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PROCESS OPTIMIZATION OF SOLVENTS ASSISTED POLYETHYLENE WASTE RECYCLING

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ABSTRACT

The global production and consumption of plastics has been increasing continuously. However, disposing of plastic wastes in landfill is becoming undesirable due to poor biodegradability. The re-utilization of plastics together with the development of green technologies is mandatory, hence chemical recycling offers a promising alternative as a potential for plastics to maintain their original properties. The scope of this research is focused on the recycling of plastic waste by the chemical dissolution method to optimize the reaction variables in the dissolution/precipitation method using a statistical experimental design. Optimization of the recycled plastics was done using response surface methodology based on box Behnken design. The optimum yield of the recovered plastic 93.4% was achieved at a temperature of 30^0C, a residence period of 30min and a solvent/non-solvent ration of 1:2 v/v. The mechanical and physical properties of the recovered plastics revealed similar value to that of the virgin polymer. The FTIR spectrum of the recycled plastics shows a less significant structure alteration when compared with the spectra of the virgin polymers. Similarly, the thermal stability of the recycled plastics determined using thermogravimetric analysis (TGA) shows a less significant difference when compared with the reported thermal stability of the virgin polymers. Therefore, the study infers that chemical recycling (dissolution/precipitation) can be an appreciable option for getting rid of waste plastics in the environment as well as providing feedstock for the reproduction of plastics.

Keywords: dissolution, precipitation, polyethylene, solvents, optimization, recycling

INTRODUCTION

To reduce the menace caused by plastic waste, a number of recycling techniques have been employed, these include; mechanical, thermal, and chemical recycling (Panda *et al.*, 2010). Due to its practicality and financial viability, processing plastic wastes into liquid goods with additional value by simple heating or a chemical process has been the preferred solution.

Global plastic output reaches 368 million metric tons in 2019; and by 2050, it is anticipated to quadruple (Tiseo 2020). Approximately, 7,300 million metric tons of plastics have been manufactured globally between 1950 to 2019 since the start of large-scale plastics production (Geyer *et al.*, 2017). In the past 70 years, the average annual plastic production around the world has increased enormously. There will likely 850 to 950 metric tons of plastic trash produced annually as the polymer sector continues to expand (Degnan and Shinde 2019). Concerns about the development of plastic trash have grown as just 9% of waste plastics are recycled, 12% are burned, and 79% end up in the landfills (Geyer *et al.*, 2017).

Since currently there is no efficient way to separate plastic garbage, some of the plastic waste that is sent to facilities for material recovery is turned away. As the rate of plastic trash generation increases, majority of discarded plastics will become ineffective as the surface area of the world is finite. Furthermore, when waste plastics are unlawfully dumped into the ocean or are transported from the land by natural phenomena, they account for 80% of marine debris (UN General Assembly 2018).

Many substances that are utilized in making plastics are known to be endocrine disruptors and carcinogens, causing neurological, reproductive, immunological, and developmental problems (Li *et al.*, 2018). The risks of human exposure to the 74,000–113,000 microplastic particles the average American consumes, drinks, and inhales each year are unknown (Cox *et al.*, 2020). Better mitigation strategies are therefore required to deal with the problem of plastic waste. Low-efficiency recycling procedures and ineffective sorting techniques are significant barriers to waste plastic recycling (Schwarz *et al.*, 2021).

Primary, secondary, tertiary, and quaternary recycling are the four main categories into which plastic recycling can be broken down (Kumar 2021). Recovering and reprocessing pure polymer streams is considered primary recycling. Sorting the polymer waste stream, reducing the material's size, and reprocessing it so that it is downcycled are all necessary steps in secondary recycling (Sahajwalla and Gaikwad 2018). The proportion of waste plastics is increasing daily in municipal solid wastes despite the several developed methods for its mitigation such as mechanical recycling, landfill and incineration. Probably, the drawbacks of these methods (water pollution due to highly polluted leaches, greenhouse gas emissions and local inconvenience, high labour cost and waste homogeneity) make such technologies counterproductive thus, the need for sustainable, environment-friendly and cost-effective methods. This research is focused on the recycling of plastic waste by chemical dissolution method to optimize the reaction variables in the dissolution/precipitation method using a statistical experimental design

MATERIALS AND METHODS

The chemicals and solvents used include methanol (99.8%, GHTECH), Xylene (99%, Sigma), n-hexane (99%, M&B), Acetone (99%, LOBA CHEMIE), Benzene (99%, CHEMSAVERS), dichloromethane (99, CDH) and Chloroform (99, LOBA CHEMIE). The chemicals were procured from a local vendor and used without further purification.

Sample Collection

The sample used is low-density polyethylene waste plastic. The sample was obtained from household wastes. The sample

was rinsed with tap water and cut into squares of approximately 1cm square in area.

Experimental design

Minitab 16 statistical software was used to design the experiment based on Box–Behnken response surface methodology. Three independent variables were investigated, each at three levels (Table 1) selected based on preliminary experiments and a literature survey (Ehimen *et al.,* 2010). Each trial was duplicated to obtain 30 trials.

Table 1: The reaction variables investigated and their respective levels

Factors	Low level	High level
Temperature (^{0}C)	25	30
Residence period (Min)	20	30
Solvent/non-solvent volume ratio	1	3

Description of the Experimental Trial

In each trial, the sample of the waste polymer (1 g) and solvent (10 cm^3) was added into a flask equipped with a vertical condenser. The flask was heated for 30 min, the flask was then allowed to cool, and the solution of the polymer was properly poured into a non-solvent (10 cm^3). The polymer was then precipitated, filtrated, washed and dried (Achilias *et al.,* 2009).

$$\text{Recovery yield} = \frac{weight\ of\ the\ dry\ ppt}{initial\ weight\ of\ the\ sample} \times 100 \qquad (1)$$

Physical Properties of Recovered Polymer

The property of the recovered polymer was determined using the America Society of Testing Materials (ASTM) standard methods. Properties determined include specific gravity and density ASTM D792-20 (2020), Water Absorption Capacity ASTM D 570-98 (2018) and melting point determination ASTM 324-16 (2016).

Mechanical Properties of the Recovered Polymer

The property of the recovered polymer was determined using the America Society of Testing Materials (ASTM) standard methods. The properties determined include tensile strength, percentage elongation and young modulus ASTM D638-14 (2014).

Characterization of the Recovered Plastic

The FT-IR spectrometer was used to characterize the chemical structure of the recovered polymer. The thermal stability of the recovered polymer was determined using TGA.

RESULT AND DISCUSSIONS

The yield was obtained from the 30 experimental runs conducted at different levels of the four reaction variables investigated. The yield varies from a minimum of about 71.4% to a maximum of 93.4%. The yield increased sharply from 71.4 to 93.4% as the temperature increased from 25^0c to 30^0c. Similarly, the yield increased from 71 to 93.4% with an increase in the reaction time from 25min to 30min. The respective changes in the reaction temperature and time appears to have a significant effect on the polymer yield.

Effect of the Reaction Variables on the Polyethylene Yield

Table 2 shows the results of the analysis of variance. In general, the model is a good representation of the experimental data (R^2 = 73.34%) with good prediction power of (R^2 =54.30%), although with a significant lack of fit (p = 0.000). Nine terms; three linear terms, three square terms and three interaction terms were analyzed. The results show that all the three linear terms are statistically significant ($p < 0.05$, at $\alpha = 0.05$). Two of the square terms are significant and two terms are statistically insignificant ($p > 0.05$, at $\alpha = 0.05$) in the two-way interaction (Table 2).

Following the elimination of the statistically insignificant terms identified in Table 2, regression analysis gave a new model (Equation 1) with only five significant terms. The new model is not only relatively simple but, with an adjusted R^2 of 73.41%, it is slightly better than the previous model (adjusted R^2 = 73.34%). It also has slightly better prediction power (R^2-pred = 64.63% vs. 54.30%) with a significant lack of fit (p = 0.000).

Table 2: Result of Analysis of Variance for the Recovery of Polyethylene

Source	DF	Adj SS	Adj MS	F-value	P-Value
Model	9	1111.12	123.458	9.87	0.000
Linear	3	901.79	300.597	24.02	0.000
Temperature (^{0}C)	1	745.29	745.290	59.56	0.000
Residence Period (min)	1	94.09	94.090	7.52	0.013
Solvent/non solvent (r)	1	62.41	62.410	4.99	0.037
Square	3	72.11	24.038	1.92	0.159
Temperature (^{0}C)*Temperature (^{0}C)	1	64.99	64.993	5.19	0.034
Residence Period *Residence Period	1	0.00	0.002	0.00	0.990
Solvent/non solvent *Solvent/non solvent	1	10.05	10.051	0.080	0.381
2-Way Interaction	3	137.22	45.740	3.66	0.030
Temperature(^{0}C) *Residence Period	1	7.22	7.220	0.58	0.456
Temperature (^{0}C)*Solvent/non solvent (r)	1	32.00	32.000	2.56	0.125
Residence Period *Solvent/non solvent	1	98.00	98.000	7.83	0.011
Error	20	250.26	12.513		
Lack-of-Fit	3	190.59	63.530	18.10	0.000
Pure error	17	59.67	3.510		
Total	29	1361.39			

Key: DF = degree of freedom, Adj SS = adjusted sum of squares, Adj MS = adjusted mean squares, F-value = F-statistics values.

Table 3: Predicted Result of Optimization and Validation of Polyethylene

Solutions	Temperature (^{0}C)	Residence Period (min)	Solvent/non solvent (v/v)	Yield (%)		Desirability
				Predicted	Experimental	
1.	30	30	1	92.45	93.00	0.959746
2.	30	20	3	91.55	92.40	0.921610

The expected outcomes for optimal solutions were attained through optimization as shown in Table 4.3 with optimal process conditions of temperature 30^{0}c and 30^{0}c, residence period of 30^{0}C and 20^{0}C and solvent/non-solvent ratio of 1:1 v/v and 1:3 v/v in the validation process, however, the solutions predicted maximum dissolution yield of 92.45% and 91.55% with the desirability of 0.959746 and 0.921610 respectively. Dissolution yield of 93% and 92.4% was obtained from validation experiments carried out at the levels of the process variables predicted in dissolutions 1 and 2. It was observed that the optimized predicted and experimentally validated results for the two solutions 92.45% and 91.55% were 93% and 92.4% respectively. Thus, with the predicted yields of 93% and 93.4% which is similarly closer to the experimental yields of 92.45% and 91.55% it indicates a relative deviation of 0.55% and 0.85% respectively. The optimization model is trustworthy, which makes it more desirable for larger-scale dissolution/precipitation of plastic waste for recycling. Optimization and validation of the recovered polymer was achieved with less solvent, in a much shorter time and at a lower temperature (30^{0}C) (Muhammad *et al*., 2019).

Table 4: Physical Properties of Recovered Polyethylene

Testing type	LDPE	
	Virgin (ASTM)	Recycled
Density g/cm^3	0.91-0.94	0.92± 0.01
Specific gravity	0.91-0.96	0.91 ±0.02
Melting point (^{0}C)	115-135	130±0.8
Water Absorption capacity (Weight Increase %)	0.06	0.04 ± 0.00

Values are expressed as mean ± standard error of three replicates

Table 4 presents some properties of the recovered polyethylene at the optimal condition in comparison with international standards of testing materials (ASTM). From Table 4; the reported density, specific gravity, melting point and water absorption capacity are however, within the ASTM standard value. Hence recycled plastic has essentially retained its relative physicochemical properties. A decrease in property lowers the price of a raw material, which in turn lowers the cost of production (Abu-Zahra 2002).

Table 5: Mechanical Properties of the Recovered Polyethylene

Testing type	Virgin (ASTM)	Recycled
Tensile strength (MPa)	9.7	8.6± 0.80
Percentage elongation %	80-850	96.9±0.08
Young modulus MPa	300	286.6±02

Values are expressed as mean ± standard error of three replicates

The mechanical properties of the recycled plastics are presented in Table 5. The results show that the tensile strength of the polymer slightly decreased after recycling, this is in line with the study of (Zhao *et al*., 2018) who reported a slight decrease in the tensile strength of recycled plastics which is possibly due to a decrease in the molecular weight of the plastic. The result is in consistent with the assertion of (Murat *et al*., 2020), who reported that, the mechanical properties of the recycled Low Density Polyethylene was found to be closer to their virgin materials after recycling, hence they have huge potential for recycling.

Characterization

FT-IR Spectrum of Polyethylene

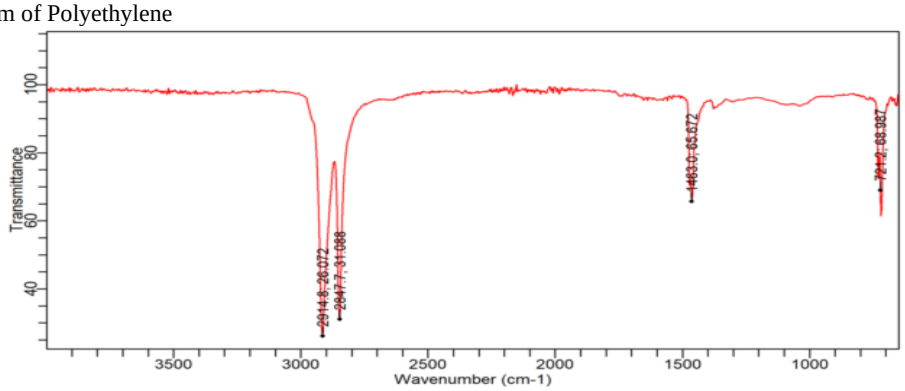

Figure 1: infrared spectrum of PE in the range of 650–4000 cm^{-1}

The infrared spectrum of PE in the range of 650–4000 cm^{-1} is shown in Fig. 4.10. The absorption peak located at 2914cm^{-1} is assigned to CH_2 asymmetric C-H stretch, absorption peak seen at 2847 cm^{-1} corresponds to CH_2 symmetric C-H stretch vibration. The absorption peak observed at 1463 cm^{-1} is assigned to $–CH_2$ bending vibration and finally Absorption peak observed at 721 cm^{-1} is assigned to C-CH_2 rocking vibration which is a strong indication of the presence of polyethylene. Similar absorption bands was also observed by (Smith 2021) who also reported absorption at 2914, 2846, 1474 and 720 cm^{-1} in the FT-IR spectrum of polyethylene.

Thermogravimetric Analysis of the Recovered Plastics

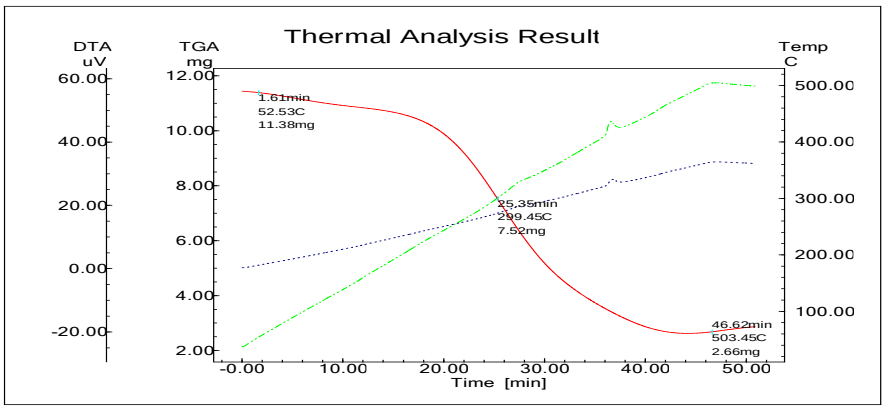

Figure 2: The thermogravimetric Analysis (TGA)

The thermogravimetric Analysis (TGA) shows that 12.00 mg of the polyethylene was reduced to 11.38mg at 1.61min and a temperature of 52.53^0c this might be due to loss of moisture and other impurities absorbed by the polymer. At 25.35 min and a temperature of 299.45^0c, the weight of the plastic was further reduced to 7.52mg. As the temperature continues to rise by a temperature of 503.45^0c and a time of 46.62 min, the weight was significantly reduced to 2.66mg. This is in conformity with the study of (Muhammad *et al.,* 2015) who reported the degradation of virgin polyethylene polymer degrading at a temperature above 480^0C.

CONCLUSION

This study investigated the recycling of plastic wastes by dissolution and precipitation method using xylene as solvent at varying temperatures and times. Xylene and methanol were found to be the most suitable solvent for the dissolution and precipitation of the plastic wastes among the tested solvents systems. The polyethylene plastic waste was successfully recycled at an optimal yield of 93.4% under a reaction temperature of 30^0C and a reaction time of 30min with 1:2 solvent/non-solvent ratio. The recycled plastics were found to have maintained their mechanical properties with only an insignificant decrease. The physical properties analyzed for the recycled plastics were found to have similar properties to the virgin plastics. The FT-IR spectra of the recovered plastics were found to have similar IR spectra of the virgin polyethylene, similarly, the TGA curves were also found to have maintained their thermal stability. The characterization of the recycled low-density polyethylene conducted in this study has provided critical insight in their recyclability potential and their suitability to be used in other applications such as battery cables, Composites bins, Egg cartons, Disposable cutlery, floor tiles, Bottle and lumbers. Consequently, by recycling these wastes, water, land and air pollution can be reduced, which can bring benefits to everyone for a greener and more sustainable future. Hence, more research should be performed on the upscaling of solvent-based techniques, including medium recovery, search for new and greener solvents to increase the potential and further explore the circular economy for plastics.

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Appendix A

Table 6: Optimization process of polyethylene monomer and the yield obtained

StdOrder	RunOrder	PtType	Blocks	Temperature	Residence Period	Solvent/non solvent	Yield%
1	1	2	1	25.0	20	2	74.6
2	2	2	1	30.0	20	2	91.2
3	3	2	1	25.0	30	2	69.8
4	4	2	1	30.0	30	2	92.4
5	**5**	**2**	**1**	**25.0**	**25**	**1**	**71.4**
6	6	2	1	30.0	25	1	86.6
7	7	2	1	25.0	25	3	83.8
8	8	2	1	30.0	25	3	86.8
9	9	2	1	27.5	20	1	73.4
10	10	2	1	27.5	30	1	90.2
11	11	2	1	27.5	20	3	83.0
12	12	2	1	27.5	30	3	86.0
13	13	0	1	27.5	25	2	83.6
14	14	0	1	27.5	25	2	83.0
15	15	0	1	27.5	25	2	86.0
16	16	2	1	25.0	20	2	72.4
17	17	2	1	30.0	20	2	89.6
18	18	2	1	25.0	30	2	74.6
19	**19**	**2**	**1**	**30.0**	**30**	**2**	**93.4**
20	20	2	1	25.0	25	1	72.6
21	21	2	1	30.0	25	1	82.4
22	22	2	1	25.0	25	3	79.6
23	23	2	1	30.0	25	3	85.6
24	24	2	1	27.5	20	1	76.4
25	25	2	1	27.5	30	1	91.8

26	26	2	1	27.5	20	3	85.2
27	27	2	1	27.5	30	3	86.4
28	28	0	1	27.5	25	2	84.6
29	29	0	1	27.5	25	2	86.4
30	30	0	1	27.5	25	2	87.8