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Review Article

A CONCISE OVERVIEW OF THE SOLVENT AIDED SEPARATION AND EXTRACTION BASED ON THE PLASTICS WASTE

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1. INTRODUCTION

Although technological advancement has been a boon to modern science, somehow it has also been the necessary evil in terms of scientific developments. A research conducted by Barba and Guitierrez in the year 2008 highlighted that most of the waste plastics are landfills or incinerations [1]. Furthermore, 87.6% of the same is contributing to the number of toxic pollutants generated from plastic waste [1]. The average global plastic recycling rate is only about 18% which is much below the standardized numerical figure of around 73.2%[1]. Since the advent of the first synthetic plastic, the usage of plastic has grown dramatically and is containing to increase. This primarily results in overconsumption, disposal, and littering of plastics which ultimately causes in pollution, thus causing serious environmental hazards. Despite there are several plastic recycling technologies that have been proposed over decades, the recycling sector stills face the challenge because of several technical difficulties involved within the recycling method and relatively insufficient profits, especially in mixed plastics [2].

As the concept of green chemistry has evolved throughout the last decade; plastic recycling by solvent extraction turns out to be one of the most favorable techniques to combat waste plastic operations and management[2]. The idea of solvent assisted plastic waste recycling prevailed in the 1980s when Drain et al [3] showed that apart from mechanical recycling, pyrolysis, and gasification, solvent recycling can also be an optimistic approach to address waste plastic recovery. Later, in the year of 1992, Vane and Rodriguez fabricated a selective dissolution process for the separation of the mixed plastics [4]. The basic working principle of plastic recycling using the solvent extraction technique is framed upon the removal of the impurities, for example, the plastic additives, homogeneous or heterogeneous dissolution, and reprecipitation or devolatilization. The polymer assembly is dissolved in the solvents followed by selective crystallization of the individual polymer. The most important aspect of the discussed process is the selection of the suitable solvent which can act as the selecting parameter because the process will be only effective when a solvent can dissolve either the specified polymer or all the other polymers except the specified one [5,6].

2. THE PHYSICAL APPROACH TO THE DISSOLUTION PROCESS

The process of dissolution is controlled by a number of parameters such as the types of polymers and solvents, the size of the polymer molecules, the molecular weight of the system along with dissolution time and concentration [7]. From a thermodynamic perspective, solvent diffusion and chain disentanglement are the two transport processes that are involved during the process of polymer dissolution [8]. As the molecular weight of the system increases, the dissolution gradually shifts to disentanglement. The plasticization of the polymer during the dissolution process results in a gel-like swollen mass with two distinct interfaces, one forming in-between the glassy and the gel phase and the second in between the gel phase and the solvent [9, 10]. The solvents subsequently push the swollen polymer into the solvent which results in an increase in the amount of solvent within the polymer fraction [10]. The phenomenon continues until there is equilibrium between the solvent dissolution and the free volume of the gel in a region of quasi-stability. The increased mass transfer rate accelerates the diffusivity of the polymer and the gradient also affects the particle size and the dissolution time [10].

A typical crystalline polymer consists of an infiltration layer, a gel layer, a liquid layer, and a swollen solid layer. The polymer hence contains numerous perforations and channels of the nanometric level. As the polymer is introduced into the solvent, the solvent fills these voids and starts the process of diffusion without deforming the original structure [11]. The decrease in the stress-energy of the gel layer results in it to disappear indicating the phenomenon of cracking instead of dissolution [12-14]. A typical dissolution process thus consists of three steps, beginning with the penetration of the solvent, followed by the swelling of the polymer and concluding with the diffusion into the solvent.

In reality pressure and temperature also plays a pivotal role in the process of infiltration. Miller-Chou and Koenig had demonstrated the occurrences of cracks in PMMA and PS using benzene as the solvent faster than in toluene due to the high vapor pressure of the system. Furthermore, under critical conditions, the swelling induced within the polymers due to the high vapor pressure of the solvents can be decreased by heating [14]. Therefore, it can be concluded that the dissolution phenomenon does depend on the optimum balance between the pressure and the temperature.

3. SELECTION OF THE OPTIMAL SOLVENTS

The solvents are selected primarily by the solubility rule, which implies that high polar polymers are dissolved in high polar solvents, while the polymers with low polarity can be solubilized by using a solvent with low polarity [15]. The more overlapping are the polarity of the polymer with respect to the solvent used, the easier it is for the dissolution to occur [16]. Apart from the solubility rule, there are other phenomena which determine the dissolution time and the solubilization of the polymer in a particular solvent [16]. For instance, if the polymer-solvent interaction is much greater than the polymer-polymer interaction, the polymer is said to be highly soluble in the respective solvent. On an additional note, if the solubility parameters (δ_D, δ_P, and δ_H) of the polymer and the solvent system are alike, the polymer is more readily soluble in the solvent system [17, 18].

Amongst all the proposed solubility parameter system developed, the Gutmann and Kamlet-Taft parameter system work best when focusing on specific material recycling from waste plastic disposals [19]. The system works by the prediction of the Hansen Solubility Parameter value, which accounts for the material interaction due to the dispersion forces, permanent dipole-permanent dipole forces and forces due to hydrogen bonding. The solubility of the polymer-solvent system is often judged by the solubility parameter distance between the polymer and the solvent (R_a) which is then compared with the relative energy difference (RED) represented as the ratio of R_a to R_0, where R_0 is the radius of sphere and R_a is the distance from a given solvent point to the centre of the sphere. Conventionally it is presumed that if the RED value of the system is less than unity, the polymer is soluble in the solvent system [19, 20].

For a real system, the Hansen Solubility Parameter theory takes into account several supplementary parameters like the electrostatic forces, viscosity, dissolution time, toxicity and the dissolution ability of the solvent [21]. In a recent study conducted by Zhao et al, it was reported that polymers such as Polystyrene, Polycarbonate, Acrylonitrile Butadiene Styrene, and Styrene acrylonitrile co-polymer responded well with this method. The study also demonstrated that each step of isolation of the polymer in the solid state needed a specific solvent [22]. The selected solvents may differ from the ideal one based on the plastic additive composition. Further, it was also noted that density was also one of the prime factors which link the solubility of the polymer to the solvent system in case of supercritical fluidic system apart from high pressure and high temperature [23].

The empirical relationship between the solubility of the material and the density of the solvent can be represented as

$C=\rho k^{[(a/T) + b]}$, where C is defined as the concentration of a solute in the solvent; ρ is the density of the supercritical fluid being used; k is the association number of the solute, and a and b are the solvent and the solute variables respectively [24].

The solubility of the polymer increases with either higher density or higher temperature in the case of the supercritical fluidic system [25]. Finally, if we compare the technologies, supercritical fluid extraction technology is much more efficient than pyrolysis in terms of energy consumption, output efficiency, and environmental hazards [26].

4. A PREFACE TO THE PLASTIC RECYCLING THROUGH SOLVENT EXTRACTION TECHNOLOGY

The basic principle of this technology uses the selective dissolution technique using targeted solvents or supercritical fluids [27]. Usually, the process revolves around two categories of solvent, the first one being the strong solvent which has a positive solubility vale as compared to the specified polymer and the second, termed as the weak solvent having the solubility value less than zero [28, 29]. The primary advantage of this method is that the process effectively removes the additives and insoluble contaminants along with providing superior properties of the recycled product compared with virgin products. A supercritical fluid is a novel approach for segregating polymers from waste including supercritical antisolvent precipitation [30]. Theoretically, the supercritical fluids exhibit the characteristics in-between liquid and gases [31]. These fluids provide with a greater advantage as compared to the conventional solvents in terms of mass and heat transfer, less extraction time and minimal residues in the final product [31]. Till recent times, these fluids are extensively used in selective depolymerization. The recent burgeoning progress in science has made these exclusive classes of fluids to particulate n various extraction and mechanical process. Carbon dioxide is used as one of the most efficient supercritical fluid because of its efficacy and non-toxic nature [32]. The polymers are dissolved in these solvents and subsequently extracted by the solvent extraction process [33]. One of the major setbacks which these supercritical fluids suffer from is its high cost. Since these solvents are green solvents, despite the cost being higher, the efficiency can be compensated by low energy consumption, minimal solvent loss, and high output rate [34].

5. REVIEWING THE QUALITY AND THE EFFICIENCY OF THE SOLVENT EXTRACTION PROCESS

For examining process efficiency, parameters such as the composition, the physiochemical material degradation, and the morphological changes have to be evaluated. Therefore several strategies, for instance, restabilization, rebuilding, compatibilization, and the addition of elastomers and fillers are often studied to promote the efficacy of the process.

5.1. Polymer Identification and Testing

The primary technique used to identify the composition of the material present in the waste plastic is by Fourier Transform Infrared Spectroscopy (FTIR) [35]. The spectrum is then compared with a normalized graph produced from the virgin model of the target polymer. Apart from FTIR, DSC, NIR and Raman Spectroscopy may also be used to detect the composition of the waste plastic without any sample pretreatment [36]. In case of a mixed polymeric system, HPLC is used to predict the probable structural changes of the sample [37]. The most common method used to predict the molecular weight distribution of the recovered polymer is either by the viscosity method or by GPC. While DSC can be employed to study the thermal degradation of the polymer system, TGA can be used to estimate the composition of the fillers, additives, volatile agents and the nonpolymeric toxic agents. Further, the stability in terms of thermal and oxidation can be evaluated by chemiluminescence [37].

5.2. Compatibility of the Polymer Blends

Morphology and the mechanical properties of the polymer products depend heavily on the composition of the plastic blends within them. Furthermore, the different recycling routes also affect the strength and the overall elongation of the material. DeSouza et al had verified the same by inferring that a plastic blend of ABS and HIPS (20:80 in weight) recycled via different routes showed a variation in their tensile stress and elongation at break [38]. This may be attributed to the fact that the higher purity within the blend system had enhanced the self-compatibilization property which in turn supported the enhancement in the mechanical properties. This implies that the purity of the recovered plastics blends combines the mechanical properties within each of them to yield a unique enhanced overall mechanical property [38, 39]. Hence, as the efficiency of the recycling process is increased, the gross cost of the solvent extraction process is decreased simultaneously. In reality, the blends of the recycled polymers such as PVC and ABS can still be recommended at the commercial level because of its overall improved mechanical properties and low cost. In the same way, PVC and PS are two immiscible polymer components because of their high interfacial energy and repulsive interaction. Using a compatibilizer such as Ethylene methyl Acrylate can covert the same into a miscible blend yielding comparable mechanical properties [40].

5.3. Additives and Impurities

During its service life, the plastic product absorbs several types of additives and impurities like brominated flame retardants, thermal stabilizers, UV resistors, clarity modifiers, and impact modifiers [41]. These, especially brominated flame retardants can cause serious toxic effects in the recovered polymer if not separated. Although Soxhlet extraction is one of the most widely used separating technique, newer developments such as extraction with supercritical fluids, solvothermal treatments, ionic liquids or microwave-assisted extraction have proved to be efficient than the conventional process. Li et al had fabricated a method based on microwave-assisted extraction which successfully separated brominated flame retardants from Polystyrene using isopropanol/n-hexane as the solvent [42]. Zhang et al had demonstrated the extraction of tetrabromo bisphenol A from e-waste plastic via the solvothermal process in the liquid to solid ratio of 15:1 (mL/g), at 90°C for 2 hours. Several of these techniques have been commercialized such as Creasolv process which can be implemented at a scale of 10000 tons/year. Arias et al had used Supercritical CO_2 at 45Mpa and 80°C to extract additives such as Irganox 1076 and Irgafos 168 from LDPE and LLDPE films. Anour et al studied the separation of additives in food grade polyolefin and proved that as compared to the classical solvent extraction technique, the supercritical fluid extraction methods proved to be simple, fast and efficient [43].

The low molecular weight organic dyes which are added in the colored plastics can be separated by the precipitation of the polymer with a non-solvent since the solution possesses entropy of higher order than that of the polymer. Nowadays, pigments can be recovered by the amalgamation of selective dissolution and evaporation from Polypropylene and polyvinyl butyral films. Additives such as titanium dioxide, chromium (III) oxide in acrylonitrile-butadiene-styrene can be extracted via the Creasolv process using acetone/water as the solvent for the medium [44].

5.4. Consecutive recycling cycles

Poulakis and Papaspyrides et al had carried out an experiment which aimed at reviewing the mechanical properties of the polymer after multiple recycling cycles [45]. Surprisingly, no significant differences between the mechanical properties of the recycled polymer and the virgin polymer were noted. Considering a system of acrylonitrile-butadiene-styrene, no changes were observed at a low strain value, but at the high strain, the yield stress and the impact strength decreased during the first couple of recovery, due to the butadiene degradation and the low molecular weight of the styrene-acrylonitrile. However, these values when subjected to high strain remained constant for the rest of the cycles. Drawing the same analogy from this experiment, the elongation at break and the melt flow index of Polypropylene increased from the fifth cycle because of the exhaustion of the stabilizers in the primary recycling cycles but henceforth remained constant throughout up to the tenth cycle.

At times, the molecular weight distribution plays a crucial role in determining the mechanical properties of the plastics [46]. Several hypotheses have been projected which relates the molecular weight distribution to the number of recycling cycles and the mechanical properties. On one hand, polycarbonates with high molecular weight distribution improve the toughness and the environmental stress cracking resistance; while the low molecular weight distribution improves the processing mechanisms. While on the other, the molecular distribution of Polyethylene Terephthalate has no role to play in terms of the product's number of recycling cycles and mechanical properties [47, 48].

5.5. Forms of the recycled plastics

Various polymers in different solvents result in various final forms such as agglomerates, powder, granules, and jelly polymer lumps [49]. For instance, the accumulation of a specific polymer resisting the separation between the two phases of solvent solid results in the formation of the gel. Not only low-temperature evaporation results in the polymer forms to alter, but there are also several mix solvents available which can alter the precipitation form. Speaking generically, the most approved form of the recycled plastics is in the form of powder and granule. Poulakis and Papaspyrides had designed an experiment in which they had dissolved polypropylene in Xylene at 135°C that was collected as small grains in its final form. The same possessed excellent recoveries of both polymer (100%) and solvents (>80%) without any degradation or decomposition. Several investigations about the particle size confirmed that smaller particle size had elevated tensile strength and improved tensile strength. Additionally, Poulakis and Papaspyrides had discovered that as the grain size of the (94% recycled) plastic material decreased below 1nm, the recycled polymer mass was found suitable for further processing without any fall in term of dimensional stability [49-50].

6. FUTURE SCOPE AND OUTLOOK

Although the plastic waste recycling technique involving solvent extraction is more effective in terms of energy and environment friendliness, it still faces certain challenges and problems with respect to the practical usability of the method [48]. Generally, most of the wastes plastic that is generated is a mixed plastic component. Hence, the major challenge is to extract and recycle the individual plastics separately. As on one hand, the dissolution of these mixed plastics is very complicated in either pure or mixed solvents because of the difference in their solute particle interactions; on the other hand, the anomalous phase formations such as gel or jelly phase hinders the extraction process due to the difficulties in centrifugation and filtration [51]. This is why there has been a recent growth in the researches and investigations focusing on the multi-plastic extraction and recovery process. The presence of the solvents along with traces of impurities in the extracted plastic product is also a matter of concern considering these extraction technologies [51]. Moreover, a continuous inflow of atmospheric or vacuum distillation gradually degrades the quality of the recycled product. In reality, as the waste product does not possess a consistency in the composition, the same gets reflected in the extracted or recycled plastic products causing the efficiency and the homogeneity of the entire process to drop. Taking into consideration, all these factors, the purity and the efficiently of the process should be carefully mapped bringing an optimization between these two aspects of the technology.

Analyzing the life cycle of the polymer, which consists of processing, shelf life and disposal should be carefully analyzed because the degradation of the polymer during its shelf life affects the life cycle of the polymer after it has been recycled. From the environment's point of view the removal of the toxic materials especially the flame retarders should be eliminated in the primary stage while recycling the plastic waste materials. Economically, after all these recycling stages, the properties of the finished products should be at par with the virgin polymer, so as to be acceptable in the competitive market of plastics [37].

To address this market pricing for recycled products, several scientists and engineers have developed numeral cost-effective methods of recycling plastic wastes along with upbringing of the backdated technologies. Apparently, the high costs of the recycled plastics can be compensated by novel technologies and innovative processes[21]. Although the selection of the perfect solvent is very important for the extraction process to be feasible, the reduction of the extraction time is also crucial at the same time. Choosing non-toxic and nonpoisonous solvents which are less costly for (natural solvents such as terpene oil) can be the way forward an improved recycling methodology. If the cost can be compensated by the process, Green solvents like supercritical fluids are a great choice for the solvent extraction method of plastic recycling. If the solvents that are being used for the extraction process can be completely recovered, the process will turn out to be very cheap and convenient.

The upcoming decades will definitely experience a rise in the plastic recycling process, methodology, and technology in a more economic friendly way, keeping the growth in the usage of plastic products and the waste generated in the background.

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