CHEMICAL ENGINEERING TRANSACTIONS

VOL. 94, 2022

A publication of AIDIC

The Italian Association of Chemical Engineering
Online at www.cetjournal.it

Guest Editors: Petar S. Varbanov, Yee Van Fan, Jiří J. Klemeš, Sandro Nižetić

ISBN 978-88-95608-93-8; **ISSN** 2283-9216

DOI: 10.3303/CET2294209

Plastic Waste Circularity with Data-Driven Approach Considering Polymer Heterogeneity

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Plastic debris has been a consistent issue in the global environment, and plastic waste recycling is the most promising option to avoid further accumulation. The waste quality serves as a crucial restriction for recycling planning and requires proper definition. This work aims to extend the previously developed data-driven approach to quantifying the recyclability clusters of plastic waste to consider polymer heterogeneity in the evaluation. Heterogeneity estimation is conducted by identifying the compatibility between polymers based on their surface tension. The applicability of the polymers mix (targeted and non-targeted polymers) can then be decided based on the Q-value approach. The method identifies the quality class of the plastic mixture based solely on the compatibility of the polymer, where the quality trend varies for each identified compatibility class. This gives insights into the suitability of mixing different polymers type prior to recycling. The recycling potential/circularity of the plastic waste can then be identified based on the Plastic Pinch Analysis, which outputs the ideal maximum external plastic demands with a certain threshold grade of the plastic (Pinch Quality). A case study is shown using three types of polymers: Polyethylene Terephthalate (PET), Polyethylene (PE) and Polypropylene (PP), to showcase the polymers heterogeneity evaluation. The results show that around 32.4 % of disposed PP waste could potentially be mixed with PE to have a compatible mixture. However, it is also crucial to check the properties of the mixed polymers to fulfil the demands of site requirements prior to recycling.

1. Introduction

Plastic waste accumulation has been a worldwide issue for decades and requires considerable attention to solve the problem systematically. The main priority is to design a systematic plastic reusing and recycling framework to reduce further the environmental impacts of plastic waste, especially when this issue became more alarming during the COVID-19 pandemic due to high levels of personal protective equipment use, overpackaging of delivered goods, and mass vaccination (Klemeš et al., 2021).

Previous plastic recycling studies can be found. Huysman et al. (2017) proposed that the compatibility between polymers plays the main role in deciding the recycling options for plastic waste. The waste substitution ratio is also mainly decided by the compatibility between polymers. Gradus et al. (2017) found that the cost of plastic treatment in Dutch recycling facilities can be reduced using post-separation of waste rather than source separation. Eriksen et al. (2019) derived several categories for plastic waste mainly based on their application types, including food application, medicine or toys. Their assessment is mainly dependent on two main properties: polymeric composition and other general residues of the polymers. Faraca and Astrup (2019) analysed the waste in Denmark facilities and categorised the waste in terms of applicability, impurity, lifetime, and polymeric compositions. Brouswer et al. (2020) proposed a qualitative categorisation of plastic package waste depending on the physical and chemical properties, also mainly depends on the application categories for the plastic packages. Min et al. (2020) have collected the critical factors for ocean plastic degradation from various literature sources and produced a decision support model to evaluate the critical factors in identifying

Paper Received: 26 May 2022; Revised: 05 June 2022; Accepted: 13 June 2022
Please cite this article as: Chin H.H., Varbanov P.S., Klemeš J.J., Tan R.R., Aviso K.B., 2022, Plastic Waste Circularity with Data-Driven Approach Considering Polymer Heterogeneity , Chemical Engineering Transactions, 94, 1255-1260 DOI:10.3303/CET2294209

plastic degradation. Varbanov et al. (2021) have recently proposed a data-driven prototype for quality-oriented plastic waste recycling, and extension by Chin et al. (2022) to consider various properties. The approach gathers different complex properties of plastic waste and identifies the clusters based on the available data set, where each cluster represents quality grades. This can be subjective to different opinions as there are various properties that can be considered the critical factor.

A critical gap remains in the lack of a well-defined plastic waste recyclability index considering the heterogeneity of polymers. Huysman et al. (2017) demonstrated a possible method for evaluating the compatibility between polymers, but it was not indicative enough for the quality of plastic waste as the properties of the polymers are still the major restrictions. Plastic waste trading is still restricted by the current recycling capacity due to the lack of a plastic chemical information system and compatibility assessment. This work aims to extend the work from Chin et al. (2022) in quantifying the qualities of the plastic waste and coupled with the compatibility estimation from Huysman et al. (2017) to derive a proper plastic waste grading system.

2. Data-driven Plastic Pinch Analysis

The use of a data-driven framework to quantify the quality clusters for plastic waste streams is proposed. The plastic waste samples are first distinguished into various polymer types. The property data samples for each polymer type are then fed into the Machine Learning model to learn their quality patterns. The Pinch Analysis method followed up by Machine Learning allows users to gain crucial intuitions on the resource quality bottlenecks of the current production or recycling system. The quality grades from the Machine Learning model can be used as the quality indicator in the Pinch Analysis framework. Pinch Analysis is an efficient framework to target minimum external resource requirements, and the solutions can be presented in a graphical manner – see Figure 1. More information on the Machine Learning approach can be found in Chin et al. (2022).

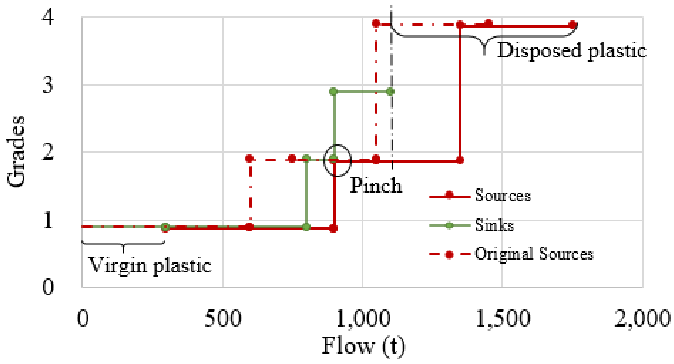

Figure 1: Plastic Pinch Analysis framework in evaluating minimum virgin plastic and disposed waste

3. Polymer heterogeneity

Plastic Pinch Analysis could potentially identify the minimum virgin plastic flow, but it ignores the heterogeneity of the polymers mix. In reality, it is difficult to achieve full separation of the plastic waste streams into individual polymer types. Different polymer types can have different chemical natures. Mixing them could cause an immiscible mixture which can jeopardise the properties of the waste, especially mechanical properties (Lei et al., 2009).

Huysman et al. (2017) proposed the waste quality prediction approach mainly based on the compatibility between polymers in a mix. The assumptions of the approach are the mixture is binary (two polymer types), the effect of thermo-mechanical degradation is not accounted for, and no additional agent is added to the mixture. The compatibility is determined by the interfacial tension between polymers in a molten state. Other approaches are available such as Flory-Huggins parameters, Hildebrand solubility parameters or mixing enthalpy function. The interfacial tension method is more straightforward and can identify the miscibility between two fluids easily. Other approaches can be used, and results comparison can be made.

The interfacial tension can be regarded as the surface energy (γ_{ij}) for the polymers 'i' and 'j', which are contributed by the intermolecular forces, dispersion forces and polar forces (Fowkes, 1964). Wu (1971) has proposed a geometric average mixing rule for surface tension estimation for binary polymers to account for the dispersion (γ_{id}) and polar components (γ_{ip}) of the polymers, where 'i' represents the polymer types – see Eq(1), where γ_{12}, γ_1, γ_2 represent the respective surface tension for mixture, polymer 1 and polymer 2. The estimation has an accuracy of close to 96 % as the polar interaction is accounted. This mixing rule is more commonly used for predicting the surface energy of polymers as compared to other available methods.

$$\gamma_{12} = \gamma_1 + \gamma_2 - 4\left(\frac{\gamma_{1d}\gamma_{2d}}{\gamma_{1d} + \gamma_{2d}}\right) - 4\left(\frac{\gamma_{1p}\gamma_{2p}}{\gamma_{1p} + \gamma_{2p}}\right) \quad (1)$$

Huysman et al. (2017) conducted the calculation of the binary surface tension between the commonly found polymers, and their results are presented in Table 1. They have derived various compatibility classes based on the range of interfacial tension γ_{12} for binary mixture, i.e., perfectly compatible, reasonably compatible, limited compatible and incompatible. In addition to the surface tension estimation, they have proposed a quality value called the Q-values for each compatibility class. The Q-value represents the quality of the plastic waste based solely on the polymer compatibility. The range of surface tension for each compatibility class is presented in Table 2. For example, the perfectly compatible mixture is the LPE/PE since their surface tension falls on the 'perfectly compatible' class. PP/PE pair is still reasonable compatible, but PP pairs with PS have limited compatibility. The completely not compatible mixture is PE, PP and LPE with PA.

Table 1: Surface tension for common polymers (γ_{12}), in mN/m, data obtained from Huysman et al. (2017)

	Low-density polyethylene (LPE)	Polyethylene (PE)	Polypropylene (PP)	Polystyrene (PS)	Polyvinyl chloride (PVC)	Polyamide (PA)	Polyethylene Terephthalate (PET)
LPE	-	0.002	0.477	6.121	2.192	14.15	9
PE	0.002	-	0.413	6.109	2.236	14.12	9.001
PP	0.477	0.413	-	6.4	3.27	14.09	9.46
PS	6.121	6.109	6.4	-	2.413	3.16	0.574
PVC	2.192	2.236	3.27	2.413	-	9.68	4.657
PA	14.15	14.116	14.092	3.165	9.681	-	0.012
PET	9	9.001	9.46	0.574	4.657	0.012	-

Table 2: Compatibility class for different ranges of binary surface tension. Data obtained from Huysman et al. (2017)

Compatibility	Min γ_{12}	Max γ_{12}
Perfectly compatible	0	0.1
Reasonably compatible	0.1	1
Limited compatible	1	10
Not compatible	10	-

In this work, the Q-value approach is utilised to analyse the potential sources and sinks in the plastic recycling system. Based on the Plastic Pinch Analysis, the disposed of plastic waste from different polymers could be mixed to achieve a potential recyclable plastic for other processes. This study proposes and demonstrates the evaluation of polymers heterogeneity based on the results from previous Plastic Pinch Analysis. Figure 2 shows the potential integration of both methods, where the disposed waste identified from Pinch Analysis can be mixed to produce a new stream of sources that can be used for other processes.

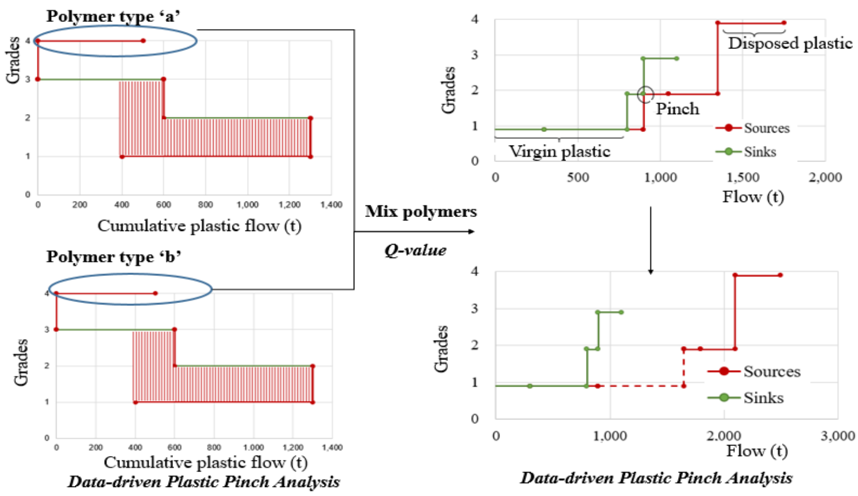

Figure 2: Potential Integration of Plastic Pinch Analysis and Q-value approach

The Q-value is dependent on the number of contaminated polymers. Depending on a certain range of the Q-value, the recycling options for the plastic waste can be decided, and they have suggested four classes for the plastic waste per compatibility class. Figure 3 shows the graphical correlations of Q-value between polymers contamination for each compatibility class. Closed-loop represents the waste that can fully substitute virgin plastic to manufacture the same product. Waste in a semi-closed loop can partially substitute virgin plastic for the same product. Waste in an open-loop cannot be reused for the same product but can be used for other products, while incineration represents the waste is not reusable/recyclable. It is worth noting that Q-value drops at a faster rate when the compatibility is poorer. Note that the Q-value is based on the results from previous literature and should be validated with an empirical approach in the future. However, this preliminary model could shed light on the heterogeneity of polymers. Note that the perfectly compatible mixture is assumed to be of high quality than can be recycled close-loop – see definition of the recycling options in Huysman et al. (2017).

(a) Reasonable compatible

(b) Limited compatible

(c) Not compatible

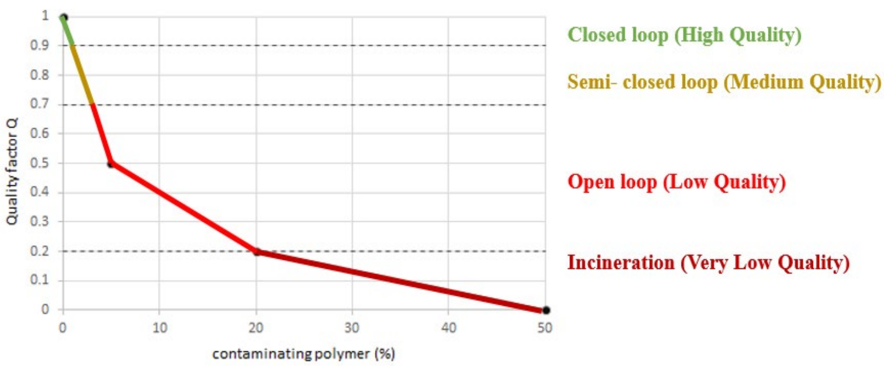

Figure 3: Q-values vs contaminating polymers for each compatibility class, adapted from Huysman et al. (2017).

4. Case study demonstration

A simple demonstration of the Q-values approach in evaluating the compatibility between waste sources is conducted based on the results of the Pinch Analysis from Chin et al. (2022). The authors have identified the minimum external plastics required for a case study considering three polymer types: PE, PET and PP, where each polymer type has disposed of plastics waste that is not usable for the demands. The disposed waste can be mixed, and their compatibility can be evaluated. Not only the disposed waste, the sources that are recycled can also be mixed to enhance the polymer grades if possible, and in turn reduce further the virgin plastic demands``. This work considers different scenarios of a plastic mix between polymer types. Table 3 below shows the extracted plastic source data used in this work.
The data from Table 3 indicates the results from Pinch Analysis, where about 500 t of PET is disposed of, and 100 t of PP is disposed of, while PE is fully reused. The disposed waste can be mixed to check for the compatibility class. However, since PET and PP have limited compatibility due to their binary surface tension being 9.001 – see Tables 2 and 3. Mixing them should not produce a mixture that can be fully recycled. Figure 4 shows the Q-values of the PET/PP mixture when the mixture is mixed with 50 % of SR3-PET and 50 % of SR3-PP. It can be clearly seen that the mixture falls into the 'incineration' class, which seems it is unwise to mix them. A similar case is observed with mixing SR1-PE even with grade 1. For a good quality PET/PE/PP mixture, one of PET or PE/PP should be as low as 10 % to be usable.

Table 3: Sources data with their polymeric compositions. Data retrieved from Chin et al. (2022). Binary mixture surface tension between two major polymers is considered only

	Targeted types	polymerGrades	Compositions (wt %)			Q-value
Sources			PET	PP	PE	
SR1-	PET	2	0.661	0.008	0.330	0.2-0.7 (Open loop)
SR2	PET	2	0.930	0.001	0.070	0.7-0.9 (Semi-closed)
SR3	PET (Disposed 500 t)	3	0.968	0.000	0.031	0.7-0.9 (Semi-closed)
SR1	PP	1	0.001	0.845	0.154	0.7-0.9 (Semi closed)
SR2	PP	1	0.001	0.925	0.074	0.9-1 (Closed)
SR3	PP (Disposed 100 t)	4	0.000	0.984	0.016	0.9-1 (Closed)
SR4	PP	1	0.001	0.914	0.085	0.9-1 (Closed)
SR1	PE	2	0.189	0.001	0.811	0.7 – 0.9 (Semi closed)
SR2	PE	1	0.046	0.000	0.954	0.9 – 1 (Closed)
SR3	PE	4	0.047	0.001	0.952	0.9 – 1 (Closed)
SR4	PE	2	0.437	0.087	0.476	0.2 – 0.7 (Open)

As for the option of mixing disposed PP with PE waste (SR1-PE), it can be seen that the mixture (with a 50:50 mix ratio of two streams) is still considered to be open-loop recycling waste even though they are reasonably compatible. In order to achieve a quality of waste to be at least usable for semi-closed loop recycling, the PP waste streams should be reduced. Based on Figure 5, the amount of SR3-PP should be reduced to 32.4 %.
Mixing different polymers could cause compatibility issues, but mixing polymers with the same types should not be a major issue. However, the mixture compatibility is still to be checked since they are contaminated with other polymers. Mixing SR3-PP with the high-quality SR2-PP seems to produce a compatible and high-quality waste, even with a 50:50 mix ratio between two streams- see Figure 5. In this case, the disposed waste can be reduced. However, this analysis only shows the compatibility evaluation, and the mixture properties are still to be experimentally validated, and the final properties of the plastic mixture should be checked prior to recycling.

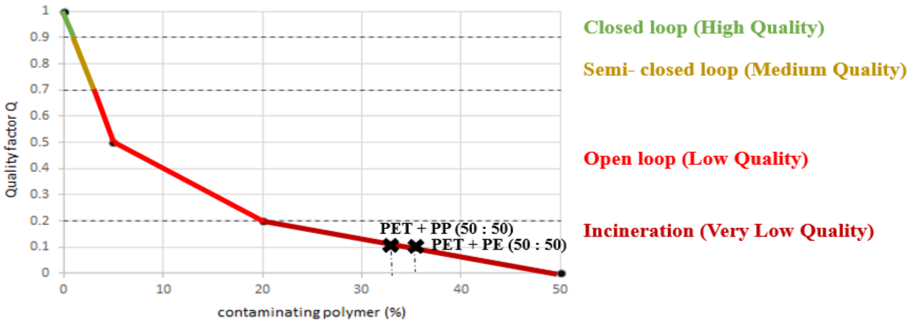

Figure 4: Q-values of mixing PET waste streams with PP or PE streams

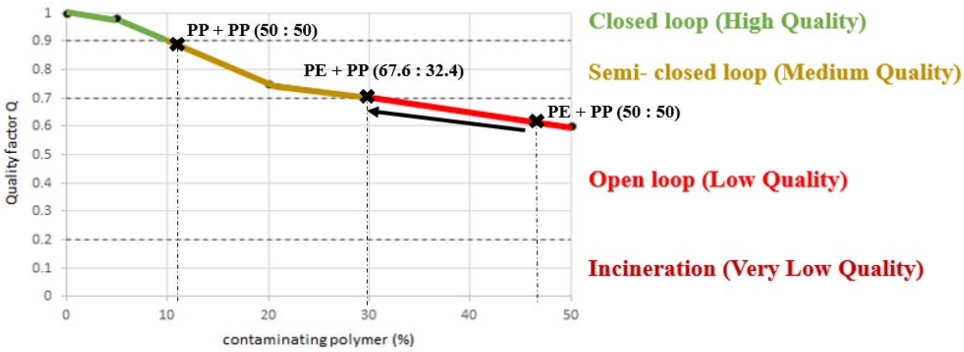

Figure 5: Q-values of mixing PP waste streams with PP or PE streams

5. Conclusions

This work has developed a simple evaluation framework to evaluate the compatibility of the polymers between two waste streams. The method is based on the Q-values as a function of contaminated polymers. To facilitate a systematic recycling planning, the data-driven Plastic Pinch Analysis can be integrated with the Q-values approaches to identify the quality predictors of the plastic waste streams, which in turn ideally minimise the virgin plastic demands for any recycling system. From the case study, it is shown that mixing PE with PP is plausible, but the mass share of the PP waste streams should be reduced to about 32.4 %. Mixing two limited compatible waste is possible but only with a minute amount. However, this framework is preliminary and requires more advanced improvements and experimental validations. Mixtures with multi-polymers with proper surface tension mixing rules should be investigated in the future. The Q-value approach mainly accounts for polymers compatibility. The final properties of the plastic waste should be evaluated prior to the recycling decision.

Acknowledgements

Gratefully acknowledged Support from the Grant Agency of the Czech Republic under Project No. 21-45726L in a bilateral collaborative project with the Ministry of Education, Science and Sport of the Republic of Slovenia (project No. 5442-1/2018/106), the Slovenian Research Agency (research core funding No. P2-0412 and P2-0421 and project No. J7-3149).

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