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Comparative Analysis of Recycled PVC composites reinforced with nonmetals of printed Circuit Boards

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Abstract: Resource recovery is an important aspect for environmental sustainability in a developed and developing economy. Poly vinyl chloride (PVC) is one of the most common and widely used construction material and its production process results in generating a great amount of waste. Luckily this waste can be recycled by combining it with other wastes such as Non-Metallic Fractions (NMF) of printed circuit boards (PCBs) which are usually land filled after the recovery of precious metals. This work has looked into the pulverization of NMF of PCBs from local scrap recyclers which were then utilized as reinforcing filler in waste and recycled PVC to make PCB-PVC composites. After the production of these composites the thermal stability, mechanical strength and fracture surface morphology of fabricated composites were investigated and compared among themselves to look for better composite composition. The obtained results revealed that using the NMF of paper fabric PCB slightly increased the thermal stability of PCB-PVC composites over its pure PVC composite counterpart. The use of pure PVC resulted in total weight loss of 87.8% however the addition of 25 wt% NMF decreased the total weight loss of 83.5% at 600 ^{0}C. There was significant improvement in the mechanical properties (such as tensile strength and modulus, bending strength and modulus) by increasing the NMF %age to a certain limit and/or threshold value, which was recorded as 20 wt% NMF. The use of this threshold amount of NMF increased the tensile and bending strength by 10MPa and 7MPa respectively in comparison with the use of pure recycled PVC. Comparative increase in tensile and bending strength modulus was recorded as 0.7 GPa and 0.9 GPa at the identified threshold value. Further increase in NMF percentage resulted in a decrease in the mechanical strength properties.

This study suggested that adding the threshold amount of NMF of PCBs as reinforcing filler in the recycled PVC is a potentially effective way to improve the mechanical and thermal properties, simultaneously resulted in effective waste resource reutilization. This study form the basis for its extension to the utilization of other polymeric material which will lead to a promising way of recycling the NMF of PCBs and trashed polymeric material resulting a sustainable waste reduction and reuse.

Keywords: Waste PVC, Waste PCB, NMF, sustainable recycling, Polymer Composites, Thermal stability, Mechanical properties.

1. INTRODUCTION

Polymeric materials such as plastic is a low cost widely used commodity in our daily lives having different applications such as coating, packaging, covers, bags, films and containers. Its large and increasing application has resulted in its increased production tonnage, resulting in generating huge amount of municipal plastic waste (Al-Salem, *et al.,* 2009). Poly vinyl chloride (PVC) is one of the widely used plastic which is mostly used in applications that have much longer application periods in comparison to other plastics. Its applications include but are not limited to its use in building infrastructures, actual buildings and housing decorations (Seki, *et al.,* 2014). (Dioxins 2012). (Wielgosinski, 2010). The worldwide production capacity of PVC was recorded as 54 million tons in 2012, which is increasing thereafter. In 2012 in terms of global plastic consumption PVC use stood at third position Poly vinyl chloride. Kunststoffe international, Munich (2013).According to the 2013 vinyl plus report, 37 million tons of PVC was produced worldwide. In Europe alone 5.5 million tons of PVC is made making it the third most used plastic VinylPlus,: VinylPlus Progress Report 2013..The suitability of PVC products for mechanical recycling has made it more advantageous in comparison to other polymers (PWMI Newsletter (2013) however PVC has poor thermal stability and low strength (Wang, *et al.*, 2010). and thus requires special care and processes for its recycling and reuse. In this work Non-Metallic Fractions (NMF) of Printed Circuit Boards (PCBs) were added to the reclaimed and recycled PVC in order to investigate its effects on thermal stability and mechanical strength properties of the resulting composite.

Production of Electrical and Electronic Equipment (EEE) is reported to be a fastest growing sector especially in the last two decades (Hadi, *et al.*, 2015). (Cui, and Zhang, 2008). The increased production of EEE has also resulted in its increased disposal

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frequency (Hadi, *et al.,* 2015). (Cui, and Zhang, 2008). (Niu, *et al.,* 2007). in both developed and developing economies due to increased product competition and advances. The latest near past reported estimates shows that 45 million tons of E waste was produced globally (Ghosh, *et al.*, 2015) (Ogunseitan, (2013).. in 2010 and this tonnage is ever increasing since then. PCB is an integral component of all EEE providing a base to connect all other elements (Ghosh, *et al.,* 2015) (Chi, *et al.,* 2011)for its proper functioning. PCB are comprised of different materials such as Cu, S, Al, Fe, Sb, Pb and other non-metals such as fiber glass, paper fibers, and thermo-set resins (Zheng, *et al.*, 2009) (Sum, 1991). Recycling of these used PCB is important due to its complex nature and imposed potential hazards to humans by land filling. Thus the recycling of these materials is important for sustainable waste management by recovering the valuable materials for reuse (Zheng, *et al.,* 2009) (Sum, 1991). (Goosey, Kellner, 2003). (Veit, *et al.*, 2006). (Cui, *et al.*, 2003). Barba- *et al.*, 2008). The current past disposal of NMF was by combustion and land-filling. However, the combustion of NMF is reported to be an environmental hazard due to the generation of highly toxic polybrominated dibenzodioxins and dibenzofurans (Guo, *et al.*, 2009). (He, *et al.*, 2006). (Lee, *et al.*, 2004) Land filling leads to a secondary pollution source due to the leaching of heavy metals and BFRs to contaminate the ground water (Guo, *et al.*, 2009). (He, *et al.*, 2006). (Lee, *et al.*, 2004). The NMFs could be safely recycled by two methods, physical and chemical. In physical method the various PCB components are separated out principally by the physical property differences utilized in a mechanical separator (Guo, *et al.*, 2009). Since the use of physical separation method does not require any pre- and post-treatment thus this results in decreased environmental loads on the process and it's mainly due to this consideration a physical separation method was used to separate the various PCB components. The separated Non-Metallic Fractions (NMF) from Metallic Fraction (MF) of PCBs has various applications such as its use as filler in the polymer composites, strength enhancer in concrete structures and to improve the temperature stability of various visco-elastic materials (Guo, *et al.*, 2009). (He, *et al.*, 2006). (Lee, *et al.*, 2004) (Guo, *et al.*, 2009) NMF can also be used as a reinforcing filler additive in the thermo-plastics and thermo-sets according to their main organic matrix **(**Liu, *et al.*, 2010). (Hadi, *et al.*, 2015).

In this work the Non-Metallic PCB fractions were recycled and used as reinforcing filler in the thermo-plastic polypropylene (PP) to improve its mechanical properties. NMF treated with silane coupling agent was mixed and extruded with has reported the use of NMF modified with pimelic acid in acetone solution. Their results showed an improvement in tensile, flexural and impact properties in comparision to the virgin PP (Hadi, *et al.*, 2015). Xu, *et al.*, 2014). (Xu, *et al.*, 2014). NMF utilized in High Density Poly-Ethylene (HDPE) by muniyandi et al., revealed that the addition of NMF up to a certain extent significantly enhances the mechanical strength properties (Hadi, *et al.*, 2015). (Muniyandi, *et al.*, 2013). (Guo, *et al.*, 2010). Suggested that the replacement of wood powder by NMF in wood polymer composite can improves the flexural and tensile strengths (Hadi, *et al.*, 2015). (Guo, *et al.*, 2010). The NMF reinforced composite plates were formed by NMF in PP by yin (Hadi, *et al.*, 2015). (Guo, *et al.*, 2010). has used the NMF of paper based PCBs as an additive in the waste PVC and their result showed that the certain extent NMF replacement can significantly improve the mechanical properties (Wang, *et al.*, 2010).Based on the literature cited in this section it is evident that PVC has numerous beneficial commercial uses and due to its large production tonnage the PVC waste is available worldwide making it a good source for sustainable recycling. Using NMF as a filler material in the recycled PVC is not only good for sustainable waste management but it will also save a significant amount of energy with markable improvement in the mechanical and thermal strength properties of recycled PVC.

2. MATERIAL AND METHODS

a. Composites processing

Recycled and size reduced PVC was obtained from recycling unit of a local PVC pipe industry in Hayatabad Industrial State of Peshawar, KPK, Pakistan. The material used in this work was processed once by this industry. PVC formulation used by the industry for pipe manufacturing before processing is given in **(Table 1).** Crushed PVC was screened in a vibratory sieve shaker [RETSCH AS 300] to obtain a uniform mesh size for making the composites.

Table 1: PVC formulation before processing

Constituents	Parts (By weight)
PVC	100
$Caco_3$	3
Thermal stabilizer	2
Titanium dioxide	0.4
Carbon black	0.15

PCBs collected from the local scrap store were washed for cleaning and then dried. Large metallic parts were manually separated and the remaining components were cut to a fine particle size range by using a sharp rotatary cutting mill. NMF from the mixture were separated out by using the dry based vertical vibration separation equipment (Habib, *et al.,* 2013). NMF were then screened in a vibratory sieve shaker (RETSCH AS 300) to get the required NMF particle size of 500, 250 and 150µm. No surface modification and/or chemical treatments were used at any stage of the process.

PCB and PVC powders were then mixed in a screw mixer in different weight ratios to make different composites. 500µm NMF particles were mixed with recycled PVC in the ratios of 5%, 10%, 15%, 20%, 25% and 30wt%. These compositions are tabulated in (**Table-2)**.

Table 2: PCB-PVC Composites formulation for processing

Sample	NMF %	NMF weight(gm)	PVC %	PVC weight(gm)	Total weight(gm)
S_0	0	0	100	140	140
S_5	5	7	95	133	140
S_{10}	10	14	90	126	140
S_{15}	15	21	85	119	140
S_{20}	20	28	80	112	140
S_{25}	25	35	75	105	140
S_{30}	30	42	70	98	140

A flat plat pressing mould with external heating system was designed for composites processing which had the internal mould dimensions of 170mm x 100mm x 35mm as shown in **(Fig. 1)**.

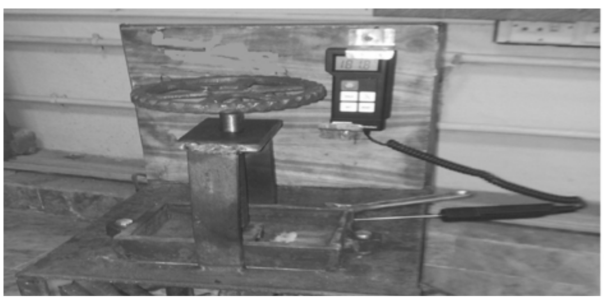


Fig. 1. Flat plat pressing mould for PCB-PVC composites processing

Upper and lower plates of the mould were lubricated with high viscosity index lubricant to prevent the material adhesion. Once the material was poured in the mould a pressure of 10 MPa was applied with heating temperatures range of 170-200^{o}C for 15 minutes. The resulting composite plates were then removed after cooling for 10 minutes at same pressure. Composite plates of dimension 170 mm x 100 mm x 4 mm were then obtained.

b. Material Testing

The TGA analysis was then carried out by using the Diamond TG/DTA Perkin Elmer, USA equipment. The tests were carried in an inert environment by using the nitrogen gas flow rate set at 20 ml/min. Initially the samples were held in the TGA equipment for 1 min at 30^0C, then heated from 30- 600 ^{0}C at the rate of 10^0C/min.

Mechanical tensile and bending strength tests were carried out by using the Universal Strength Testing Machine (UTM, Testometric model M500-100KN) with a test speed of 1mm/min. For the tensile tests a 4mm thick composite plate was cut into standard shape as shown in (**Fig.-2 and 3)**.

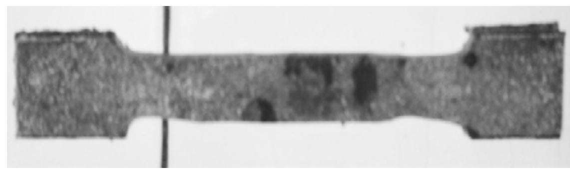


Fig. 2. Standard shape for tensile test

Fig. 3. Standard shape for three point bending test

In the standard shape for bending tests the minimum required length was 10 cm. A test speed of 0.5mm/min with a span of 80mm was used.

3. RESULT AND DISCUSSION

a. TGA Analysis

TGA analysis were carried out to investigate the thermal stability of recycled PVC and to see that how the thermal stability is affected by the addition of NMF. PCBs used in this study were paper fiber based that will usually degrade near 230^0C. PVC decomposition was expected to take place in 2 weight loss steps **(Table 3)** shows the TGA data for different PCB-PVC composites.

Table 3: **TGA Data for PCB-PVC composites**

Sample	Heating rate (^{0}c/min)	1st wt Loss stage		2nd wt Loss stage		Total wt loss at 600^{0}c (%)
		1st weight Fall Temp(^{0}c)	1st Wt Loss % (wt loss at start of stage - wt loss at end of stage)	2nd weight Fall Temp(^{0}c)	2nd Wt Loss % (wt loss at start of stage - wt loss at end of stage)	
S_0	10	278	52.75-4.4=48.35	429	85.8-55.5=30.3	87.84
S_{10}	10	270	52-5.5=46.5	430	85.3-54.8=30.5	85.3
S_{20}	10	261	51-6.4=44.6	429	84.4-55.2=29.2	84.4
S_{25}	10	258	49.8-5.5=44.3	415	83.5-54.1=29.4	83.5

In **(Tble-3)** it can be seen that the first weight fall temperature decreased as we increased the amount of NMF in PCB-PVC composites. This may be due to the presence of large amount of paper fibers which started to decompose near 230^{0}C. As for as the TGA results were concerned the addition of paper fiber based NMF had a slight effect on the thermal stability of PCB-PVC composites. There was a slight observed decrease in the weight loss percentages as we increased the amount of NMF in both stages. At low temperature the pure PVC composite were more thermally stable due to their lower water absorption capacity in comparison to the paper fiber based NMF. **(Fig. 4)** shows the combined TGA curve for composites which clearly shows the weight loss trend at various temperature stages.

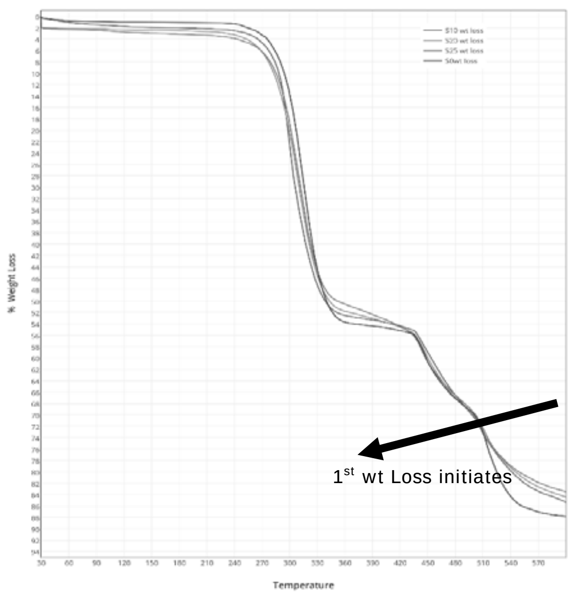


Fig. 4 Combined TGA cures of PCB-PVC composites

b. Strength Analysis

NMF-PVC composite specimens were tested for tensile, bending strength and modulus. Results revealed that both tensile and bending strengths increased with increasing the NMF ratio. However there was a certain limit and/or a threshold value at which the strength values reached their maximum and no further increase in strength was observed. Figure 5 shows a trend in tensile and bending strength variations with varying NMF weight ratios. Maximum values of strength were recorded at the threshold NMF amount of 20wt%. At this threshold NMF amount an increased Tensile and bending strength value of 10MPa and 7MPa were respectively recorded in comparison to the pure PVC sample. Further increase in NMF wt% weakened the adhesion capacity of PVC and PCB particles resulting in decreased strength measurement as projected in (**Fig-5).**

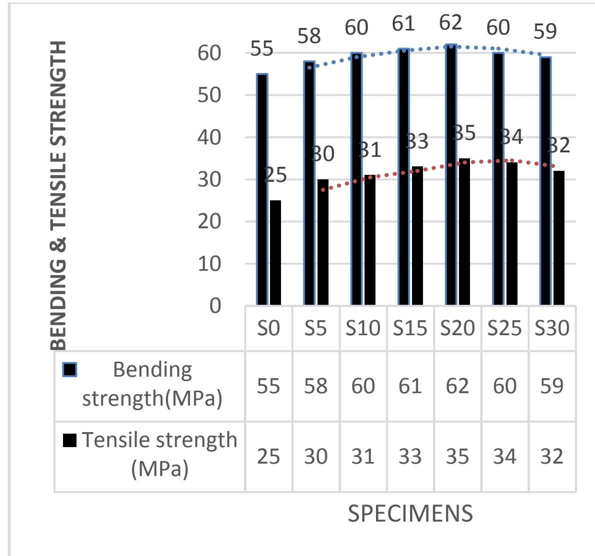


Fig 5 Variation of tensile and bending strength with change in NMF weight ratio

Another important property related to the strength is the tensile and bending modulus. Results showed that both tensile and bending modulus increased with increasing the NMF ratios. It was revealed from the obtained results that the addition of rigid particles like NMF improved the composite stiffness by improving the tensile and bending modulus. In line with the identification of threshold value in strength analysis an optimum modulus values were also recorded, beyond which the modulus values decreased. Addition of threshold amount of NMF increased the tensile and bending modulus by 0.7GPa and 0.9GPa respectively, over the use of pure PVC sample. Figure-6 shows these variations of tensile and bending modulus with changing NMF ratio in PCB-PVC composites.

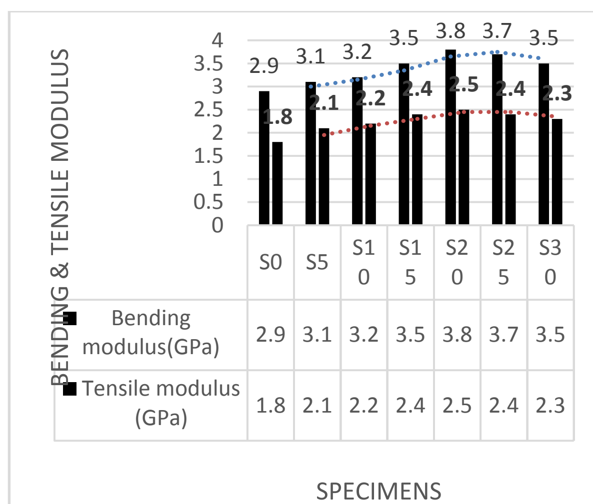


Fig 6 Variation of tensile and bending modulus with change in NMF weight ratio

c. NMF and Fracture surface SEM analysis

Fig.-7 shows the Scanning Electron Microscope (SEM) monograph of 500μm size NMF particles of PCBs after the separation of metallic fractions.

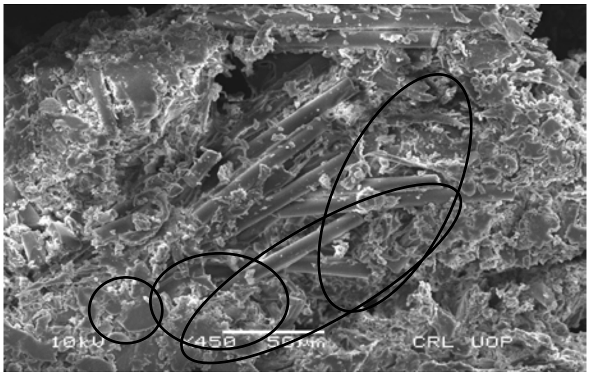
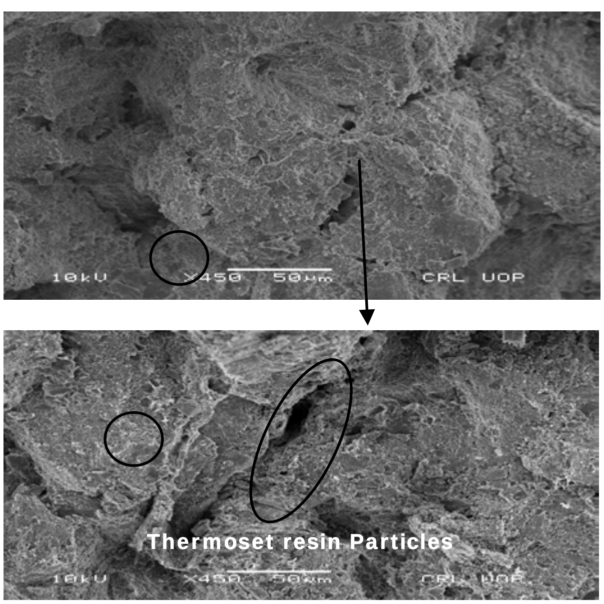


Fig. 7 SEM photograph of 500 micron NMF of PCBs

2nd wt Loss initiates

NMF of PCBs was mostly comprised of fibrous material like cellulosic paper fibers, woven glass fibers and thermo-set resins (epoxy resin or phenolic resin) SEM monograph analysis showed that the NMF powders mostly consisted of fibers and thermo-set resin particles. It can be seen in SEM monographs that the NMF particles were irregularly shaped due to their preparation in the cutting mill.

SEM monographs of fractured surfaces (after the tensile strength tests) of composites were obtained to investigate the NMF particles distribution and its effect on surface morphology after the fracture and surface roughness. **(Fig. 8)** shows the fractured surface SEM monographs of Pure PVC (8a), PCB-PVC having 15 % NMF (8b) and PCB-PVC having 20 % NMF (8c) composites. These all reported monographs were taken at X450 magnification.



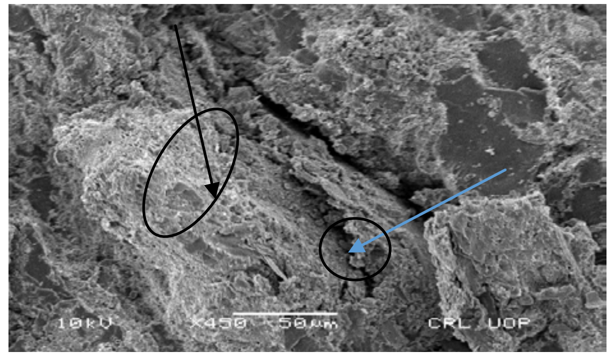


Fig. 8 SEM photographs of fracture surfaces, Pure PVC (8a), PCB-PVC having 15 % NMF (8b) and PCB-PVC having 20 % NMF (8c) composites

SEM monographs showed that the NMF particles were uniformly distributed in the composite structure. However the particle segregations zones were also present in some areas of the produced composite. NMF particles seemed to be strongly interlocked in the PVC particles showing a strong adhesion capacity in between them. From the reported monographs it is clear that by increasing the NMF in PVC increases the roughness of the fractured surface of PCB-PVC composites. Pure PVC composite fracture surface seemed to be very smooth as compared to composites having NMF. This increased roughness is potentially due to the greater extent of inter-locking among the NMF-PVC particles which caused the surface fracture to be more drastic and forceful. A large rupture holes can also be observed in PCB-PVC composites in Fig.-8 (a b c).

4. CONCLUSION

This study was aimed to analyze the effects of using the NMF as reinforcing filler in recycled PVC and to identify any improvements in the thermal stability and mechanical strength of the resulting composite. The TGA analysis revealed that there is a slight decrease in weight loss percentages as we increase the weight percentages of NMF in the composite. This study has shown that the NMF addition to the recycled PVC significantly improves the thermal stability and strength of the resulting composite. The obtained results have shown that the addition of identified threshold amount (20wt%) of NMF gives the maximum strength. This identified threshold amount also resulted in increased tensile, bending strength and modulus values. Any further addition over and above the identified threshold value resulted in decreased measure of all kind of strengths due to poor particle adhesion.

Recycling NMF of PCBs as filler in recycled PVC by a physical method is very cost effective, environmentally friendly and good for resource reutilization. This utilization method is therefore highly recommended in comparison to land filling and incineration which have proved hazardous for the environment.

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