



The reuse of nonmetals recycled from waste printed circuit boards as reinforcing fillers in the polypropylene composites

Yanhong Zheng, Zhigang Shen*, Chujiang Cai, Shulin Ma, Yushan Xing

Beijing Key Laboratory for Powder Technology Research and Development, Beijing University of Aeronautics and Astronautics, Beijing 100191, People's Republic of China

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ABSTRACT

The feasibility of reusing nonmetals recycled from waste printed circuit boards (PCBs) as reinforcing fillers in the polypropylene (PP) composites is studied by using both mechanical and vicat softening temperature (VST) tests. The concentration of Cu leached from the composites is also tested. The mechanical test shows that both tensile and flexural properties of the nonmetals/PP composites can be significantly improved by adding the nonmetals into PP. The maximum increment of tensile strength, tensile modulus, flexural strength and flexural modulus of the PP composites is 28.4%, 62.9%, 87.8% and 133.0%, respectively. As much as 30 wt% nonmetals recycled from waste PCBs can be added in the PP composites without violating the environmental regulation. The VST test shows that the presence of nonmetals can improve the heat resistance of the nonmetals/PP composites for their potential applications. The optimum particle is the fine or medium nonmetals recycled from waste PCBs, and the optimum content of the nonmetals is 30 wt% basing on the comprehensive consideration. All the above results indicate that the reuse of nonmetals as reinforcing fillers in the PP composites represents a promising way for recycling resources and resolving the environmental pollutions.

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1. Introduction

Printed circuit boards (PCBs) are the typical and fundamental component for almost all electronic products and they contain diversiform materials, including metals such as Cu, Al, Fe, Sn, Sb, Pb, etc. and nonmetals such as thermoset resins, glass fibers, etc. [1–2]. Recycling of PCBs is an important subject not only for the treatment of waste but also for the recovery of valuable materials as the amount of deserted PCBs is dramatically increasing [1–8]. Recycling may be inferred as separating the metals and plastics from the printed circuit boards and reusing them in their native applications. The lack of non-polluting separation methods to separate metals and nonmetals from these sources drives many researchers to go into mechanical separation methods [3,7–13]. Air classification is a cleaner separation method that does not use any polluting medium for separation [11].

After being mechanical separated, recycled metals such as Cu, Al, Fe, Sn, Sb, Pb, etc., are sent to recovery operations and the processes are already quite mature. However, significant quantities of nonmetals (up to 70%) especially present a huge challenge for reusing. The main components of the nonmetals are thermoset resins and

reinforcing materials. The main obstacle to recycle is coming from the crosslinked structure of the thermoset resins, which makes its melt impossible. So the conventional technologies used for thermoplastics resins are not applicable. Traditionally, these nonmetals are landfilled or incinerated without further disposing or reusing, which will cause resource waste and potential environment problems. Recently, many scientists and engineers are exploring to reuse these nonmetals in a more profitable and environmentally friendly way. The nonmetals could be used as fillers for epoxy resin products, such as paints, adhesives, decorating agents and building materials [14–17]. Hong and Su used nonmetals as reinforcing fillers in the polyester composite [18]. Mou et al. presented new methods that nonmetals were used to make formative models, compound boards or related products [19]. Xu and co-workers [20,21] reported that nonmetals were used for a kind of plate with a few additives and also used in production of phenolic moulding compound to replace wood flour. Franz [3] reported that the use of the nonmetals for thermoplastics would be the perfect recycling solution. The approach was reuse of nonmetals to make thermoplastic housing. Unfortunately, this approach is in complete opposition to ongoing trends in product miniaturization.

To our knowledge, there is little published information about using nonmetals recycled from waste PCBs as reinforcing fillers in the thermoplastic polypropylene (PP) resin and evaluating mechanical properties of the PP composites. PP as one of the most important commodity polymers is widely used in technical applications.

* Corresponding author. Tel.: +86 10 8231 7516; fax: +86 10 8231 7516.
E-mail address: shenzhg@buaa.edu.cn (Z. Shen).

Because of its good processibility, great recyclability and low cost, PP has been found a wide range of applications in the packaging, textile, automobile industries and furniture industries [22–24]. However, owing to its low strength, low modulus and high notch sensitivity, the usefulness of PP as an engineering thermoplastic is still limited. To expand the range of applications, the challenge of increasing the strength and modulus of PP has aroused considerable interest. Filling PP with rigid inorganic particles is an effective, economical, and convenient way to enhance the strength and rigidity [25–27].

In this article, the objective of the research is to reuse the nonmetals recycled from waste PCBs in the PP composites, with the aim to recycle the resources in a more profitable and environmentally friendly way. The feasibility of using nonmetals as reinforcing fillers in PP composites is studied by using mechanical and vicat softening temperature (VST) tests. The concentration of Cu and Br leached from the composites is also tested. All the results show that the reuse of nonmetals as reinforcing fillers in the PP composites represents a promising way for recycling resources and resolving the environmental pollutions.

2. Experimental

2.1. Preparation of nonmetals

In this study, the waste PCBs are from a personal computer PCBs factory (without electronic elements) and consist of a woven fiberglass mat impregnated with thermoset resins (epoxy resin or phenolic resin etc.). The process technology for mechanical recycling PCBs contains two-step crushing and air separation. The PCBs are firstly pulverized in a process consisting of a coarse-crushing step and a fine-pulverizing step. Then, air classifier is used to separate the metals from the nonmetals. After being separated, the nonmetals are screened by a series of sieves with holes of 25, 80 and 150 meshes. To ensure the surface smoothness, three fractions of the nonmetals with particle sizes of 25–80 (coarse), 80–150 (medium) and less than 150 meshes (fine) are selected and compounded in the PP composites.

2.2. Preparation of the nonmetals/PP composites

The nonmetals particles, which are 25–80, 80–150 and less than 150 meshes, are selected for making composites. To improve the dispersion of nonmetals particles in PP matrix and the compatibility between the nonmetals and matrix, all the nonmetals are modified with 1.0 wt% content of silane coupling agent KH-550 (γ -Aminopropyltriethoxysilane, Nanjing Shuguang Chemical Group Co. Ltd., China). PP powder S1003 (Beijing Yanshan Petrochemical Co. Ltd., China, melt flow rate 3.6 g/10 min) is used as the matrix

polymer. The PP powders and the modified nonmetals particles are dried at 80 °C for 2 h. Then, the dried nonmetals particles and PP powders are stirred and mixed by using high speed mixer (SHR-5A, Zhangjiagang Qiangda Plastics Machinery Co. Ltd., China). The nonmetals/PP blends are extruded into thread with a screw extruder (TE-35, Coperion Keya (Nanjing) Machinery Co. Ltd., China). The extrudate is pelletized, dried for 2 h at 90 °C, and injected into standard samples. Fig. 1 shows the whole fabrication procedure of the nonmetals/PP composites.

2.3. Measurements

The content of Cu in the nonmetals is determined by inductively coupled plasma-atom emission spectrometer (ICP-AES, Vista-MPX, Varian Co., American) after digesting by microwave digestion oven. The nonmetals are heated in a muffle furnace at 500 °C for 3 h to volatilize the resin and other organic materials. The weight contents of resin etc. and glass fiber are then obtained by weighing. The concentration of Cu and Br leached from the nonmetals/PP composites is tested by ICP-AES according to GB 5086.1-1997 (national standards of the People's Republic of China) for solid waste.

The tensile and flexural properties of the pure PP and nonmetals/PP composites are measured on an electronic universal testing machine (DXLL-10000, No.4 Chemical Machinery Plant of Shanghai Chemical Equipment Co Ltd., China) at room temperature (23 °C) with a cross head speed of 50 and 2 mm/min according to ISO 527-2:1993 and ISO 178:1993 standards, respectively. VST of the composites is determined by vicat heat resistance instrument (XRW-300M, Chengde Jinjian Testing Instrument Co., Ltd, China) at heating rate of 50 °C/h at load of 10 N according to ISO 306:1994 standard. The physical characterisation of the nonmetals and micrographs of the tensile fracture surfaces are observed by the scanning electron microscopy (SEM, LEO-1450, Germany). All of the specimens are carbon-sputtered before SEM test.

3. Results and discussion

3.1. Characterization of the nonmetals

3.1.1. Physical characterization

The main components of the nonmetals recycled from waste PCBs are thermoset resins and reinforcing materials. Physical characterisation of the nonmetals is complicated by the presence of both fibrous and particulate fractions. The shapes and compositions of the nonmetals vary with different particle sizes through careful observations on SEM as shown in Fig. 2. Microscopic observation reveals that the coarse nonmetals contain predominantly fiber-particulate bundles, with the majority of fibers being encapsulated in thermosetting resin as shown in Fig. 2(a). Single glass fibers and thermosetting resin powders are not seen. The ther-

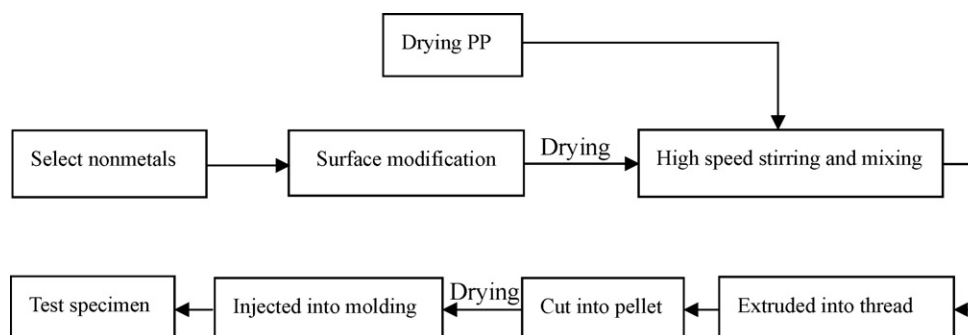


Fig. 1. The process of the preparation for the nonmetals/PP composites.

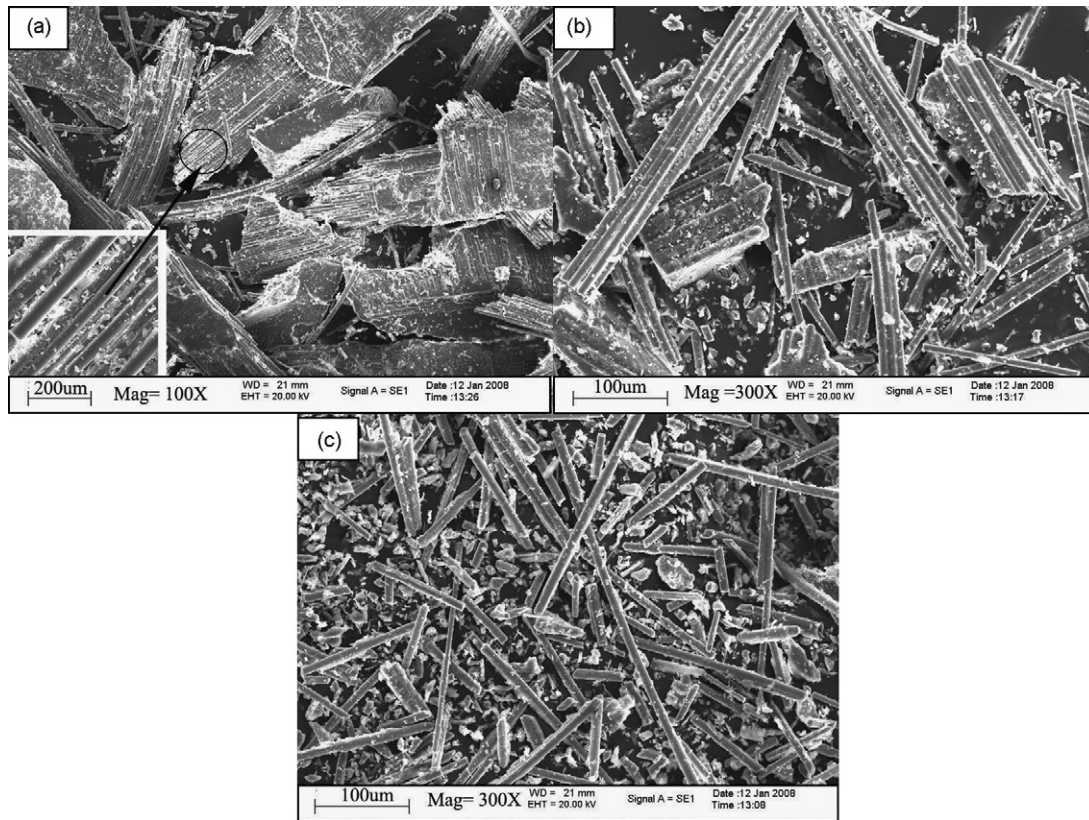


Fig. 2. SEM micrograph of the nonmetals with different particle size ((a) coarse, (b) medium and (c) fine).

mosetting resin gets stuck inside the glass fibers, forming a large fiber-particulate bundle of loosely entangled fibers. One part of the bundle marked by the circle is enlarged on the left-bottom of the Fig. 2(a). The medium nonmetals contain fiber-particulate bundles, single glass fibers and thermosetting resin powders as shown in Fig. 2(b). The fine nonmetals contain predominantly single glass fibers and resin powders as shown in Fig. 2(c). The differences of shapes and compositions among nonmetals with different particle sizes are determined by intrinsic structure of waste PCBs and the physical recycling processes.

3.1.2. Chemical characterization

The chemical composition and contents of three particle sizes of the nonmetals recycled from waste PCBs are listed in Table 1. It shows that the highest Cu content is in the fine nonmetals and reaches 1.64 wt%. So metals separating efficiency by air separation are very high. Meanwhile, the lowest resin etc. content is in the fine nonmetals and is 26.86 wt%. The highest glass fiber content is also in the fine nonmetals and reaches 71.5 wt%. The finer the nonmetals particles are, the higher the glass fiber content is. The microstructure of the glass fibers (volatilizing residues of the fine nonmetals by heating in a muffle furnace) is observed by using SEM as shown in Fig. 3. Compared with the microstructure of the fine nonmetals (Fig. 2(c)), most of them are single glass fibers. Glass fibers possess

many inherent characteristics such as high length diameter ratio (L/D ratio), high elastic modulus and low elongation. They are one of the most widely used reinforcements in thermoplastics [28–29].

3.2. Mechanical properties of the nonmetals/PP composites

Figs. 4 and 5 show the tensile and flexural properties of the nonmetals/PP composites with various additions of modified nonmetals (0–30 wt%), respectively. The content of the coupling agent was 1 wt%. The particle sizes of the fillers are 25–80 (coarse), 80–150 (medium) and less than 150 (fine) meshes.

Fig. 4 shows that the tensile properties of the nonmetals/PP composites are improved significantly by adding the fine, medium and coarse nonmetals particles (0–30 wt%), at room temperature. The

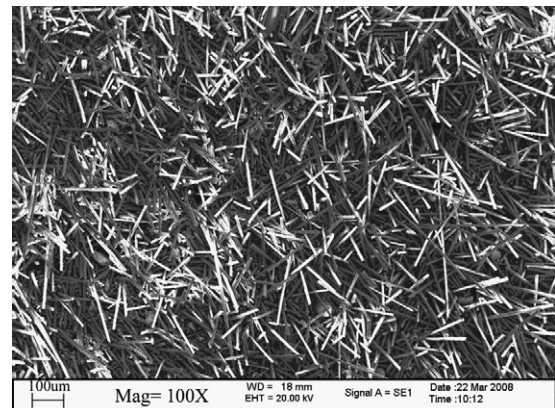


Fig. 3. SEM micrograph of the combustion residue of the fine nonmetals.

Table 1
The material content of the nonmetals recycled from waste PCBs

Specimen	Nonmetals	Cu content (wt%)	Glass fiber content (wt%)	Resin etc. content (wt%)
1	Fine	1.64	71.50	26.86
2	Medium	1.05	55.50	43.45
3	Coarse	0.48	46.00	53.52

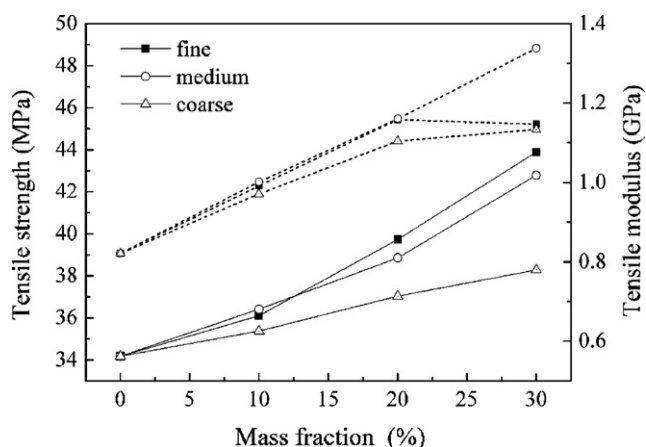


Fig. 4. (—) Tensile strength and (---) tensile modulus of the nonmetals/PP composites.

contents of the nonmetals added in the PP matrix can affect the tensile properties of the PP composites. It is evident that the tensile strengths and tensile moduli of the PP composites adding the fine, medium and coarse nonmetals almost increase with increasing filler contents. The maximum increment of tensile strength and modulus of the composites is 28.4% and 62.9%, respectively. However, as the content of fine nonmetals increase to 30 wt%, the tensile modulus of the PP composites suddenly decreases. That is mainly because the fine nonmetals have smallest particle size but a largest specific surface area that there is optimum interfacial contact area between the filler and PP matrix. The size of nonmetals also can affect the tensile properties of the composites. The tensile strengths of the PP composites adding the fine and medium nonmetals are both greater than those of the composites adding the coarse ones (0–30 wt%). It is evident that the size of the nonmetals is an important factor in affecting the tensile strengths of the nonmetals/PP composites. Decreasing particle size results in dramatic increases in the specific surface area of particles, which leads to an increase in interfacial contact area between the filler and PP matrix. The increase in interfacial contact area would be beneficial to transfer the stress from the matrix to particles, therefore resulting in higher tensile strength of the composite. But there is the optimum content of the nonmetals for the best tensile properties of the PP composites.

Similar to that of the tensile properties, it is shown in Fig. 5 that the flexural properties of the nonmetals/PP composites are

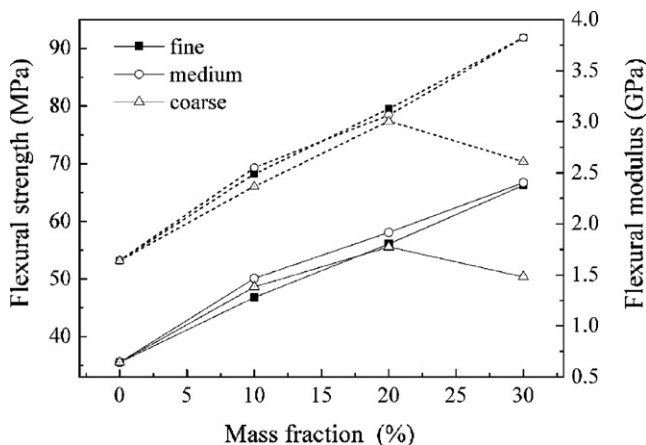


Fig. 5. (—) Flexural strength and (---) flexural modulus of the nonmetals/PP composites.

improved significantly by adding the fine, medium and coarse nonmetals particles (0–30 wt%), at room temperature. Moreover, the increases are greater than that from tensile properties. This is due to the fact that the crack initiating processes in the composites are different between tensile and flexural tests. The contents of the nonmetals added in the PP matrix can affect the flexural properties of the PP composites. When the filler is the fine or medium nonmetals, the flexural properties of the PP composites both increase with increasing filler contents. When the content of fillers increases to 30 wt%, the maximum increment of flexural strength and modulus of the composites is 87.8% and 133.0%, respectively. However, when the filler is the coarse nonmetals, there is an optimum content (20 wt%) for the best flexural properties of the PP composites. As the coarse nonmetals filler content increase to 20 wt%, the flexural strength and modulus further improve and reach a maximum value of 56.5% and 83.1% in the experiment, respectively. When the content of coarse nonmetals increases to 30 wt%, the flexural strength and modulus suddenly decrease. That is mainly because the coarse nonmetals contain predominantly fiber-particulate bundles that can easily induce the formation of higher stress concentration points in the nonmetals/PP composites with higher content filler during the bending and shear process. In addition, the interfacial adhesion between the particles and matrix is weak because of the irregular fiber-particulate bundle in the composites.

The changes in these properties mean that the tensile and flexural properties of the composites increase simultaneously by adding the nonmetals particles. In other words, the tensile and flexural properties of the nonmetals/PP composites are both greater than that of the pure PP. It is clearly shown that the nonmetals recycled from waste printed circuit boards can be used as reinforcing fillers in the polypropylene composites. In this paper, based on comprehensive consideration of the mechanical properties, economy, environment and technology, the optimum particle is the fine or medium nonmetals recycled from waste PCBs, and the optimum content of the nonmetals is 30 wt%.

3.3. Vicat softening temperature of the nonmetals/PP composites

VST tests are measured to determine the heat softening characteristics of the nonmetals/PP composites. This value is particularly important for the conversion of the material into the product for their potential practical application. Fig. 6 shows the characteristics of VST with nonmetals content and nonmetals size in the nonmetals/PP composites. No matter using the fine, medium or coarse nonmetals particles, the VST of the nonmetals/PP com-

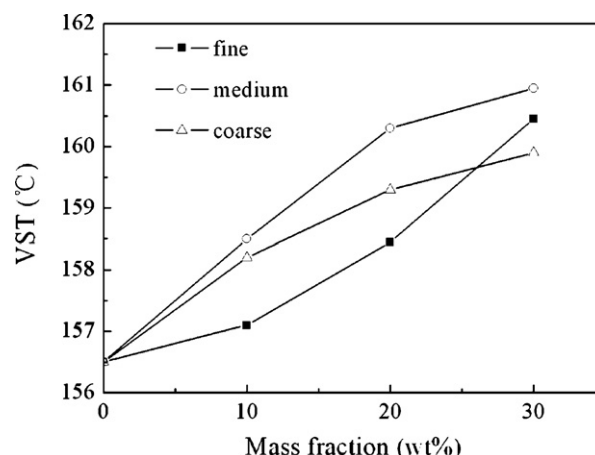


Fig. 6. Vicat softening temperature of the nonmetals/PP composites.

posites are improved significantly by adding nonmetals particles (within 0–30 wt%). The increasing effects from the nonmetals increase with increasing the filler content. The size of nonmetals fillers also can affect the VST of the composites. The VST of the PP composites with the medium nonmetals are better than those of the composites with the fine or coarse nonmetals. In a word, the VST of the nonmetals/PP composites increase 3–5 °C obviously compared with that of the pure PP.

The presence of nonmetals improves the heat resistance or heat softening characteristics of the nonmetals/PP composites for their potential practical application in an elevated temperature. The increase of the VST of the nonmetals/PP composites may be attributed to the nonmetals fillers possessing low thermal conductivity. In a word, the thermal properties of the PP material are improved by adding the nonmetals recycled from waste PCBs.

3.4. Tensile fracture surfaces observation and analysis

The mechanical properties results show that strength and rigidity of the composites are improved simultaneously when the nonmetals are filled into PP. That is mainly because the glass fibers in the nonmetals possess inherent characteristics such as high length diameter ratio, high elastic modulus and low elongation. Every dispersed particle triggers effective stress concentrations and leads to mass crazes so that the weak point cannot be formed in the composites. Thus, the polymer matrix properties are improved through the interaction of high strength particles and matrix. In this study, the effect of nonmetals is observed and analyzed under SEM on the fracture surfaces of the selected nonmetals/PP composites samples. All results are summarized as follows.

Fig. 7 shows the SEM micrographs of the tensile fracture surfaces of the nonmetals/PP composites with three particle sizes of the nonmetals recycled from waste PCBs (30 wt%). Dispersion of the nonmetals in the PP matrix seems to be good in Fig. 7. The tensile strength of the composite depends on the crack initiation and propagation characteristics through the matrix. While the crack initiation and propagation depends on the shape, orientation of the reinforcements in the matrix and interfacial adhesion between the particles and matrix. Fig. 7(a) shows the SEM micrograph of the tensile fracture surface of the nonmetals/PP composite with the fine nonmetals. On the tensile fracture surface of the composite, single glass fibers and resin powders are intimately mixed in the PP matrix. The glass fibers exhibit a large extent of pull-outs (see the pulled out fibers and the pull-out holes) and fibers breakage in the nonmetals/PP composites. Meanwhile, the fibers being pulled out have a rough surface, indicating the strong adhesion and good compatibility between the fibers and the matrix. Fig. 7(b) shows the SEM micrograph of the tensile fracture surface of the nonmetals/PP composite with the medium nonmetals. Single glass fibers and fiber-particulate bundles exhibit a large extent of pull-outs and breakage in the nonmetals/PP composites. Meanwhile, the presence of fiber-particulate bundles may be the result of stress concentration at the weakly bonded interface of the fiber-particulate bundle and PP matrix, which makes it easy for the crazes initiation and particles to detach themselves from the polymer matrix. Therefore, the composite sample with medium particles shows a little lower tensile strength compared with that of the composite with fine ones. Fig. 7(c) shows the SEM micrograph of the tensile fracture surface of the nonmetals/PP composite with the coarse nonmetals. The status of fillers mainly exhibit as larger fiber-particulate particles. There are some fiber-particulate bundles detached themselves from the PP matrix but almost no glass fiber pulled out from the matrix.

Results show that the fine nonmetals particles act as the concentration of stress which leads to the formation of cracks in the matrix

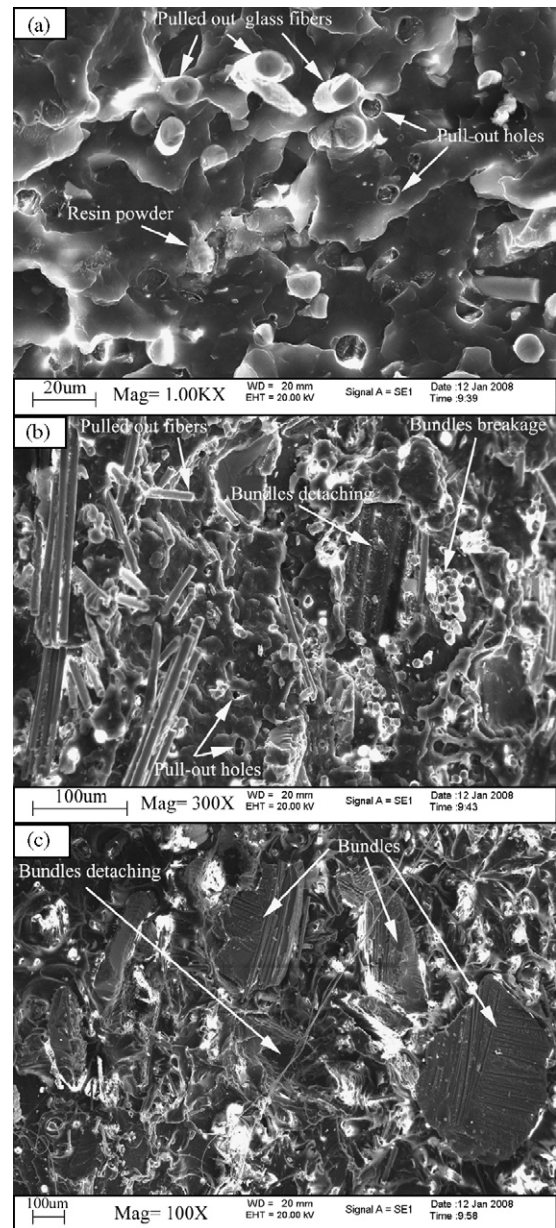


Fig. 7. SEM micrograph of tensile fracture surfaces of the nonmetals/PP composites (30 wt%), (a) fine, (b) medium and (c) coarse.

when they are dispersed in the PP matrix. The single glass fibers in the fine nonmetals possess high elastic modulus, low elongation and can first undertake the loading when the cracks propagate to the surface of particles. Meanwhile, the fine nonmetals have a larger specific surface area leading to more interfacial contact area between the filler and PP matrix. The more interfacial contact area would be beneficial to transfer the stress from the matrix to particles. The process of the crack initiation, propagation, fiber breakage and fibers pull-outs dissipate a great amount of energy. Therefore, the tensile strengths of the PP composite adding the fine nonmetals are improved. While the coarse nonmetals particles are filled into PP matrix, the interfacial adhesion between the particles and matrix is weak because of the irregular fiber-particulate bundle. The cracks can easily propagate along the weak interface, and the capability of particles to terminate the crack propagation and undertake the loading can be weakened. Thereby resulting in the interfacial debonding and then the fiber-particulate bundles

Table 2
The concentration of Cu and Br leached from nonmetals/PP composites

Specimen	Fillers	Concentration of leached Cu (ppm)	Concentration of leached Br (ppm)
1	Fine	0.030	6.32
2	Medium	0.018	4.88
3	Coarse	0.010	3.37

detached themselves from the PP matrix easily. Hence, the composite sample with coarse particles shows lower tensile strength compared with that of the composite with fine or medium ones. But the tensile strength is still greater than that of the pure PP. This is because the process of the crack initiation, propagation, interfacial debonding and bundles detaching dissipate a great amount of energy. Therefore, the tensile strengths of the PP composite adding the coarse nonmetals are improved.

3.5. The concentration of Cu and Br leached from the nonmetals/PP composites

The typical metal composition of PCB is copper ~16%, solder ~4%, iron, ferrite ~3%, nickel 1–2%, silver 0.05%, gold 0.03%, palladium 0.01%, others (bismuth, antimony, tantalum, etc.) <0.01% [2]. And the PCB is known to contain brominated flame retardants. So the nonmetals recycled from waste PCBs may contain brominated flame retardant and various metal elements, among which, Cu with the highest contents. Therefore, the concentration of Cu and Br leached from the nonmetals/PP composites is tested according to GB 5086.1-1997 for solid waste.

The PP composites with maximum content of the nonmetals in the experiment (30 wt%) are selected and tested the leach concentration of Cu and Br. Table 2 lists the concentration of Cu and Br leached from the nonmetals/PP composites. It shows that the composites with fine, medium, and coarse nonmetals leach Cu about 0.030, 0.018, and 0.010 ppm, respectively. As expected, the highest lead concentration of Cu is from the PP composites with fine nonmetals. That is mainly because the fine nonmetals have a highest Cu content and smallest particle size but a largest specific surface area of Cu, so that the capable of Cu being extracted from the PP composites is the highest.

Like the lead concentration of Cu, the highest lead concentration of Br is from the PP composites with fine nonmetals. Although the fine nonmetals have the lowest resin content in, the fine nonmetals have a smallest particle size with a largest surface area of Br that capable of being extracted from the PP composites is the highest. In addition, the coarse nonmetals contain predominantly fiber-particulate bundles with the most of resins encapsulating in the glass fiber bundle. As a result, the possible Br concentration, leached from the brominated compounds (such as brominated flame retardant) of the coarse nonmetals in the PP composites, is lowest. The concentration of Br leached from the nonmetals/PP composites is trace and below 6.32 ppm. However, there is no relative identification standard.

In a word, all the PP composites with 30 wt% nonmetals recycled from waste PCBs have leached Cu concentrations below 0.030 ppm. They are all complied with the identification standard for hazardous wastes (far lower than 50 ppm according to GB 5085.3-1996 of China). Therefore, the nonmetals recycled from waste PCBs can be successfully reuse in the PP composites.

3.6. Economic considerations

In China, the profit from recycling of waste PCBs is attained by selling recycled metals, and the profit can offset the cost of the recycling system, which includes management policies, a chain from production to consume, and behavior concepts.

Waste PCBs recycling enterprises have to pay 2000 Yuan/t when nonmetals are sent to the landfill [21], so it will be happy to convey nonmetals to PP producer if the price of transportation is much less than filling fee. Therefore, the price of nonmetals is zero when accounting the production costs. While the price of mineral fillers (such as CaCO₃, talc and mica) powder with different particle sizes is from 250 to 1000 Yuan/t according to the market price in China. In addition, the nonmetals recycled from waste PCBs can highly improve both mechanical properties and heat resistance of the PP plastic products. Increasing demand market for getting both higher performance and cheaper materials will cause a raise in nonmetals/PP consumption.

It is assumed that the adding content of nonmetals instead of mineral fillers is 30 wt% and the price of mineral fillers is 500 Yuan/t. It is also assumed that the adding contents of other materials such as PP resin and additives are same, so these costs are not taken into account when accounting the economic benefit. Taking a PP producer with annual output of 10,000 t PP plastic products, for example, economic benefit of 1,500,000 Yuan could be attained, which was attractive for producers of PP.

Although the appearance of the recycled PCB products is not attractive, this can be remedied by applying coating on the products, sandwiching with other materials, and using in little esthetic concern areas [18]. The nonmetals, a byproduct of PCBs recycling industry, could be used to replace traditional mineral fillers to manufacture reusable container, pallets, board and so on in the packaging and furniture industries. This would not only greatly decrease the manufacturing cost of PP but would solve an environmental problem caused by nonmetals of waste PCB and achieve complete recovery of reusable resources.

4. Conclusions

The nonmetals recycled from waste PCBs can be reused as reinforcing fillers in the PP composites. The tensile and flexural properties of the nonmetals/PP composites are improved significantly by adding nonmetals particles. The maximum increment of tensile strength, tensile modulus, flexural strength and flexural modulus of the PP composites is 28.4%, 62.9%, 87.8% and 133.0%, respectively. As much as 30 wt% nonmetals recycled from waste PCBs can be added in the PP composites without violating the Cu discharged standard. And the VST test shows the presence of nonmetals improves heat resistance and heat softening characteristics of the nonmetals/PP composites for their potential applications in elevated temperatures.

Based on comprehensive consideration of the mechanical properties, economy, environment and technology, the optimum particle is the fine or medium nonmetals recycled from waste PCBs, and the optimum content of the nonmetals is 30 wt%.

This study shows the beneficial reuse of nonmetals recycled from waste PCBs. Nonmetals, a new recycled material, may bring great social and economic benefits and, at the same time, prevent environmental pollution.

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