

Heat Treatment's Effect on Properties of Polystyrene from Building Demolitions

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Polystyrene (PS) is a synthetic polymer widely used as a packaging material and in thermal insulation of buildings. At end-of-life, there are not many recycling management options of PS because of the reduced incentive and high cost. PS is non-biodegradable, and consequently, the disposal of this product causes serious health and environmental concerns. This study discusses the application of thermal treatment to modify the properties of PS waste foams. Both expanded and extended polystyrene were collected from building demolitions and subjected to different temperature treatments and duration. The effect of the treatment was investigated on the density, structure, glass transition temperature, mechanical properties (hardness, compression strength), thermal conductivity, and sound absorption of treated PS. The results showed that density increased with treatment temperature, which had a corresponding effect on the evaluated properties. The study concluded that thermal treatment is a beneficial way to improve the mechanical properties of PS waste from buildings. However, a trade-off between application and relevance still needs to be ascertained, as the thermal and acoustic insulation properties of PS decreased with the treatment.

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INTRODUCTION

With rapid population growth and urbanization, annual waste generation is expected to continuously increase in the next years. As an integral procedure of waste management, recycling is the final practice of the three Rs system- Reduce, Reuse, Recycle. Recycling depends on the quantity and quality of wastes, the market security for the recovered materials, and the quality of products produced (Nazari *et al.* 2021). Synthetic plastic materials constitute large amounts of wastes. Worldwide plastic production was estimated to be 367 million tons in the year 2020, and that included thermoplastics and thermoset plastics. An analysis of European plastic production shows an estimated quantity of 55 million tons in the same year. The distribution of polystyrene waste in plastics converter demand is rated as 6.1% (Plastics Europe 2021). The commonly seen polystyrene foam includes expanded polystyrene (EPS), extruded polystyrene (XPS), and polystyrene paper, such as food tray and cups.

Polystyrene (PS) is an inexpensive, synthetic, aromatic, thermoplastic polymer made from the monomer styrene and is one of the most widely used plastics. It is soluble in organic solvents, clear, hard, and rather brittle. It possesses a poor barrier to oxygen and water vapor and has a relatively high melting point at 212 °C (Harper 2002). PS has wide applications as protective packaging and insulation in automotive industries, appliances, electronics, food service industries, medical products, *etc.* (Samper *et al.* 2010; EPS Industry Alliance 2016). Foamed PS of low density is used in the form of panels in the building construction industry as thermal insulation and water barrier (Acierno *et al.* 2009). Despite the numerous advantages, there is a growing problem with PS disposal options because of its bulkiness and non-biodegradability. Because it is lightweight compared to its volume, PS occupies a large amount of landfill space and can quickly become a pollution hazard to land and aquatic organisms. Styrene has been linked with adverse health effects in humans and is a known carcinogen (Farrelly and Shaw 2017).

Generally, PS is often not recycled locally like other thermoplastics. This is likely due to the logistics required, since recovered material needs to be transported to a centralized plant. Its low density makes it uneconomical to collect; thus manufacturers cannot obtain sufficient scrap to recycle. Apart from occupying storage space, disposed PS foam yields only a fraction of PS for reuse (about 2%). The PS often has a high level of contamination, especially those used in food packaging applications. Consequently, there is a lack of incentive to invest in PS recycling and an overall increased recycling cost compared to other plastics. Where recycling is possible, reclaimed PS is often converted for reuse into other value-added materials. Currently, there are several methods employed to recycle PS such as thermal, mechanical, and chemical methods. Some of these include mechanical compaction to reduce the volume, reduction in particle sizes, heat extrusion in solid pellets, and catalytic degradation in supercritical solvents (Maharana *et al.* 2007; Kaho *et al.* 2020). However, recyclability can sometimes be a challenge because of the additives used in manufacturing, as these are often undesirable in new recycled products (WEKA 2022).

This situation has led to the development of methods for reusing PS in building construction materials (Aciu *et al.* 2015; San-Antonio-González *et al.* 2015). One of such methods is the thermal treatment of PS to modify the structural properties (Ellouze *et al.* 2020; Kan and Demirboğa 2009). Thermally treated EPS showed improvement in mechanical, tribological, and hygroscopic properties by creation of a polystyrene crust, which protects the cell structure against stretching (Ellouze *et al.* 2020). Density, thermal conductivity, and compressive strength increased when waste EPS was optimally treated at 130 °C for 15 min (Kan and Demirboğa 2009). Most of these studies focused on the utilization and treatment of reclaimed PS as building material composite. Thus, thermal treatment is generally applied when considering PS in new applications that require structural performance than for traditional packaging and insulation purposes. Thermal processes modify the behavior of PS to maximize its service life and mechanical properties. Moreover, untreated PS aggregates in concrete cause segregation in mixing, as they do not adhere to the cement paste and tend to float out of the matrix (Kan and Demirboğa 2009).

The aim of this research was to investigate the effect of heat treatment at different temperatures and durations on physical, mechanical, microstructural, and thermal and acoustic insulation properties of reclaimed EPS and XPS collected from building demolition works.

EXPERIMENTAL

The research material was collected from recovered insulation panels of different origin after building demolition. Four PS types, with nominal thickness of 5 cm, were investigated. Three types corresponded to three colors (white A, blue B, and green C) of XPS, and the fourth type to expanded polystyrene (EPS) of white color (D). The PS types are specified in the European Standards EN 13163 (2001) and EN 13164 (2001). The initial material was cut to prismatic samples of about $1 \times 1 \times 1 \text{ cm}^3$ in dimensions. These samples were heated in a laboratory oven at four different temperatures (80, 100, 120, and 140 °C) for 1, 2 and 4 h. The volume of samples was determined before and after heat treatment by the water displacement method to determine density (Tsoumis 1991). For each parameter combination of temperature and time, five samples of each PS type were used. Moreover, other properties, such as structural, thermal, acoustic, hardness, and compression strength, were determined for treated and untreated samples. For comparison purposes, EPS of white color (E) from packaging material was also subjected to the same treatment conditions and property evaluation. Non-treated and heat-treated PS samples were observed using a scanning electron microscope (SEM) for investigating the effect of heat treatment on the cellular structure of PS polymer. Small pieces were cut from the above samples, mounted on stubs and coated with gold using a sputter device (K550X; Emitech Ltd., Kent, UK) before examining using a XL 30 ESEM (Philips, Eindhoven, Netherlands) operated at 10 kV accelerating voltage, with images recorded digitally. Differential scanning calorimetry (DSC) analyses were performed using a STAR^e DSC 3 system (Mettler-Toledo AG, Schwerzenbach, Switzerland) to determine glass transition temperature. The tests were performed under a dry nitrogen atmosphere (50 ml/min), and the heating rate was 10 °C/min. Hardness was conducted according to EN 13163 (2001a) and EN 13164 (2001b). Compression tests were conducted at cross head speed of 1.5 mm/min according to ASTM D1037 (2013). The acoustic property of the samples was performed according to ISO 10534-2 (1998). The normal-incidence complex acoustic impedance and sound absorption coefficient were measured by using the ACUPRO system (TFAcoustics, LLC, Lexington, KY, USA) in an impedance tube at 6.8 Hz and microphone spacing of 29.21 mm. Samples measuring 34 mm in diameter were used and the test was performed in replicates. The thermal conductivity of the samples was measured using the transient plane source (TPS) method according to ISO 22007-2 (2015). The samples were paired as couples, with each sample having two surfaces. Measurements were performed for a combination of surfaces in the same sample couple.

RESULTS AND DISCUSSION

Density

The results of heat treatment of all PS types are presented in Table 1 and Fig. 1. There was no noticeable change in density between untreated samples and those treated at 80 °C. However, by increasing the temperature of heating from 80 °C, the density increased substantially across all PS samples. Figure 1 shows the density variation in PS samples exposed to different temperature and duration. Temperatures between 80 and 100 °C had little effect on density for all durations (Fig. 1). Kan and Demirboğa (2009) also found that the density of EPS was nearly constant up to 100 °C for 15 min duration. The density increased by 15.3, 11.7, 11.2, 33.1, and 23.1 times after 4 h heating for the PS materials A,

B, C, D, and E, respectively. Percent increase in density for all the samples decreased after the 120 °C treatment. This may be attributed to the increasing flow at the glass transition point of PS (Luo *et al.* 2020).

Table 1. Effect of Heat Treatment on Density of PS After 4 h Heating (mean values ± standard deviations)

Type of PS Material		Density (g/cm ³)				
Code	PS material	Controls	Temperature/ 4 h Duration			
			80 °C	100 °C	120 °C	140 °C
A	XPS (white) demolition	0.032 ±0.0005	0.033 ±0.0017	0.075 ±0.0096	0.297 ±0.0078	0.489 ±0.0308
B	XPS (blue) demolition	0.034 ±0.0003	0.033 ±0.0002	0.045 ±0.0041	0.394 ±0.0238	0.398 ±0.0226
C	XPS (green) demolition	0.030 ±0.0002	0.029 ±0.0005	0.022 ±0.0004	0.342 ±0.0051	0.486 ±0.0497
D	EPS (white) demolition	0.014 ±0.0011	0.012 ±0.0014	0.032 ±0.0032	0.364 ±0.0042	0.464 ±0.0178
E	EPS (white) packaging	0.024 ±0.0028	0.024 ±0.0031	0.039 ±0.0044	0.438 ±0.0367	0.567 ±0.0137

(XPS = Extruded PS; EPS = Expanded PS)

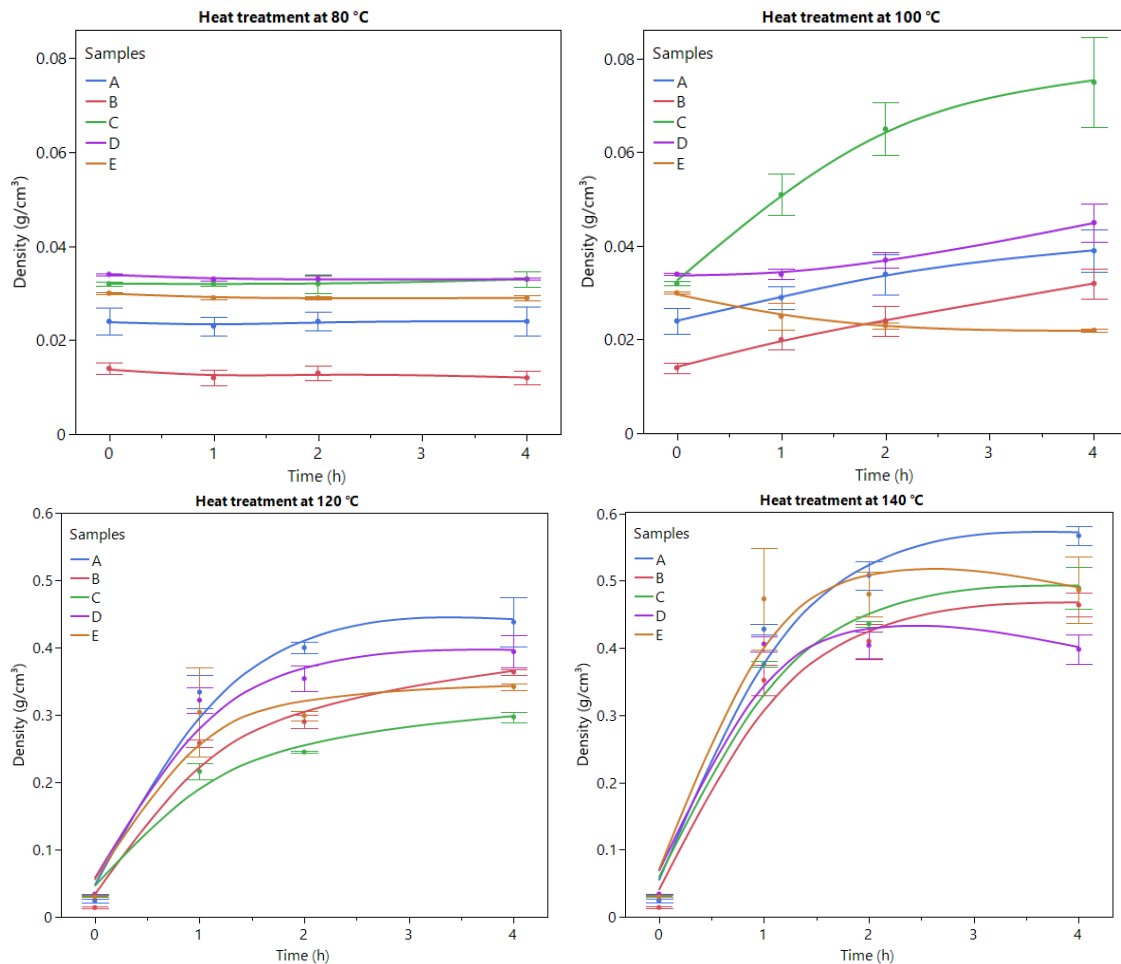


Fig. 1. Effect of heat treatment on density of five PS types at different temperatures and duration. A: XPS (white) demolition; B: XPS (blue) demolition; C: XPS (green) demolition; D: EPS (white) demolition; E: EPS (white) packaging

For the two treatments at 120 and 140 °C, the density increased substantially up to 1 h heating. As expected, the effect of heat treatment at 140 °C was greater than at 120 °C (Fig. 1). Between 1 and 4 h heating at 120 °C, density increased gradually among the samples. However, at 140 °C, samples D and E (EPS) showed a slight decrease in density after 1 h heating. When EPS foam is heated, it transforms into a plastic state. Above 130 °C, it begins to shrink with decreasing density (Kan and Demirboğa 2009). From an energy saving perspective and subsequent cost, as well as a satisfactory increase in density, heat treatment of 2 h at 120 °C was preferred for further property investigation. This treatment resulted in a noticeable increase of density due to a substantial reduction of volume (Fig. 2). As a result of densification, the properties of the materials changed. Since the behavior of the materials was similar, two representative PS types were selected, one XPS (type B) and one EPS (type E), and their properties are discussed in the sections below.



Fig. 2. An example of volume reduction of a mixture of various XPS (type B) and EPS (type E) shredded particles before and after thermal treatment at 120 °C for 2 h (bulk density of untreated particles: 0.015 g/cm³ and bulk density of treated particles: 0.242 g/cm³)

Scanning Electron Microscopy (SEM)

SEM observations of XPS (type B) and EPS (type E) samples showed clear differences in the micro-morphology of their cellular structure before and after heat treatment (at 120 °C), even though the general cellular network structure remained unchanged. For example, non-treated EPS consisted of PS beads that appeared to have a honeycomb type cellular structure (Fig. 3a-c). A similar overall cellular structure could also be seen after the heat treatment (Fig. 3d-f). However, there were considerable micro-morphological changes that occurred after the heat treatment, such as a significant size reduction of the PS beads following the shrinkage of its cells (Fig 3a vs 3d and Fig. 3b vs 3e). Furthermore, thin cell walls of the alveolar cellular structure of untreated PS beads underwent thickening during heat treatment, resulting in more than 50% of thick cell walls (asterisks in Fig. 3e) compared to that of the untreated sample (Fig. 3b). In addition, the thin outer surface layer of each PS bead from untreated samples (arrowheads in Fig. 3a and Fig. 3c) was modified during the heat treatment and transformed into a surface crust, which is a compact and much thicker surface envelop (double-headed arrow in Fig. 3e) having a rough structure (arrow in Fig. 3e and Fig. 3f). Similar micro-structural changes were also observed with XPS samples after heat treatment, such as size reduction (i.e. PS beads and their cells) following cellular shrinkage, thick cell walls and the formation of rough surface crust (Fig. 3g-i; untreated vs Fig. 3j-l; treated). These observations are in line with previous studies on EPS (Ellouze *et al.* 2020).

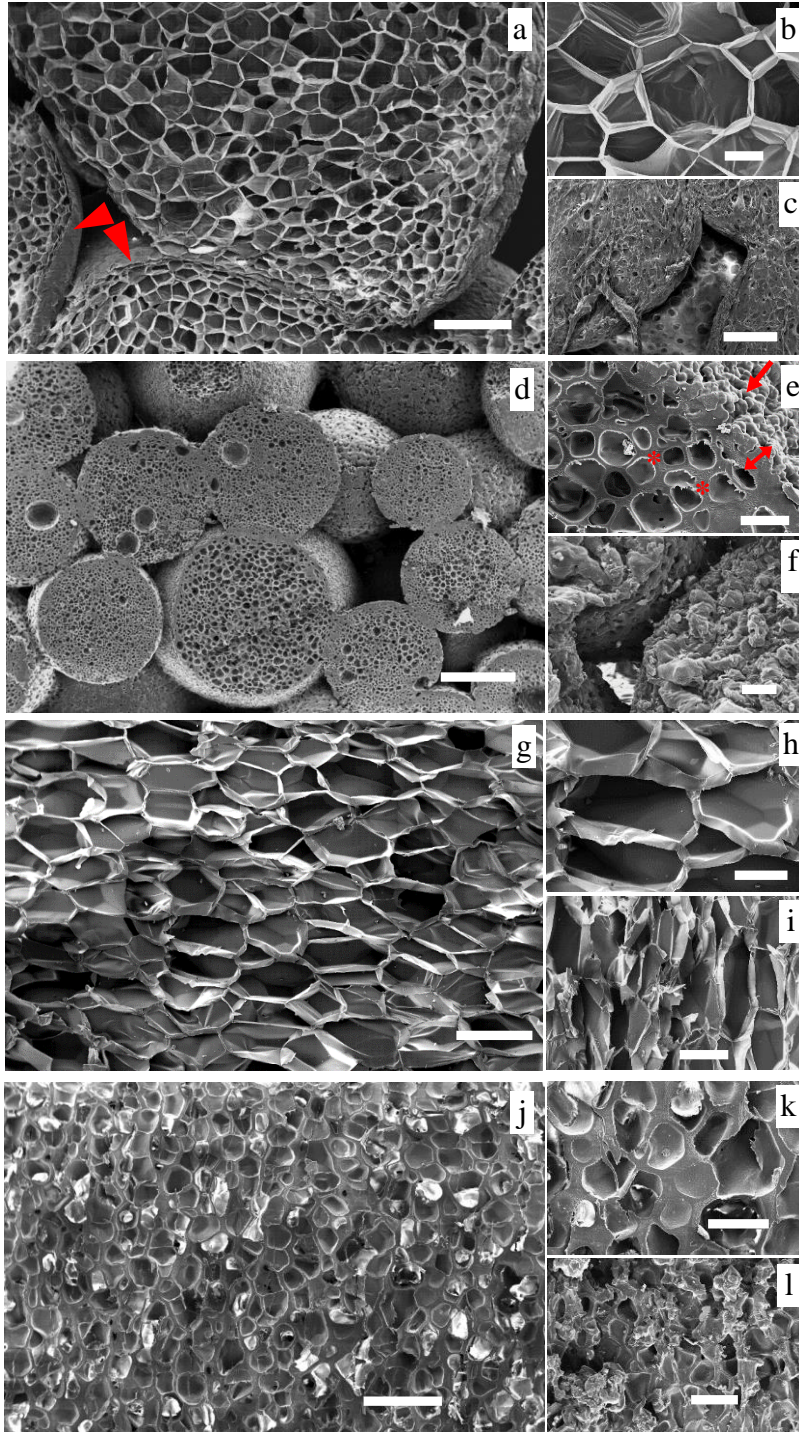


Fig. 3a-l. Scanning electron micrographs of EPS type E (a-f; a-c from untreated and d-f, treated) and XPS type B (g-l; g-i from untreated and j-l, treated) showing micro-morphological characteristics of PS before and after heat treatment at 120 °C for 2 h: (a-b) show inside of the PS beads illustrating honeycomb type cellular structure; (c) shows outer surface structure of the beads; (d-e) show the overall cellular network inside the beads that remains unchanged after the treatment; (f) shows considerable modification of the outer surface structure of the beads after the treatment; (g-h) show inside of the XPS beads; (i) shows the outer surface of XPS beads; (j-k) show inside of the XPS beads and unchanged cellular network; (l) shows the outer surface structure of the treated XPS beads. Scale bars: a,d,g,j, 500 μm ; b,c,f, 100 μm ; e, 50 μm ; h,k, 200 μm ; i,l, 300 μm

Differential Scanning Calorimetry

The glass transition temperature of the five untreated PS materials and after their treatment at 120 °C for 2h are presented in Table 2. For illustration purposes, DSC heating scans of sample A, before and after thermal treatment, are shown in Fig. 4. Before thermal treatment, all PS samples presented close values of glass transition between 100 to 105 °C. Those values were expected for polystyrene materials (Claudy *et al.* 1983) and are coherent with the effect of heat treatment on PS density as observed previously. Almost no density variation was observed for the treatment performed under the glass transition temperature in the glassy state at 80 and 100 °C, but significant changes were observed when thermal treatment was performed above the glass transition in the rubber state, at 120 and 140 °C. All the PS samples after thermal treatment presented a comparable glass transition value between 102 and 105 °C, which implied a non-significant variation to the untreated samples.

In addition, most untreated PS samples showed a small endothermic peak correlated with their glass transition (see Fig. 4). This peak is associated with the sample's structural recovery after physical ageing (Koh *et al.* 2016). This physical ageing is coherent with the origin of the materials as waste from building demolition. These peaks were absent for the PS materials after thermal treatment indicating that the structural recovery already happened during the treatment above the material glass transition temperature.

Table 2. Glass Transition Temperature of PS Before and After 2 h Heating at 120 °C

PS Material	Untreated	Treated
A	105	105
B	101	102
C	100	102
D	101	102
E	104	105

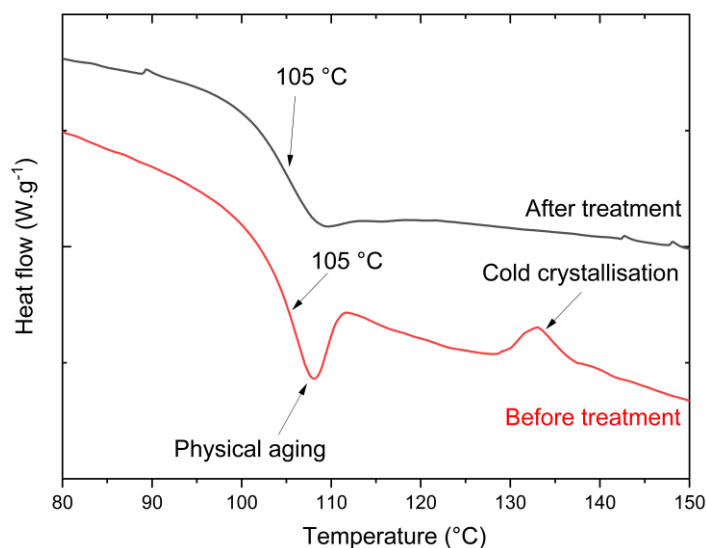


Fig 4. DSC thermographs of the XPS (white) demolition (A) samples before (red) and after (black) thermal treatment at 120 °C for 2 h

Finally, some untreated PS samples presented a small exothermic peak center at 130 °C. This small peak, which was not present in the samples after thermal treatment, is associated with a cold crystallization of the PS sample (Xu *et al.* 2003; Chen and Torkelson 2021). The absence of the peak for the heat-treated samples indicates that crystallization also occurred during the thermal treatment.

Mechanical Properties

Table 3 presents results of the mechanical properties of the PS samples after 2 h heat treatment at 120 °C. All treated samples showed higher values for the measured properties. This can be explained by the changes in morphology due to the treatments, which caused an increase in the density of the samples. For the XPS treated samples, the surface hardness increased by more than 12% compared to that of the EPS-treated samples (5%). The increase in surface hardness could be attributed to changes in the structure of the PS materials as it goes through the glass transition phase. The voids in the PS cell structure could be filled with melted material, which hardens upon cooling. In addition, small particulates may find their way to the surface through evaporation, which also increases the surface roughness (Kan and Demirboğa 2009). However, increased hardness often leads to brittleness in the structure of the PS.

Table 3. Certain Properties of EPS and XPS Types of PS Compared with those of Balsa Wood Before and After Thermal Treatment at 120 °C for 2 h

Properties	PS Material ¹		Wood Material
	XPS (Type B)	EPS (Type E)	Balsa wood
Density (g/cm ³)			
Untreated	0.015	0.028	0.16*
Treated	0.285	0.299	
Hardness (kp/cm ²)			
Untreated	7.0	13.8	45 / 100 **
Treated	89.0	70.8	
Compression Strength (MPa)			
Untreated	0.260	0.186	10 / 94 ***
Treated	3.682	3.550	
Thermal Conductivity (W/mK)			
Untreated	0.029	0.031	
Treated	0.047	0.059	

* Density at 15% moisture content;

** Hardness perpendicular to fibres = 45 kp/cm². Axial hardness = 100 kp/cm²;

*** Compression strength perpendicular to fibres = 10 kp/cm². Axial compression = 94 kp/cm²

The compressive strength increased after heat treatment compared to the untreated PS. Treatment at 120 °C increased the density, which had a positive correlation with the strength property. However, treatment beyond 130 °C had a negative correlation with compressive strength of EPS, probably because of the brittleness in the structure of EPS at elevated temperatures (Kan and Demirboğa 2009). Ellouze *et al.* (2020) showed that heat treatment of EPS caused an increase in compressive strength but with a decrease in compression plateau. The reduction was due to narrowing of cell size after thermal modification, resulting in faster densification of cells in the treated EPS. Compressive strength for the treated samples were 14 and 19 times greater than untreated samples for XPS and EPS, respectively (Table 3). This was similar to the results obtained for EPS in a related study. At a total densification of its cellular structure, the compressive strength of

treated EPS increased by 14 times compared to the untreated samples (Ellouze *et al.* 2020). The authors concluded that the created crust after treatment takes up most of the compressive stress and protects the cell structure from damage.

Thermal Conductivity

The PS foams are good thermal insulators and are thus used as building insulation materials. The thermal conductivity of the untreated PS samples ranged from 0.029 to 0.031 W/mK, while that for the treated PS samples ranged from 0.047 to 0.059 W/mK (Table 3). The thermal conductivity of the samples increased with increased density, thereby making heat treatment a less desirable option for recycling PS into insulation panels. Khoukhi *et al.* (2019) also found that the conductivity of PS insulation is affected by change in operating temperature. However, the conductivity value of EPS, which is a function of moisture content increases with reduction in material density (Khoukhi *et al.* 2019). Although XPS (type B) had a higher change in density after heat treatment, EPS (type E) showed a greater value in conductivity. Similar results were obtained by Kan and Demirboğa (2009), where conductivity increased 23% when EPS was treated at 120 °C for 15 min. A positive correlation was also obtained with apparent density for thermal conductivity in recycled EPS (Bumanis *et al.* 2023).

Sound Absorption

The average octave band sound absorption coefficients for XPS (type B) and EPS (type E) are presented in Fig. 5.

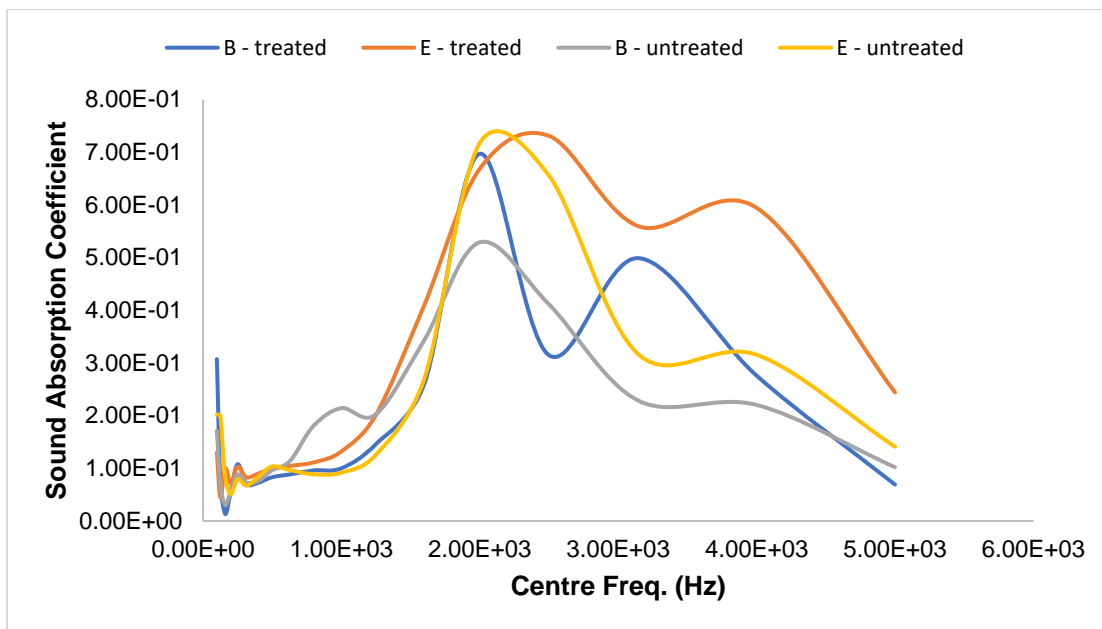


Fig. 5. Average octave band sound absorption coefficient for the treated and untreated polystyrene samples (B = XPS; E = EPS)

The change in the absorption band reflects the effect of thermal treatment on the PS samples. Compared to the untreated samples, the treated PS samples showed increased absorption band between 3000 and 4000 Hz center frequency. The treatment causes transformation of the alveolar structure in the PS into denser structure with fewer vacuums and more material (Ellouze *et al.* 2020). This is attributable to the development of a

compacted open structure in the PS samples. This irreversible change resulted in increased absorption and a corresponding decrease in the transmission of sound waves through the material. However, in the low frequency range (300 to 600 Hz), all samples showed similar band in sound absorption coefficient.

CONCLUSIONS

1. This study supports other works to demonstrate the feasibility of recycling low density polystyrene (PS) by optimized thermal treatment. This can potentially reduce the difficulty in recycling waste PS and promote a circular economy.
2. Heat treatment of recycled PS caused a noticeable change in the density of the PS when performed above their glass transition, with a corresponding increase in mechanical properties and thermal conductivity. Heat treatment also positively influenced the sound absorption property of the PS within the measured frequency range. When used as recycled PS aggregates in polymer matrix, there is the potential to enhance the properties of the basic material.
3. The modified properties following heat treatment were attributed to changes in the structural and morphological characteristics of the recycled PS. Based on the results, it is believed that this can be beneficial in structural applications with a high impact resistance. However, the increase in the thermal conductivity observed in this study presents a limiting condition when recycled PS is reused in thermal insulation applications.

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