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# EVALUATION OF SOME PLASTIC WASTES AS ADDITIVES TO REINFORCE HIGH-DENSITY FIBERBOARD (HDF)

*The use of plastic wastes in the forest product industry as an additive material is an alternative solution for reducing environmental pollution. In this study, different types of plastic wastes, polyethylene terephthalate (PET), polypropylene (PP), and polystyrene (PS), which have various characteristics and considerable potential as reinforcing materials for wood fibers, were added to high-density fiberboard (HDF) in different mixture ratios (25/75, 50/50, 75/25) with commercial fibers. Changes in some properties of the boards, including density, water absorption, thickness swelling, modulus of elasticity (MOE), bending strength (MOR), and internal bond strength, were determined. It was found that water absorption and thickness swelling ratios were lower in the boards with plastic waste additive than in the control samples. Moreover, the mechanical properties of the samples using plastic waste (except PET) were nearly as good as those of the control samples. The results indicate that PP and PS wastes can be considered for use in the reinforced HDF production process, with different mixture ratios for different usage areas.*

**Keywords:** plastic waste, reinforced HDF, dimensional stability, mechanical properties

## Introduction

The plastics industry is one that is developing rapidly, as the range of use of its products expands over time [Kaymakci et al. 2012]. Some projects have been developed over the past decade to counteract the negative effects of increasing plastic usage in the last century and to minimize the use of untreated plastic [Zarrabi-Ahrabi 2009]. The European Union, which has developed many of

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these projects, aims to increase the plastic recycling rate to 55% by 2030, and today recycled plastics are used in the manufacture of construction products, such as lumber, roof or floor tiles [Turku et al. 2018]. On the other hand, owing to technological developments, increasing consumption of limited forest products causes a decrease in forest resources [Inzani et al. 2013; Oncel et al. 2019]. The use of plastics in the forest industry, as part of a search for alternative raw materials that has been ongoing for the last 20 years, is an alternative solution both for the forest products sector and for dealing with environmental problems [Kaymakci et al. 2012; Sommerhuber et al. 2016]. Wood-plastic composite (WPC) is a new type of composite material created by combining a polymer with lignocellulosic material, which has been developed for the solution of problems relating to plastic use [Ashori 2008; Chaharmahali et al. 2008; Ayrilmis and Kaymakci 2013; Keskisaari and Karki 2018; Gulitah and Liew 2019].

The main result of research on virgin polymer WPCs is that the composite materials' water resistance and mechanical strength increase with increasing polymer content [Huuhilo et al. 2010; Ayrilmis and Jarusombuti 2011; de Cademartori et al. 2015; Rao et al. 2018]. However, plastic wastes can lead to heterogeneous density and morphology. These are major disadvantages of the use of such wastes in the production of recycled composite material, because certain production parameters, especially glass transition degree (Tg) and melting temperature (Tm), vary with density. In particular, Tg is an important parameter determining the maximum service temperature for largely amorphous plastics like polystyrene. It is also an important factor for the processing of plastics with higher crystallinity (polyethylene terephthalate (PET), polypropylene (PP), etc.). Tm represents the value of the maximum service temperature of this type of plastics [Chanda 2017].

Besides, structural damage to lignocellulosic materials occurs above 200°C. Hence, the use of polymers that can melt but not burn at temperatures between 150 and 220°C, from the group of thermoplastics – e.g. polyethylene (PE), polypropylene (PP), and polystyrene (PS) – can prevent deterioration of the wood material while producing a composite product together with the wood material [Chaharmahali et al. 2008; Najafi 2013]. Some of the most commonly used plastics and their Tg and Tm values are given in Table 1.

**Table 1. Tg and Tm values of obtained plastics [Chanda 2017]**

| Polymer type | Glass transition degree (Tg) | Melting temperature (Tm) |
|---|---|---|
| Polyethylene terephthalate (PET) | 69°C | **265°C** |
| Polypropylene (PP) | -20°C | **176°C** |
| Polystyrene (PS) | **100°C** | 240°C |

Generally, lignocellulosic-based materials, such as nut shells, sawdust, MDF waste, etc., have been used with plastics as a filler by many researchers [Jayaraman and Bhattacharyya 2004; Faruk and Matuana 2008; Karakus 2008; Ayrilmis and Buyuksarı 2010; Najafi and Khademi-Eslam 2011; Akbas et al. 2013; Ayrilmis et al. 2013; Chavooshi and Madhoushi 2013; Ozmen et al. 2014; Arnanda et al. 2017; Narlıoglu et al. 2018]. However, the wood-based composite sectors (fiberboard, particleboard, plywood, etc.) are larger than the plastic material sectors. Wood-based composites are more widely used than wood--plastic composites at home [Ritter de Souza Barnasky et al. 2020], including in flooring material, which is one of the main areas of application of WPCs [Gao et al. 2016; Machado et al. 2016]. Laminate flooring is a type of composite material which consists of several layers [Kara et al. 2016]. High-density fiberboards (HDFs), with densities above 800 kg/m$^3$ [ISO 818:1975], are usually used as the carrier panel layer for laminate flooring. Thus, the dimensional stability and mechanical strength values of HDF panels are important for laminate flooring production, and are generally subject to some additional requirements [Kara et al. 2016]. For this reason, the effects of the materials used in HDF production on these values are important for evaluating the usability of these materials. On the other hand, the wood-based composite sector directly depends on forest resources. It is therefore important to evaluate plastic wastes as a reinforcing material in HDF, as this may be a more effective solution for reducing environmental problems [Karaman et al. 2006; Tayyar and Ustun 2010; Ayrilmis and Jarusombuti 2011; Binhussain and El-Tonsy 2013; Najafi 2013; Nemati et al. 2013; Rahman et al. 2013; Lopez et al. 2018].

The main purpose of this study was to determine the usability of several plastic wastes in the production of HDF as a reinforcing material in laminate flooring. For this purpose, the most commonly used plastic materials, PET, PP, and PS, were added in different ratios to the HDF production system, and certain properties of the resulting HDF panels were observed.

## Material and methods

### HDF panel production

Commercially prepared wood fibers, manufactured by the thermomechanical pulping (TMP) method from a mixture of pine and beech wood (70:30 w/w) glued with 10% urea formaldehyde (Table 2), including 1% ammonium chloride (hardener), were supplied by the fiberboard production line of the Kastamonu Entegre A.Ş. Kastamonu O.S.B. plant (Kastamonu, Turkey). The final moisture content of the prepared fibers was approximately 10–12%, and these values were selected to correspond to the factory regimes. Plastic waste – polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS) – selected from waste materials from solid waste collection areas and recycle bins in Kastamonu province, and maleic anhydride (99.5%) were purchased from YPF Química

(Buenos Aires, Argentina). After grinding the plastic wastes with a hammer mill (Fig. 1) it was mixed with the fibers in different ratios (25:75, 50:50, 75:25 w/w), with maleic anhydride (3% w/w) as a coupling agent, in a laboratory drum mixer for a short time, and three panels were produced from each mixture. All of the panels were produced with a laboratory-scale single column press at Kastamonu University, and samples were pressed at an average pressure of 2.94 MPa for 10 min. The press factor is approximately 45 sn/mm, excluding the warm-up time of the extra press plates used to facilitate the loading and unloading of samples to and from the press. Convenient press temperatures (150°C for PS, 180°C for PP and PET mixtures) were determined according to the thermal properties of the plastic waste types (Table 1). The HDF panels were manufactured at the target density of 850 kg/m$^3$ with dimensions of 310 × 360 × 11 mm, and were conditioned at 20 ±2°C and 65 ±5% relative humidity to reach constant weight before the mechanical and physical properties were tested.

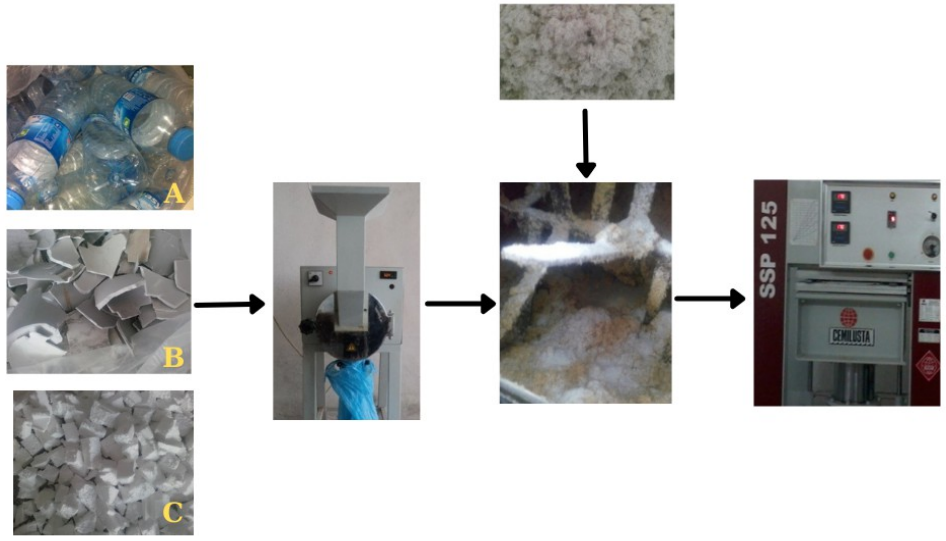


**Fig. 1. Addition of waste plastics to the produced boards (A: PET wastes, B: PP wastes, C: PS wastes)**

**Table 2. Properties of urea-formaldehyde adhesive**

| Properties | Urea-formaldehyde |
|---|---|
| Density (gr/cm$^3$) | 1.24 |
| Solid (%) | 55 |
| pH | 8.3 |
| Formaldehyde/urea molar ratio | 1.12 |
| Viscosity (20°C cp) | 63 |
| Gel time (100°C sn) | 50 |

### Characterization of HDF panels

Several mechanical features – modulus of elasticity (MOE), bending strength (MOR) and internal bond strength – and physical properties – density, water absorption, and thickness swelling – were determined according to the appropriate standards, which are EN 310 [1993], EN 319 [1993], EN 323[1993], and EN 317[1993] respectively. Water absorption and thickness swelling values were determined from weight and thickness measurements before and after 24 h water immersion of three replicate samples (50 × 50 mm) for each mixture. MOE and MOR values of samples (300 × 50 × 11 mm) were determined by a three-point loading test on a Shimadzu AG-IC machine. The applied support span-to-depth ratio (a/d) was 20, and the loading speed was 2 mm/min. Experiments were carried out with five sample replicates. Internal bonding strength tests were carried out on three 50 × 50 mm replicate samples for each mixture at a 2 mm/min loading speed on a Shimadzu AG-IC universal testing machine. Test results for all samples were analyzed by ANOVA and the Duncan test with the use of SPSS 22 (IBM Corp., Armonk, NY, USA).

Scanning electron microscopy (SEM) was applied to analyze the microstructures of the panels, using a Quanta FEG 250 (FEI, USA). To improve surface resolution HDF composites, the test pieces were coated with a thin layer of gold using a Cressington Sputter Coater 108 Auto Au-Pd Coating Machine, at Kastamonu University Central Research Laboratory, under 40 mA for 30 seconds.

## Results and discussion

Table 3 shows the results for the tested parameters of manufactured HDF panel samples. All board samples were found to have densities close to the target density depending on the polymer type and ratio. Small differences from the target density can be explained by various properties of the plastic waste used [Chanda 2017].

It was clear that, because of the structural properties, PS samples made from different mixtures with wood fiber had the lowest density, as shown in Table 3. Rahman et al. [2013] reported that board density decreased with increasing plastic contents. They also indicated that the main reason for this was the use of adaptive substances. Usually, adaptive substances, especially maleic anhydride, chemically bond between wood and plastic molecules [Faruk and Matuana 2008; Rao et al. 2018].

The ANOVA test was applied used to determine the effect of the polymer type and polymer mixture ratio on data of the HDF panels. ANOVA results are shown in Table 4. According to these results, the polymer type, the ratio of polymer to wood fiber, and the combination of these two factors were all statistically significant on MOR, while only polymer type had a significant effect on the MOE and internal bond strength. The ratio of polymer to wood

fiber was also statistically significant for the water absorption and thickness swelling values of the samples.

**Table 3. Physical and mechanical properties of boards, with standard deviations**

| | Density ($gr/cm^3$) | 24 h water absorption (%) | 24 h thickness swelling (%) | MOE ($N/mm^2$) | MOR ($N/mm^2$) | Internal bond strength ($N/mm^2$) |
|---|---|---|---|---|---|---|
| Control | 0.85 (0.03) | 67.09 (9.44) | 28.43 (3.42) | 1784.26 (624.37) | 17.76 (5.16) | 0.67 (0.11) |
| PET 25% | 0.90 (0.06) | 21.60 (3.02) | 8.90 (0.70) | 675.31 (55.53) | 12.09 (2.95) | 0.16 (0.01) |
| PET 50% | 0.88 (0.08) | 22.38 (11.88) | 7.41 (0.39) | 622.92 (394.72) | 10.80 (2.42) | 0.09 (0.02) |
| PET 75% | 0.89 (0.89) | 14.83 (2.18) | 4.18 (0.53) | 316.88 (291.71) | 2.62 (0.52) | 0.05 (0.01) |
| PP 25% | 0.83 (0.30) | 19.32 (4.33) | 8.03 (0.71) | 1652.21 (74.46) | 18.24 (3.52) | 0.60 (0.24) |
| PP 50% | 0.85 (0.08) | 18.76 (2.72) | 6.60 (0.98) | 1687.02 (300.51) | 19.71 (3.01) | 0.62 (0.17) |
| PP 75% | 0.84 (0.04) | 13.89 (2.14) | 2.10 (0.38) | 1789.06 (354.87) | 20.05 (2.62) | 0.75 (0.07) |
| PS 25% | 0.83 (0.01) | 35.33 (9.32) | 8.73 (0.97) | 1462.58 (154.52) | 17.85 (1.10) | 0.63 (0.03) |
| PS 50% | 0.84 (0.04) | 21.10 (4.65) | 5.44 (0.90) | 1775.04 (155.47) | 21.46 (1.07) | 0.74 (0.05) |
| PS 75% | 0.83 (0.03) | 16.24 (2.66) | 2.65 (0.12) | 1874.86 (557.15) | 25.64 (4.99) | 0.97 (0.19) |

**Table 4. Results of variance analysis**

| Source of variance | 24 h water absorption (%) | 24 h thickness swelling (%) | MOE ($N/mm^2$) | MOR ($N/mm^2$) | Internal bond strength ($N/mm^2$) |
|---|---|---|---|---|---|
| Polymer type (A) | ns | ns | * | * | * |
| Polymer mixture ratio (B) | * | * | ns | * | ns |
| AxB | ns | ns | ns | * | ns |

* significant, ns: not significant ($p \leq 0.05$).

The Duncan test was carried out on the tested parameters according to significant sources of variance. The results (Table 5) show the homogeneity groups of the dependent variables. The water absorption (WA) and thickness swelling (TS) values of the tested panels were statistically affected by variation in the polymer ratio. In the case of MOE and internal bond strength, the homogeneity groups were not clear.

**Table 5. Duncan test results of tested parameters according to significant source of variance**

| Mixture ratio | | | | Polymer type | | | |
|---|---|---|---|---|---|---|---|
| | WA | TS | MOR | | MOR | MOE | Internal bond |
| Control | A | A | A | Control | B | A | A |
| 25% polymer | B | B | A | PS | A | A | A |
| 50% polymer | BC | C | A | PP | AB | A | A |
| 75% polymer | C | D | B | PET | C | B | B |

Alpha = 0.05.

The 24-hour water absorption test results are shown in Figure 2. The results for WA (%) show a clear improvement with an increasing in the plastic waste content. The values for all groups with added plastic are lower than for the control groups, and meet the standard requirements. The thickness swelling results (Table 5 and Fig. 3) are similar to those for WA. Previous studies have also stated similar results for these parameters [Ayrilmis and Buyuksarı 2010; Ayrilmis and Jarusombuti 2011; Akbas et al. 2013; Ayrilmis and Kaymakci 2013].

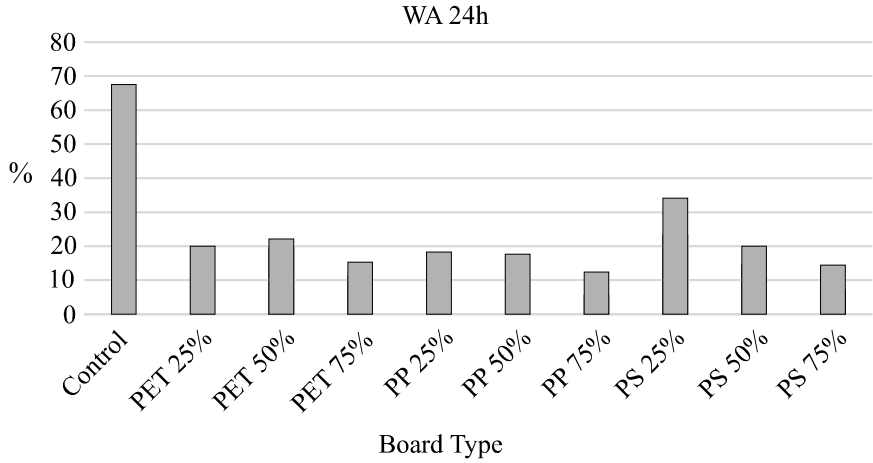


**Fig. 2. 24 h water absorption values of tested boards**

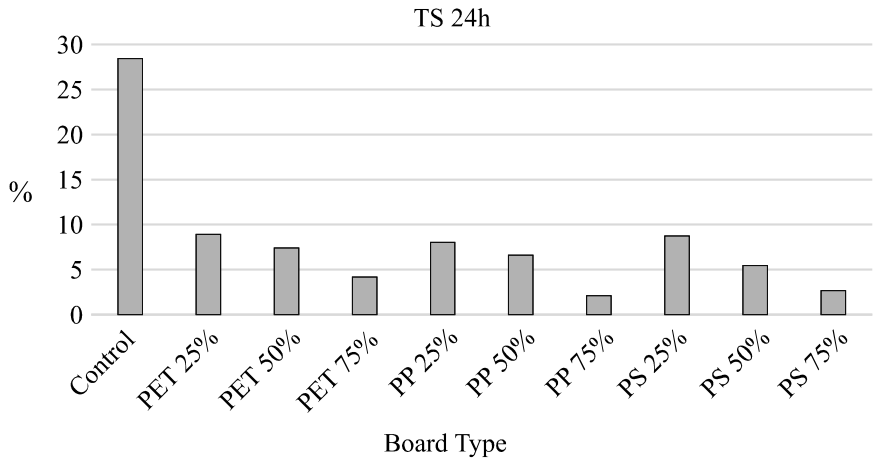


**Fig. 3. 24 h thickness swelling values of tested boards**

Although there is not too great difference between the absorption and thickness swelling values for the boards with added plastic after 24 hours, the difference is higher in the control samples. The results show that the addition of PET reduces the board's water absorption and improves its quality. Similarly to these results, Karaman et al. [2006] reported a preference for waste PET bottles in their studies, and stated that the water absorption and moisture problem in wood material can be solved by using plastic material. Ayrilmis and Kaymakci [2013] added 30%, 40%, and 50% polypropylene (PP) and 3% maleic anhydride harmonizer to wood fiber, and observed positive results for the dimensional stability values (water absorption and thickness swelling). Ayrilmis and Buyuksarı [2010] stated that water absorption values of maleic anhydride added composites were better than those of the control samples. Similarly, Binhussain and El-Tonsy [2013] tested boards made using 50% PS and 50% palm leaf composited panels. They determined the water absorption rate of the boards at 9.9%, lower than the values for hardwood samples (29.2%) and softwood samples (42.4%). The best results for WA after 24 h immersion in water were obtained using a 75% PS content. This could be due to the lower melting point of PS and a homogeneous distribution of PS particles between the wood fibers.

The bending strength (MOR) values for the test samples increased with increasing waste plastic content, except for the groups using PET (Fig. 4). The first reason for this may be the incompatibility of PET and wood fibers. Karakus [2008] reported that for bonding lignocellulosic materials with polymer materials, a coupling agent should be used. Secondly, a sufficient temperature to melt the PET wastes may not be reached, and the composition of the PET wastes used can be very heterogeneous. As shown in Table 1, the Tm temperature for PET is 265°C. This means that wooden fibers will burn before the PET Tm point is reached [Rahman et al. 2013]. Thus, PET is not a good filling material for reinforcing HDF production. For the groups containing PP and PS, the MOR

results were similar to those reported in previous similar studies [Jayaraman and Bhattacharyya 2004; Ayrilmis et al. 2013].

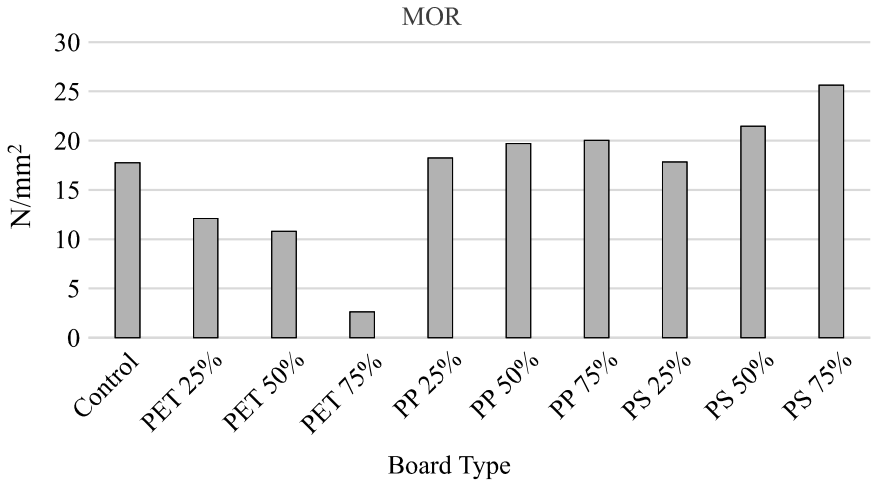


**Fig. 4. Bending strength values of tested boards**

In the case of modulus of elasticity (MOE) (Fig. 5), the values for groups using PP and PS waste were not significantly changed compared with the control group, but the MOE values for the groups with PET significantly decreased, similarly to the MOR values. Karaman et al. [2006] reported that mechanical properties are decreased for the polymer content varying from 30% to 100%. The main reason for this may be that in order to activate the functional groups of PET, the press temperature needs to reach the Tm value, but this is theoretically impossible for the HDF production line. In a study similar to the present one, Tayyar and Ustun [2010] reported that PET and HDPE were homogeneously distributed in the board, and when the PET content increased, the percentage value of strain under maximum load decreased. Muehl et al. [2004] also reported that the mechanical properties of panels made from PP were better than those of panels made with PET. Moreover, Nemati et al. [2013] concluded that the PS matrix led to positive changes in the mechanical properties of the wood-based composite product and provided significant improvements in its quality.

Results for internal bond strength perpendicular to the surface are similar to the other mechanical test results (Fig 6). It has been reported in previous studies that generally the mechanical properties of composite materials are improved by increasing the polymer content [Karaman et al. 2006; Ayrilmis and Kaymakci 2013; Ayrilmis et al. 2013]. According to the Duncan test results (Table 5), the PP and PS wastes data showed no statistically significant differences from the control samples. The groups using PET had significantly lower MOR and MOE values. This shows that the bonding between PET and the wood fibers was very

poor. Also, PP and PS wastes are more suitable for using for HDF production with different mixture ratios.

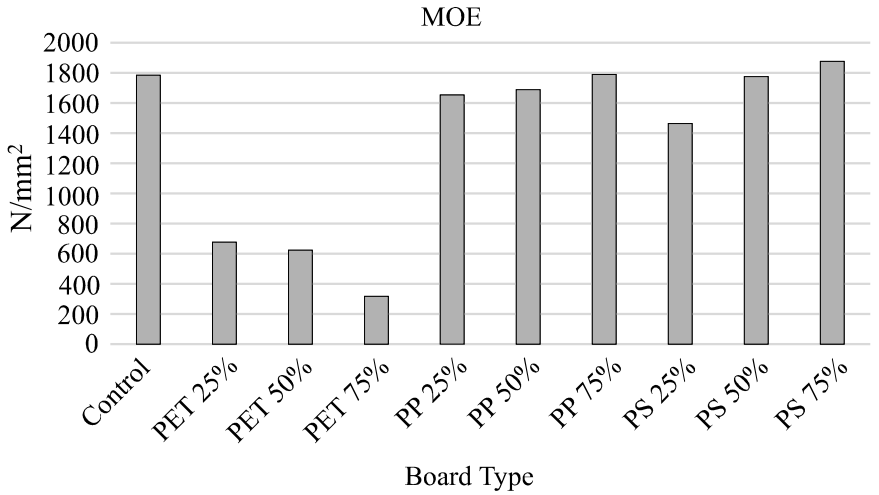


**Fig. 5. Modulus of elasticity values of tested boards**

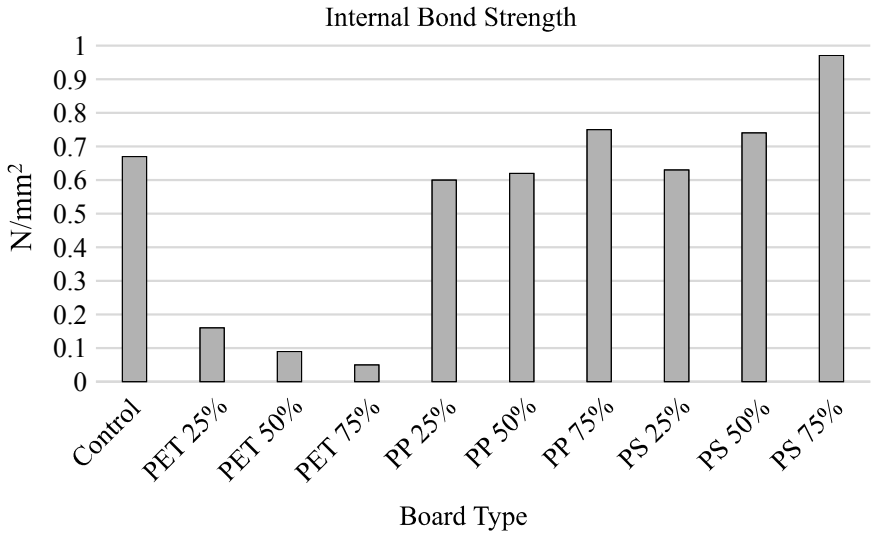


**Fig. 6. Internal bond strength values of tested boards**

SEM photographs of the produced boards are shown in Figure 7. When the microstructures are examined, it is clear that the fibers in the control group sample (Figure 7 A) are regularly distributed and their morphology is well preserved. As regards the groups containing waste plastic, it is observed that those made using polystyrene (Fig. 7 H, I, J) are quite well distributed as

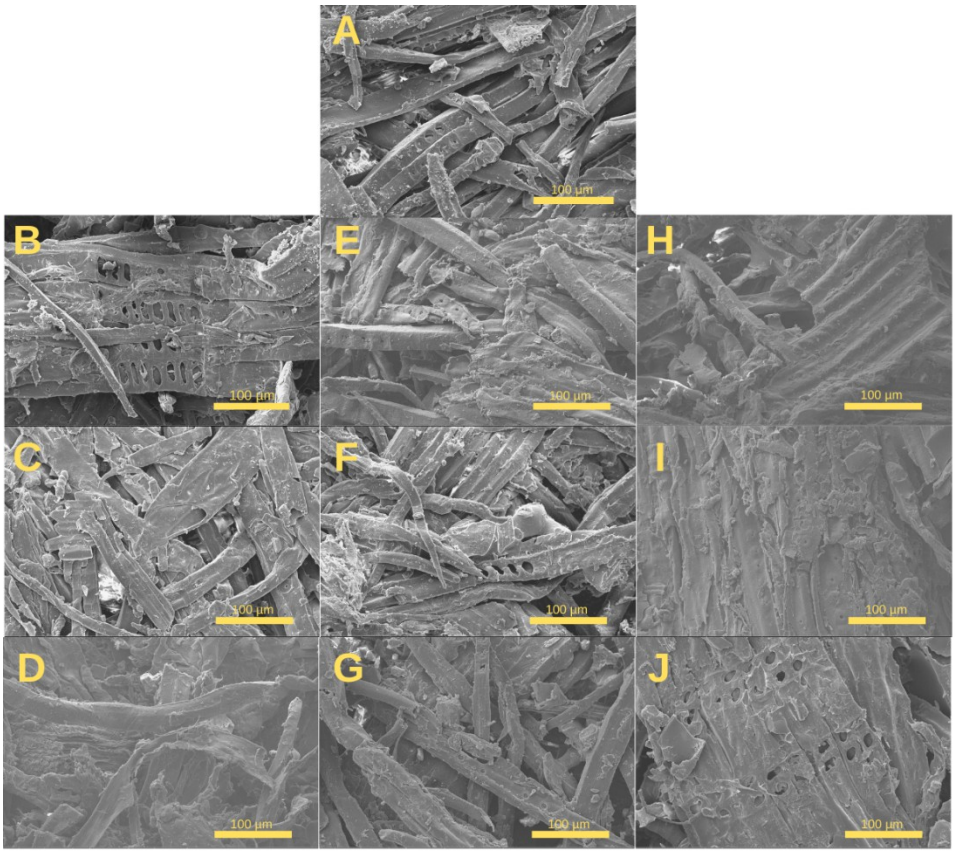


**Fig. 7. SEM micrographs of boards (A: control, B: 25% PET, C: 50% PET, D: 75% PET, E: 25% PP, F: 50% PP, G: 75% PP, H: 25% PS, I: 50% PS, J: 75% PS)**

a matrix on the boards. However, in the waste polypropylene and polyethylene terephthalate groups, whole polymer particles with different shapes are found between the fiber clusters; they act as a filling material that fills the gaps between the fibers rather than as a matrix. The microstructures of the boards support the notion that PET and PP wastes are heterogeneous in boards and PET wastes must reach their Tm point in order to form a bonding matrix between the fibres. It is also thought to be possible to improve the matrix distribution of PP wastes in the plate by changing the press temperature. In conclusion, according to the microstructure photographs, PS waste has better conformity with the fibers than PET and PP.

## Conclusions

The properties of HDF used in the production of laminate parquets can be reinforced with the use of plastic wastes, especially PS and PP. Thus, it is

possible to reduce pressure on forest resources by reducing both environmental pollution and wood requirements.

Because of the heterogeneous characteristics of plastic wastes, it will be possible to evaluate their potential for using as filling materials in HDF production, apart from some plastics such as PET, which has high crystallinity and higher Tm points.

According to the MOE and MOR tests, the addition of PS in particular will be advantageous in reinforced HDF production. In addition, since the Tg point of PS is low, it is possible to reduce the energy costs of the hot press by lowering the press temperatures used in HDF production.

Proceeding from this study, it is concluded that more studies on different plastic types and press temperatures, including those not covered by this study, and on behavior in a factory production environment are required to enable the incorporation of these materials into the industrial process.

In the case of PET wastes, a major cause of environmental problems, there should be more studies on wood composite production with using of different coupling agents.

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## List of standards

**EN 310:1993** Wood-based panels – Determination of modulus of elasticity in bending and of bending strength

**EN 317:1993** Particleboards and fibreboards – Determination of swelling in thickness after immersion in water

**EN 319:1993** Particleboards and fibreboards – Determination of tensile strength perpendicular to the plane of the board

**EN 323:1993** Wood-based panels – Determination of density

**ISO 818:1975** Fibre building boards – Definition – Classification

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