurethane [20–24].

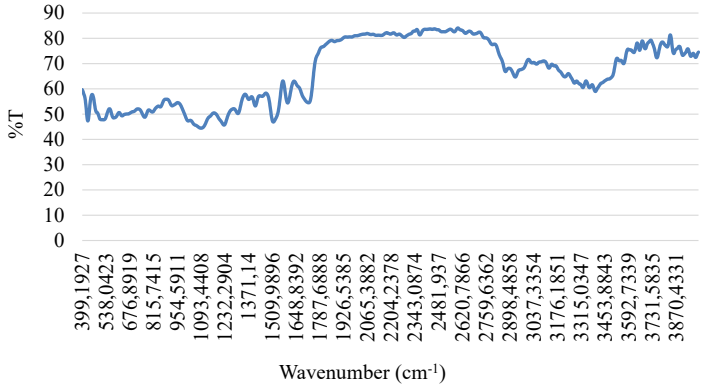


Fig. 5. FTIR spectroscopy of polyurethane

Table 1
Absorption bands for synthesized polyurethane

Functional group	Bond	Wavenumber (cm^{-1})
urethane amino	N–H stretch	3362–3290
carbonyl of urethane	C=O bond	1720
aliphatic methyl symmetric	C–H stretch	2972
aromatic methylene	C–H stretching	3010
aromatic	C–C stretching	1600
urethane group	C–O bond	1079

3. 2. Mechanical properties

The empirical results indicate a decrease in the tensile strength behavior at the 10 % percentage, with a value of 2 MPa [25], which indicates that the polymer mixture at this percentage has high elasticity. When the polymer is in its pure form, that is, at a percentage (0 %), the tensile strength is large, reaching 2.56 MPa, as shown in **Fig. 6**. Whilst the practical results indicate that the maximum tensile strength was at 35 %, with a value of 2.86 MPa. This showed that coal powder at this percentage improves the rigidity through homogeneous distribution between the polymer chains in this mixture, thus enhancing strength. The tensile strength has a direct relationship with the percentage of fillings, especially at weight percentages of 15 % and 35 %. Experimental results indicate a direct relationship between the weight percentages of coal powder and tensile strength behavior, indicating that increasing the weight percentages of fillers enhances the hardness of the mixture [26].

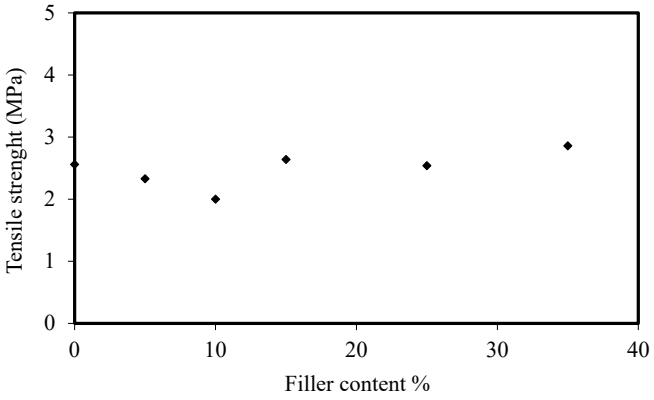


Fig. 6. The relationship between tensile strength and weight ratios coal powder additive to polyurethane polymer

Fig. 7, indicates the effect of filler (coal powder) on the elongation. The behavior of elongation begins at the greatest value at 0 %, which is (6.7 %), and then decreases at the ratio of 10 %, which is (2 %), because the polymer is flexible at this percent and the filler fills the chain polymers, leading to a limitation of chain movement and a decrease in elongation. As indicated by the experimental results, a decrease in the mix elongation at the percentage (25 %) which is (1.8 %), that the polymer has high hardness with low flexibility. The fillers work to reinforce polymers by forming an association with the polymer chains, limiting movement [27].

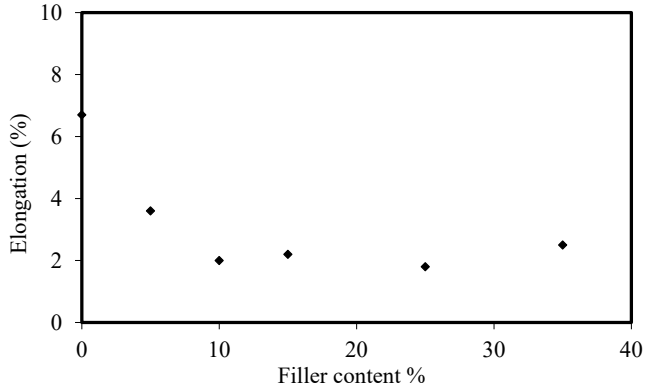


Fig. 7. The relationship between elongation and weight ratios coal powder additive to polyurethane polymer

Fig. 8, depicts the effect of coal powder on the Young modulus (elasticity modulus). The modulus of elasticity is the maximum stress to maximum elongation. The experimental results showed the largest value of the Young modulus at the percentage (25 %), which is (1.41) MPa, while the lowest value of the Young modulus at the ratio (5 %) is (0.64) MPa. There is a positive effect on the modulus of elasticity when the filler (coal) content is increased, because the coal powder increases the hardness of the polymer, and therefore its elasticity decreases [28].

The effect of coal powder on compressive strength is shown in **Fig. 9**, compressive modulus is the capability of the material's resistance to compressive strength perpendicularly for rigid materials. The empirical study indicated that the behavior of the compressive strength is considerably low at the percentage (0 %), about 1.08 MPa, while at the ratio (10 %), the value is about 2.98 MPa, which is the greatest value [29]. It is a fact that the fillers impose interphase uniformity on polymer chains, which affects the polymer's hardness. Though, the compressive modulus behavior starts to decrease at large ratios of the additional fillers, particularly at the ratio (35 %) [30], where the compressive modulus was recorded at nearly (1.99) MPa [32, 33].

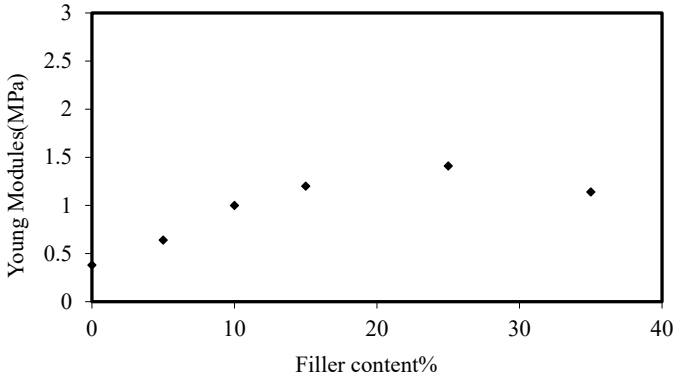


Fig. 8. The relationship between Young modules and weight ratios coal powder additive to polymer

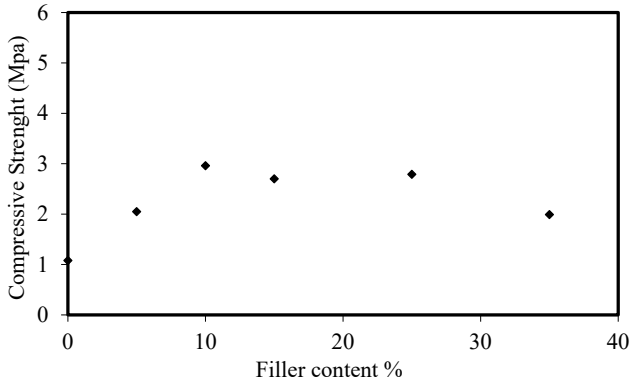


Fig. 9. The relationship between compressive coefficient and weight ratios coal powder added to polyurethane polymer

The correlation between the resistance of bending and weight ratios coal powder added to polyurethane polymer is illustrated in **Fig. 10**, where the bending resistance behavior is shown at the pure polymer (12.6 MPa), i.e., the hardness of the specimen is high and the elasticity is low.

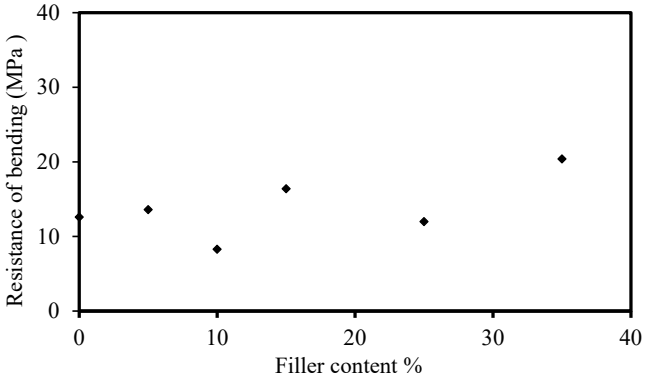


Fig. 10. The relationship between bending resistance and weight ratios coal powder added to polyurethane polymer

The inclusion of the inflexible filler material between the polymer chains constrains the mobility of the chains, leading to reduced flexibility and increased hardness. And then decreases at the ratio 10 %, which is 8.29 MPa, due to the polymer's flexibility at this ratio the filling of coal powder within the primary polymer chains, leading to the limitation of chain movement and

a decrease in bending resistance. The observation is readily apparent when the ratio of filler to polymer chains reaches 35 wt %, indicating a strong affinity between the polymer and filler [32, 33].

3. 3. Thermal conductivity

Fig. 11, shows the relationship of thermal conductivity with the weight ratios of coal powder added to polyurethane. The discrepancy in the results is sometimes attributed to the inhomogeneous distribution of the coal powder with the polymer, which leads to asymmetry in the number of gaps per unit volume, and thus the difference in the results, which leads to a difference in the values of the thermal conductivity coefficient for some unit series samples. The figure also shows that the thermal conductivity coefficient increases with increasing weight percentage of the additives, and that the thermal conductivity coefficient of the pure polymer (without any addition) has the lowest value, which is (0.036 W/m °C), then it starts to increase with the increase in the concentration of the additive, especially at (10 %), which is (0.114 W/m °C) [30]. The increase in the coefficient of thermal conductivity of the polymer with filler concentrations is attributed to two main reasons: First, increasing the concentration of coal powder with the polymer works to reduce the gaps and pores inside the sample. Second, the additives have a higher coefficient of the thermal conductivity than that of polyurethane [34], so they increase the thermal conductivity coefficient of the resulting samples [35].

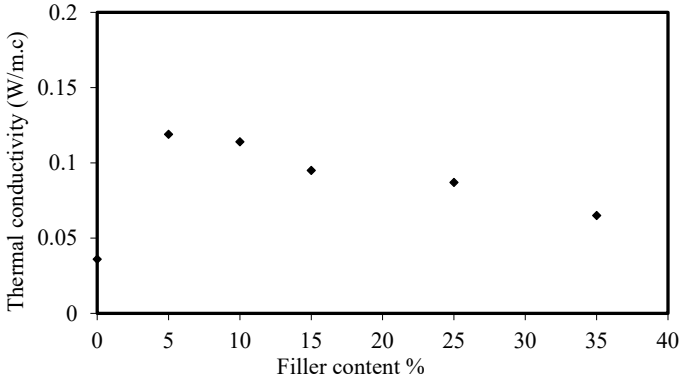


Fig. 11. The relationship between thermal conductivity and weight ratios coal powder added to polyurethane polymer

3. 4. Limitations of the study and future directions

One of the limitations and obstacles to the work in this research is the difficulty of dealing with the base polymer material (polyurethane) because it requires precision in the work, as well as the difficulty of obtaining particles for the additive (coal) with a size of less than 125 μm.

The disadvantage of the resulting samples (fillings with the polymer) used in this research is their shortage of resistance to combustion, as the models prepared from the polymer with the fillings are highly flammable because the polymer and the fillings are highly flammable. This practical study cannot be used in applications that require resistance to combustion.

The research can be developed by supporting it with combustion-resistant fillings to apply the research applications that require combustion resistance. The research can be developed by increasing the thermal and electrical conductivity with increasing the fillers (coal) to apply the research in heat and electrical applications. The additives (coal) have a higher coefficient of thermal conductivity than that of polyurethane [34], so they increase the thermal conductivity coefficient of the resulting samples.

4. Conclusions

The values of thermal conductivity coefficients and compressive strength decrease with increasing weight percentage of additives to the polymer. It is preferable that the coal powder addition percentage does not exceed approximately 35 %. The practical study indicates that

a percentage (35 %) improved the thermal insulation properties. The addition of these percentages also led to an increase in the values of the mechanical properties represented by the tensile strength at (35 %), which is 2.86 MPa, the compressive strength at (25 %), which is 2.79 MPa, and the value increase in flexural resistance at the percentage 35 %, which is 20.4 MPa. Determining the optimal weight ratios at which good thermal insulation is obtained, if it does not affect the mechanical properties in any way. The effect of coal powder on the polymer led to an increase in the coefficient of thermal conductivity in general compared to the pure polymer. The results indicate that the addition of the coal powder lead to enhance the ability of thermal conductivity special at the ratios (5 %, 10) wt. % with values equal to 0.119 W/m ºC and 0.114 W/m ºC respectively, via increasing the value of the thermal conductivities of the samples. Additionally, mixing coal powder filler with polyurethane is a viable method especially in thermal conductivity applications, especially at 10 %.

Conflict of interest

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

Financing

The study was performed without financial support.

Data availability

Data will be made available on reasonable request.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating the current work.

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