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Laboratory simulations of the mixed solvent extraction recovery of dominate polymers in electronic waste

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ABSTRACT

The recovery of four dominant plastics from electronic waste (e-waste) using mixed solvent extraction was studied. The target plastics included polycarbonate (PC), polystyrene (PS), acrylonitrile butadiene styrene (ABS), and styrene acrylonitrile (SAN). The extraction procedure for multi-polymers at room temperature yielded PC, PS, ABS, and SAN in acceptable recovery rates (64%, 86%, 127%, and 143%, respectively, where recovery rate is defined as the mass ratio of the recovered plastic to the added standard polymer). Fourier transform infrared spectroscopy (FTIR) was used to verify the recovered plastics' purity using a similarity analysis. The similarities ranged from 0.98 to 0.99. Another similar process, which was denoted as an alternative method for plastic recovery, was examined as well. Nonetheless, the FTIR results showed degradation may occur over time. Additionally, the recovery cost estimation model of plastics in e-waste, especially with a higher proportion of PC and PS, can achieve a lower cost than virgin polymer product. It also reduced 99.6%, 30.7% and 75.8% of energy consumptions and CO₂ emissions during the recovery of PC, PS and ABS, and reduced the amount of plastic waste disposal via landfill or incineration and associated environmental impacts.

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1. Introduction

Millions of tons of plastic are produced around the world, and less than half of it is recycled (Kaya, 2016; Rochman et al., 2013). For example, approximately 78 million tons of plastic waste were generated in the world's three largest economic powers in a one year period (the United States in 2012, the European Union in 2012, and China in 2011) (Ni et al., 2016). Most of the plastics were incinerated for energy recovery or sent to landfills; both disposal methods can release toxic materials into the environment (Chung et al., 2010; Lea, 1996). Plastics recycling is economical and ecofriendly (Wu et al., 2013), and it may have a positive effect on global warming (Astrup et al., 2009). Because of this, plastics recycling has become a growing concern (Nishida, 2011; Wager and Hischier, 2015; Wang and Xu, 2014). However, there has not been any substantial growth in recycling technology for plastics because of the high recovery requirement and cost of disposal (Perrin et al., 2016; Stein, 1992). In this context, developing economical recycling technology for plastics has become essential for environmental and economic reasons.

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Electrical and electronic wastes (e-waste) are swiftly growing in volume (Widmer et al., 2005; Zhou and Qiu, 2010). E-waste has a high plastic content (20-25%) (Alston et al., 2011), and PC and ABS account for the major portion of e-waste plastic (approximately 85%) (Stenvall et al., 2013; Weeden et al., 2015). Current options to manage plastic wastes are mainly recycling, incineration and landfill (Burange et al., 2015). Recycling e-waste plastics can be helpful in controlling the growing plastic problem (Bhaskar et al., 2002; Hooper et al., 2001) and achieving maximum recirculation (Clark et al., 2016). There are several methods for polymer recycling, such as direct recovery, which keeps the polymer structure intact, reuse of monomers and polymer degradation (Antonakou and Achilias, 2013; Gutierrez et al., 2013; Papaspyrides et al., 1994). Recently, repurposing polycarbonate (PC) via conversion into the more valuable poly(aryl ether sulfone)s was reported (Jones et al., 2016). In most cases, plastic waste forms a complex mix, and e-waste plastic contains numerous different resins (such as styrenics, polyolefins, engineering plastics, and thermosets) and various additives (including stabilizers, flame retardants, colorants, pigments, plasticizers, fillers, etc.) (Buekens and Yang, 2014). This makes the efficient separation of different types of plastics from e-waste very difficult (Bhaskar et al., 2008). Current work has been primarily limited to the recovery

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of one or two types of plastic at a time. In an effort to develop the resourceful and economically ways of recovery, methods of plastic recovery include: mechanical recycling, solvents recycling (secondary mechanical recycling), pyrolysis, and gasification (Wang and Xu, 2014). Recycling of plastics by segregating mixed wastes into a homogenous polymer (e.g., mechanical recycling) leads to broader application and higher value (Braun, 2002). In light of life cycle assessment, mechanical recycling is more attractive in environmental terms (Dodbiba et al., 2008). In particular, solvent recycling is proved as a promising and widely used application (Cervantes-Reyes et al., 2015). Comparing with other methods, major advantages of solvent recycling are the simplicity of equipment and operation for separation, and a high purity of recycled plastic (Deferm et al., 2016). For example, a selective dissolution technique was applied for polystyrene (PS) recycling (Gutierrez et al., 2013). Arostegui et al. developed a dissolution-based recycling technique for acrylonitrile butadiene styrene (ABS) (Arostegui et al., 2006).

In 2015, a room temperature sequential extraction method using mixed solvents for the recovery of polycarbonates (PC) and other polymers from e-waste was proposed (Weeden et al., 2015). PC was recovered efficiently, but ABS was insoluble in dichloromethane (DCM) and PS formed a gel, which led to difficulty in separation according to the authors (Weeden et al., 2015). To our knowledge, a satisfactory and inexpensive separation and recovery method for the dominate e-waste plastics has not been established at room temperature.

To avoid the separation and recovery process from being disturbed by other components in e-waste plastic, we first attempted to develop a practical recycling procedure at room temperature via a model study. The mutual action of different resins and various additives during recovery was eliminated using laboratory simulation research. New knowledge and methodology for plastic recovery was obtained and can be used as the basis of recovering plastic from e-waste.

In the present study, a mixed solvent extraction system was used with imitation e-waste to create a recovery method for the four dominate plastics in e-waste and is further proposed for future study of e-waste plastic recycling. We chose PC, ABS, PS, and SAN as the target plastics in our lab simulation. Specifically, a synthetic e-waste plastic sample (SEPS) was mixed with virgin PC, ABS, SAN, and PS. The plastics were in the form of pellets, except for PS, which was in powder form. The proposed process comprised of dissolution in the "strong" solvent and reprecipitation in the "weak" solvent for the plastics. The polymers yielded were washed and dried thoroughly. The important tasks of the model study were to determine key factors that affect the recovery efficiency and how to avoid or reduce a significant reduction in the values of the polymers.

2. Potential solvent selection

A previous article (Weeden et al., 2015) showed that no single solvent can selectively dissolve all of the components in e-waste plastics. Sequential extraction with two or three solvents is the only possible choice. To reduce the workload, the Hansen solubility parameters (HSPs) were used to select the potential solvents for plastic recovery. The HSP values depend on the following three types of interactions: dispersion forces (δ_D), permanent dipole–permanent dipole forces (δ_P), and hydrogen bonding (δ_H). In addition, the HSP values also depend on temperature (Srinivas et al., 2009). In the present study, only the HSP values at room temperature were considered to help reduce the complexity of the recovery and the cost. The HSP correlation equation used is as follows (Hansen, 2007):

$$(Ra)^{2} = 4(\delta_{D2} - \delta_{D1})^{2} + (\delta_{P2} - \delta_{P1})^{2} + (\delta_{H2} - \delta_{H1})^{2}$$
(1)

where *Ra* the "distance" between the polymer and solvent; δ_{D} , δ_{P} , and δ_{H} are the three interactions between the polymer (subscript 1) and the solvent (subscript 2). The RED number reflects the relative energy difference:

$$\operatorname{RED} = \frac{R_a}{R_0} \tag{2}$$

where R_0 the experimentally measured radius of the polymer solubility sphere (Table S1). Theoretically, the extent to which the polymer is soluble in the solvent is indicated by RED (RED < 1, the polymer is soluble in the solvent; RED > 1, insoluble; and RED = 1, partially soluble (Achilias et al., 2009; Weeden et al., 2015)). In addition to RED, miscibility, boiling point, cost, toxicity, operability and environmental impact were considered in the solvent selection. Using the HSP for polymers in common solvents, the RED values were calculated and are listed in Table 1. The RED values in DCM for PC, PS, ABS and SAN are 0.17, 0.70, 0.24 and 0.54, respectively; in acetone (ACE), the RED values are 1.25, 1.77, 0.42 and 0.95, respectively; and in acetonitrile (ACN), they are 2.43, 2.88, 0.96 and 1.36, respectively.

There have been inconsistent statements on the solubility of ABS in DCM in previous studies (Achilias et al., 2009; Weeden et al., 2015). Weeden et al. stated that ABS has a negligible solubility in DCM, and DCM can be used to extract PC from ABS (Weeden et al., 2015). However, Achilias et al. (2009) suggested that DCM should be a "strong" solvent that easily dissolves both ABS and PC. In reality, the estimated RED values in DCM for ABS (1:2:4) (Peng et al., 2009), ABS (40SAN) (Peng et al., 2009), ABS-CR (Hansen, 2007), R-ABS (Hansen, 2007) and PC (Hansen, 2007) were 0.64, 0.49, 0.68 and 0.24, respectively. These results indicated that these four ABS are soluble in DCM. For example, R-ABS performs similarly to PC (RED = 0.17 (Weeden et al., 2015)), which suggested that DCM might extract PC from ABS poorly.

The HSP values of the mixed solvents are linearly associated with the solvent component (Srinivas et al., 2009). Hence, the HSP value of the solvent mixture was used to determine the optimum solvent mixture for extraction of the specific polymers. However, the experimental observation was not always consistent with the estimation based on the HSPs. Negligible parameters may affect the HSP predictions, e.g., hydrogen bonds between the solvent pairs (Kamlet and Taft, 1976). For instance, ABS is insoluble in ACN (RED = 0.96), and SAN is soluble in ACN (RED = 1.36). ABS can be extracted via centrifugation after it is dissolved in ACE (21.8%), and the RED value of ABS is 0.42. The Hansen theory may not be applied in some cases based on previous work (Weeden et al., 2015).

3. Dissolution experiment

PC, PS, ABS and SAN dissolved well in DCM. Specifically, 1 g of PC, PS, or SAN dissolved in 10 mL DCM, and 1 g of ABS dissolved in 15 mL DCM with a modest viscosity. For the solubility of the polymers in ACE, 1 g of ABS partially dissolved, i.e., formed suspended particles, in 15 mL of ACE, and 1 g of SAN was completely soluble in 10 mL of ACE, resulting in a transparent solution. PS forms a gel that adheres to the vessel containing ACE. The PC pellet was insoluble in ACE (Table S2). To simplify the recovery procedure, the PC pellet was used in this study instead of PC powder. ACN can dissolve SAN to leave ABS, PC, and PS in their solid form, but PC is partly dissolved in ACN.

ABS is a family of engineering thermoplastics. The mass ratio of monomers in the ABS may affect its solubility in DCM. Moreover, the source of the DCM is also likely to exert an influence on the solubility of ABS. Room temperature solubility experiments with ABS

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