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## Recycling of ultrahigh molecular weight polyethylene waste used for preparing high performance synthetic paper

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**ABSTRACT**: Due to their widespread applications, the production and consumption of plastics have increased sharply and also brought about a lot of waste. However, only one-fourth of plastic solid waste is effectively recycled, and the rest is almost entirely disposed in landfills. In this article, high performance synthetic paper used for printing was successfully prepared from ultrahigh molecular weight polyethylene waste adopting thermally induced phase separation method. Silicon dioxide was added as filler and mineral oil was used as diluent. Whiteness, chemical resistance, tensile strength, thermal stability, surface morphology, and inkjet print of synthetic paper was investigated, respectively. It is indicated that the resulting synthetic paper possesses excellent printing effect. Its whiteness equally distributes around 80% and the highest tensile strength is up to 5.5 MPa. They also reveal good resistance to chemical corrosion and have good shape maintenance. As a result, the preparation of high performance synthetic paper for industrial applicability is expected to perform by recycling plastics waste. © 2016 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2016**, *133*, 44159.

KEYWORDS: application; characterization; membrane; preparation; recycling

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#### INTRODUCTION

The production and consumption of plastics have increased sharply. There was 280 million tons of plastic produced in 2012. According to current consumption rates, the planet will hold another 33 billion ton plastic by 2050.<sup>1</sup> Increasing cost and decreasing space of landfills are forcing people to find alternative options for plastic waste disposal.<sup>2</sup> Treatment, recycling and recovery methods for plastic waste are expected to be economically and environmentally viable, including landfill, incineration, chemical recycling, and mechanical recycling.<sup>3</sup> Usually, landfill takes large space, while incineration stimulates the growing emission of harmful, greenhouse gases, for example, NOx, SOx, COx, and so forth. Recycling is asked to decrease the waste volume and reduce environmental issues.

Al-Salem *et al.*<sup>2</sup> reviewed recycling and recovery routes of plastic solid waste, covering primary (re-extrusion), secondary (mechanical), tertiary (chemical), and quaternary (energy recovery). In recent years, there has been an increasing interest in seeking the potential applications of recycled mixed plastic wastes in building and construction sectors. Jafar *et al.*<sup>4</sup> selected recycled waste plastics as partial aggregate replacement in

bituminous mix product. Chemical recycling conforms to the principles of sustainable development and leads to the formation of the raw materials.<sup>5–7</sup> Wong *et al.*<sup>8</sup> discussed the current status and future prospects of plastic waste as source of fuel, with the main focus in pyrolysis, as well as co-pyrolysis of plastic waste with other materials. Any solution related to plastic waste problem should, of necessity, include the development of the recycling techniques. Plastic waste can be used as a raw material in the fabrication of new goods, thus reducing its environmental impact.

Consumption of traditional cellulose paper around the world currently leads to deforestation. To solve this problem, a special film that accepts writing or printing like cellulose paper, fabricated from thermoplastic polymers called synthetic paper is produced.<sup>9,10</sup> Synthetic paper can meet the requirements for light weight and reveal good resistance to weather, acids, and corrosion. Thus, it is increasingly replacing cellulose paper. At present, the preparation process of synthetic paper includes film method (YUPO, ALINDA, Q-per, etc) and fiber method (Tyvek) based on the polymer like PP, HDPE, PVC, and so forth.<sup>11</sup> Nowadays the film method is more commonly used. YUPO synthetic paper developed by Yupo Corporation is made from

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Figure 1. The processing route of internal paper-manufacturing method. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

PP by internal paper-manufacturing method, whose processing route is showed in Figure 1. However, the current methods just focus on the surface porosity, their indispensable postprocessing always brings about the complicated process. The development of synthetic paper still has many limitations. One reason is the complex process and another is the high cost.

Some researches on synthetic paper prepared of plastic waste have been conducted. Santana *et al.*<sup>10</sup> used postconsumer PP to make synthetic paper and examined the influence of surface treatment by CD on composite films. Correa *et al.*<sup>12</sup> prepared synthetic paper based on blends of post-consumer HIPS, as well as HDPE in various proportions for graphic printing. Santi *et al.*<sup>13</sup> used films of post-consumer polypropylene composites for the support layer in synthetic paper. These studies on transforming plastic waste into synthetic paper are beneficial to plastic waste disposal and reducing cost.

Ultrahigh molecular weight polyethylene (UHMWPE) is a high performance engineering thermoplastic with many repeat units of  $-C_2H_4$ —. It has incomparable properties among plastics caused by its physical chemistry stability. UHMWPE-based composite materials are widely used in medical, military, industrial, and consumer applications.<sup>14,15</sup> However, UHMWPE presents in a rubbery state in melting resulted from its ultrahigh molecular weight (>1.0 × 10<sup>6</sup>). Furthermore, UHMWPE has a small frictional coefficient and low critical shearing rate.<sup>16</sup> All of these properties make UHMWPE hard to process. Thereby, it is seldom made into membranes with routine methods such as melt stretching. Thermally induced phase separation (TIPS) is a suitable method for UHMWPE being processed into microporous membranes.<sup>15,17–22</sup> Even UHMWPE has more and more widespread application, its recycle had to be further developed.

In our research, UHMWPE waste was used as raw material of fabricating synthetic paper adopting TIPS method. The synthetic paper was characterized by thermogravimetric analysis (TGA), scanning electron microscopy (SEM), tensile testing machine, color measurement spectrophotometer, and inkjet printer. And the application performances were also compared with that of the virgin UHMWPE products.

#### EXPERIMENTAL

#### Materials

UHMWPE resin was collected from post-consumer film or leftover bits and pieces. SiO<sub>2</sub> (OK500: industrial grade; particle size, 6  $\mu$ m; density, 1.9 g/cm<sup>3</sup>) was kindly supplied by Shanghai Aile Imaging Materials (Shanghai, China) used as a filler. Mineral oil (alkane mixtures of C15 to C40, kinematic viscosity 65  $\times 10^{-6} - 71 \times 10^{-6}$ m<sup>2</sup>/s, industrial grade) was obtained from China Petro-chemicals Group, Hangzhou Refinery (Zhejiang,



**Figure 2.** Process flowchart of preparing synthetic paper from UHMWPE waste. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

China). Absolute ethyl alcohol (AR grade) obtained from Sinopharm Chemical Reagent (Shanghai, China) was used as an extraction agent.

Cellulose paper (A4 size, 80 g) was collected from common office papers. Virgin UHMWPE (number-average molecular mass  $2.0 \times 10^6$ ) was purchased from Shanghai Lianle Chemical Industry Science and Technology (Shanghai, China).

#### Preparation of Synthetic Paper from UHMWPE Waste

Figure 2 illustrates the process of manufacturing synthetic paper based on UHMWPE waste. In Figure 2, the UHMWPE waste was the resin material for preparing synthetic paper.  $SiO_2$  was used as filler to improve the opacity and ink receptivity of paper. Mineral oil played a role as diluent to dissolve the polymer at a high temperature.

Melt and Extrusion. The UHMWPE waste were precut into chips (Side length: about 0.5 cm) first. Then, washed in clean water at room temperature, and dried in a fanned oven. UHMWPE chips, mineral oil and SiO<sub>2</sub> were manually premixed. After premixing, the mixture was extruded from a conical SJSZ20/40 twin-screw extruder at the speed of 6.0 rpm (temperature program: 200/180/160/140 °C) with the UHMWPE dissolved in the mineral oil forming a homogeneous solution. The extruded blend was air cooled to induce phase separations and then granulated.

The weight ratio of UHMWPE chips, mineral oil,  $SiO_2$  particles was listed in Table I. The mass fraction of mineral oil in all mixtures was fixed at 75 wt % while the mass ratio of  $SiO_2$ /solids was variable containing 0 wt %, 6.7 wt %, 13.3 wt %, 20 wt %, 26.7 wt %, 33.3 wt %, respectively.

Table I. Composition of the UHMWPE/Mineral Oil/SiO2 Mixture

Sample	UHMWPE (g)	OK500SiO <sub>2</sub> (g)	Mineral oil (g)	SiO <sub>2</sub> / Solids (wt %)
U1	15	0	45	0.0
U2	14	1	45	6.7
U3	13	2	45	13.3
U4	12	3	45	20.0
U5	11	4	45	26.7
U6	10	5	45	33.3





**Figure 3.** CIE Whiteness of synthetic paper from UHMWPE waste (U1, U2, U3, U4, U5, U6), virgin UHMWPE (a) and cellulose paper (b). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

**Preparation of Membranes.** The blend was placed between a pair of stainless steel plates and pressed into membranes with a flat vulcanizing machine (XLB-400  $\times$  400  $\times$  2, Qicai, Shanghai), at 200 °C and 8 MPa for 180 s. Then, the stainless steel plate with the membrane together was taken out quickly and air cooled. Finally, take the regular membrane out of the mold.

**Extraction and Drying.** The diluent in membranes was extracted by being immersed in ethanol for 24 h. The resulting membranes were dried in vacuum drying chamber at 60  $^{\circ}$ C for 6 h, and then, the synthetic paper was obtained.

#### Characterization of Synthetic Paper

Whiteness. The CIE Whiteness of the synthetic papers was evaluated by color measurement spectrophotometer (Datacolor650, Datacolor, America) according to the ISO 11475: 2004 standard, which specifies the procedure used for determining the whiteness of papers or boards. When the samples are viewed under the CIE D65 daylight illuminant, the resulting values can correspond to the visual appearance of white papers or boards, which is with or without fluorescent whitening agents.

**Chemical Resistance.** Representative acid and alkali solutions, including hydrochloric acid (38 wt %), nitric acid (68 wt %), sulfuric acid (98 wt %), sodium hydroxide (40 wt %) were adopted to test the chemical resistance. The synthetic paper were soaked in above mentioned solutions for 48 h at room temperature, and then dried in vacuum oven at 40 °C for 24 h. Evaluate the chemical resistance by observing appearance and calculating the weight loss according to the eq. (1).

$$\gamma = (M - m)/M \times 100\% \tag{1}$$

where M represents the mass of synthetic paper before soaking, and m is the mass after soaking and drying.

**Tensile Test.** The tensile test was evaluated with a digital tensile testing machine (CZ-8010, Zhongzhi, China) at room

temperature with a humidity of about 50% at a tensile speed of 5 mm/min. The initial gauge length and width were 50 and 10 mm, respectively.

**Thermal Shrinkage Measurement.** The thermal shrinkage was evaluated by measuring one-dimensional length changes in the axial and transverse directions after being kept in vacuum drying chamber (DZG-6020 Shanghai Science, China) under different temperatures for 30 min. All the tested samples were squares with 3 cm  $\times$  3 cm dimensions. The thermal shrinkage was calculated according to the eq. (2).

Thermal shrinkage(%) = 
$$(L_1 - L_2)/L_1 \times 100\%$$
 (2)

where  $L_1$  and  $L_2$  represent the side length of the squares before and after heating.

**Thermogravimetric Analysis.** TGA was conducted on a thermal analyzer (TG209F1, NETZSCH, Germany) under nitrogen atmosphere at flow rate of 20 mL/min. Usually, about 5 mg samples were heated from room temperature to 900  $^{\circ}$ C at a heating rate of 20  $^{\circ}$ C/min.

**Morphology Observation.** The surface morphology of the synthetic paper was tested using SEM (TM-1000, Hitachi, Japan). The samples were coated with gold in a rarefied argon atmosphere (20 Pa) using an Emitech K550 Sputter Coater, with a current of 12 mA for 50 s.

**Inkjet Printing.** The printing of synthetic paper was evaluated by a regular laser jet (HP1020, Hewlett-Packard Development Company, America).

#### **RESULTS AND DISCUSSION**

#### Whiteness

Paper with higher whiteness causing excellent light reflection ability tends to exhibit more vibrant colors, thus presenting better visual effects. But a visual fatigue may occur after a long time staring. The resulting whiteness indices of the synthetic paper prepared from UHMWPE waste are displayed in Figure 3. It is found that all the synthetic paper based on UHMWPE



Figure 4. Mass loss of synthetic paper from UHMWPE waste and virgin UHMWPE after soaking S-1 (40% sodium hydroxide), S-2 (38% hydro-chloric acid), S-3 (68%nitric acid), and S-4 (98%sulfuric acid).





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**Figure 5.** Appearance of synthetic paper from UHMWPE waste before (O) and after soaking (T1: 40% sodium hydroxide; T2: 38% hydrochloric acid; T3: 68% nitric acid; T4: 98% sulfuric acid). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

waste have similar values at around 80%, except for the sample U2 prepared with  $SiO_2$  content at 6.7 wt %. The results confirm that inorganic particles in synthetic paper are independent of the variation in whiteness.

The variety of impurities and degradation byproducts in the waste would impart a variable degree of yellowing to the membrane and cause not only the differences in whiteness from paper to paper but also the relatively lower values of whiteness compared to those of synthetic paper made of virgin UHMWPE [Figure 3(a)] and cellulose paper [Figure 3(b)]. Even so, the whiteness (80%) of synthetic paper prepared from UHMWPE waste is still suitable for printing and visual sensation if regarding of the commercial synthetic paper Teslin with 76% whiteness.<sup>11</sup>

#### **Chemical Resistance Characterizations**

The mass loss of synthetic paper based on UHMWPE waste and virgin UHMWPE after soaking is shown in Figure 4, respectively. It can be seen that the mass loss of all the synthetic paper are below 5% in different solutions. It is inferred that these samples have good chemical resistance regarding of the common acid and alkali. The filled  $SiO_2$  also shows relatively good acid and alkali resistance. Compared with virgin UHMWPE, synthetic papers prepared from UHMWPE waste possess more mass loss. It is attributed to the physical chemistry stability of virgin UHMWPE. However, synthetic paper prepared from UHMWPE waste suffers about 5% mass loss in sulfuric acid, indicating that the impurities in the waste are prone to be



**Figure 7.** Thermal shrinkage of synthetic paper from UHMWPE waste with different  $SiO_2$  content (0 wt %, 6.7 wt %, 13.3 wt %, 20 wt %, 26.7 wt %, 33.3 wt %) treated with different temperature. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

corroded and form defects. Synthetic paper prepared from virgin UHMWPE reveals no mass loss in sulfuric acid. It may be because there appears a phenomenon-like passivation.

Figure 5 shows the appearance of synthetic paper based on UHMWPE waste before and after soaking in chemical solutions. It is found that synthetic paper takes on little change in appearance except in sulfuric acid.

#### The Effect of SiO<sub>2</sub> Content on Tensile Strength

The results for tensile test of synthetic paper samples are shown in Figure 6(a). The tensile strength increases with the increase of inorganic filler, which plays a role as filler toughening the composite system. It reaches the maximum of 5.5 MPa when the content of  $SiO_2$  is 20 wt %. But with the  $SiO_2$  continuing to increase, the tensile strength decreases. This may be on account of the agglomeration of  $SiO_2$  during preparation, which causes uneven crystallization and results in weak skeleton of pores with poor homogeneity and stress defects.



**Figure 6.** Tensile strength of synthetic paper from UHMWPE waste with different SiO<sub>2</sub> content (0 wt %, 6.7 wt %, 13.3 wt %, 20 wt %, 26.7 wt %, 33.3 wt %) (a) and stress–strain ( $\sigma$ – $\epsilon$ ) curves of different typical SiO<sub>2</sub> content (0 wt %, 20 wt %, 33.3 wt %) (b). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]



Figure 8. TGA curves (a) and residual weight (b) of synthetic paper from UHMWPE waste with different  $SiO_2$  content (U1, U4, U6). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Figure 6(b) shows the stress–strain curves of different typical SiO<sub>2</sub> content (0 wt %, 20 wt %, 33.3 wt %). The  $\sigma$  and  $\epsilon$  value of the sample with moderate SiO<sub>2</sub> (20 wt %) are the maximum, indicating better performance with higher strength and ductility. But with the further increasing of SiO<sub>2</sub> content, the  $\sigma$  and  $\epsilon$  both tend to decrease due to the particle agglomeration.

If comparing with the synthetic paper made from virgin UHMWPE, whose tensile strength is in the range from 4 to 8 MPa,<sup>11</sup> the strength of synthetic paper made from UHMWPE waste decreases due to the impurities and degradation byproducts. According to literature,<sup>11</sup> PP/ethylene propylene rubber synthetic paper owns a highest tensile strength of 3.0 MPa. It is indicated that the synthetic paper obtained from UHMWPE waste can meet the general application requirements.

#### **Thermal Shrinkage Studies**

Paper is expected to meet requirement of being applied at high temperature. The thermal shrinkage is a proper characterization of thermal stability.

Figure 7 presents the thermal shrinkage of synthetic paper prepared from UHMWPE waste treated under different temperatures from 80 to 120 °C for 0.5 h. It is shown that they easily lost dimensional stability after being exposed to over 100 °C. The synthetic paper almost has no shrinkage at 80 °C proving that they possess good stability under ambient temperature. When the temperature increases to 120 °C, all samples show a significant shrinkage percentage of 8%. It is indicated that synthetic paper from UHMWPE waste still keep good feasibility of the application at a relatively high temperature blow 80 °C.



Figure 9. SEM images of synthetic paper from UHMWPE waste (a) U1, (b) U2, (c) U3, (d) U4, (e) U5, (f) U6.



**Figure 10.** Ink-jet printing of synthetic paper from UHMWPE waste (a) U1, (b) U2, (c) U3, (d) U4, (e) U5, (f) U6, A4 cellulose paper (g), and synthetic paper made of virgin UHMWPE (h). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

It can be inferred from Figure 7 that the thermal shrinkage may be affected by  $SiO_2$  content. Usually,  $SiO_2$  plays a role as nucleating agent which promotes the crystallization process of UHMWPE known as semi-crystalline polymer in preparation. As a result, the skeleton of the microporous membrane gets enhanced. However, the agglomeration of too much  $SiO_2$  will lead to the worse stability of synthetic paper. Therefore, the thermal shrinkage cannot decrease with the increase of  $SiO_2$ content.

#### Thermogravimetric Analysis

Figure 8 illustrates the TGA curves and residual weight of synthetic paper from UHMWPE waste (U1, U4, U6) and the initial degradation temperature were 277.4 °C, 287.9 °C, and 281.3 °C, respectively. The TGA curves indicate that synthetic paper without filling (U1) starts to decompose first. It is found from Figure 8(a) that the thermal stability of the systems increases with the increase of SiO<sub>2</sub> content. When SiO<sub>2</sub> content is 33.3 wt % (U6), the initial degradation temperature decreases because of agglomeration. Serious agglomeration of SiO<sub>2</sub> brings about the unevenness of crystalline. Therefore, the strength of the resulting membrane is reduced.

From Figure 8(b), it can be seen that there is difference among the residual weights of the samples with different  $SiO_2$  contents. This is due to the impurities existed in the UHMWPE waste, which cannot decompose even under high temperature. Furthermore, part of  $SiO_2$  that affecting crystallization is extracted combined with the diluent during preparation.

#### **Morphologies Analysis**

SEM micrographs of synthetic paper from UHMWPE waste, with and without filling, are shown in Figure 9. By comparing, it is shown that all of the samples have almost similar surface morphologies. The microvoids are on the surface with different size and shape. But with the increasing of SiO<sub>2</sub> content, the agglomeration causes the unevenness of crystalline. As the pores' skeleton is weakened, part of them will collapse when being extracted. From the SEM image of U6 with the highest SiO<sub>2</sub> content, it is found that there are different between the pores size and distribution.

Usually, polymers are nonporous and have a chemically inert surface with low surface energy, which resulting in weak adhesion to other materials. But the synthetic paper based on microporous material prepared using TIPS method was expected to achieve the printing expectation.

#### **Inkjet Printing Characterizations**

Figure 10 illustrates the inkjet printing results, which also can be confirmed by Supporting Information. It is indicated that all the synthetic paper from UHMWPE waste have good adherence, including the one that not filled with SiO<sub>2</sub>. U6 is of best printing behavior when the SiO<sub>2</sub> content reaches a maximum value of 33.3 wt % [Figure 10(f)]. This may be owing to the filling of SiO<sub>2</sub>, which can improve the paper opacity.

It also can be seen that the synthetic paper made from virgin UHMWPE [Figure 10(h)] shows excellent printability. Similar excellent print quality and ink adhesion are observed when plain A4 cellulose paper [Figure 10(g)] is tested in the same way. It is inferred that synthetic paper from UHMWPE waste possesses good printing performance.

#### CONCLUSIONS

Synthetic paper based on UHMWPE waste was successfully prepared using TIPS method. Whiteness of the resulting synthetic paper was in the range from 70 to 90%. The tensile strength can reach up to 5.5 MPa when the SiO<sub>2</sub> content was 20 wt %. Moreover, the resulting synthetic papers show good thermal



stability. All samples have good adherence resulting in good printing effect. In addition, the synthetic papers have good resistance to chemical corrosion and reveal good shape maintenance. UHMWPE waste is expected to be recycled for preparing synthetic paper, which can meet some special requirements in different surroundings that cellulose paper is not applicable.

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