

Discrete Element Simulation and Economics of Mechanochemical Grinding of Plastic Waste at an Industrial Scale

Elisavet Anglou^a, Yuchen Chang^a, Arvind Ganesan^a, Sankar Nair^{a,b}, Carsten Sievers^{a,b}, Fani Boukouvala^a

^aDepartment of Chemical and Biomolecular Engineering, Georgia Institute of Technology, Atlanta, GA 30332 USA

^bRenewable Bioproducts Institute, Georgia Institute of Technology, Atlanta, Georgia 30332, United States

Abstract

Efficient and sustainable chemical recycling pathways for plastics are vital for addressing the negative environmental impacts associated with their end-of-life management. Mechanochemical depolymerization in ball mill reactors is a new promising route to achieve solid-state conversion of polymers to monomers, without the need for additional solvents. Physics-based models that accurately describe the reactor system are necessary for process design, scaling up, and reducing energy consumption. Motivated by this, a Discrete Element Method (DEM) model is developed to investigate the ball milling process at laboratory and industrial scales. The lab-scale model is calibrated and validated with data extracted from videos using computer vision tools. Finally, scaled-up ball mill designs capable of depolymerizing varying feeds of PET waste were simulated, and their capital and operating costs are estimated to assess the economic potential of this route.

Keywords: ball milling, plastic recycling, discrete-element-method, technoeconomics

1. Motivation and background

Traditional economies rely on linear production processes in which raw materials are manufactured into useful products and later discarded as waste. However, this economic model places a significant strain on natural resources and the environment, particularly in the case of single-use plastics. In 2018, 360 million tons of plastics were produced globally, with 80% ending in landfills or in the sea, whereas only 10% were recycled (Tricker et al., 2022). To alleviate the environmental stress on natural resource degradation, there is increasing interest in transitioning to a circular economic model in which waste materials, such as plastics, will be recycled back into the economy.

Plastic recycling methods can be categorized into mechanical and chemical pathways. Most recycling infrastructures currently focus on mechanical processes in which waste is physically reshaped into new products from a melt. However, mechanical and thermal stresses acting on the polymer melt during processing degrade the integrity of the plastic product; therefore, each plastic product can only be recycled a limited number of times, and only if mixed with large quantities of virgin polymers (Tricker et al., 2022). A promising alternative is the chemical recycling of polymers, which bypasses the limitation of material degradation entirely, by converting polymers directly to monomeric molecules through depolymerization. Common chemical recycling pathways such as pyrolysis and solvolysis involve high energy consumption or the use of solvents, therefore hindering their economic viability. The development of alternative depolymerization

processes for plastic waste is crucial for implementing sustainable practices and reducing negative environmental effects. In a previous study (Tricker et al., 2022), it was shown that mechanochemical depolymerization in ball mill reactors is a promising route for converting solid polymers into monomers without the need for additional solvents. Mechanochemical depolymerization pathways utilize the mechanical energy supplied by collisions between grinding bodies (balls and walls) of a ball mill to drive the reaction between solid reactant particles. Thus, accurately simulating the movement and interactions between these entities can provide crucial information for explaining mechanisms of mechanochemical depolymerization and for optimizing process design. Two modeling frameworks have predominantly been used to simulate ball mill systems: semi-empirical population balance and mechanistic Discrete Element Method (DEM) models. DEM models for ball milling have received increasing attention owing to their ability to describe the kinematics of moving entities and the corresponding energy involved in their collisions during ball milling. Additionally, ball milling operations at the industrial scale can be energy-intensive; hence, even a marginal improvement in efficiency can lead to a significant reduction in expenses. For instance, approximately 110 kWh of electric energy is consumed during the production of 1 ton of cement, of which 70% is required for comminution, whereas only 1-5% is explicitly used for particle breakage; the remaining energy is wasted (Boemer & Ponthot, 2017; Tavares, 2017).

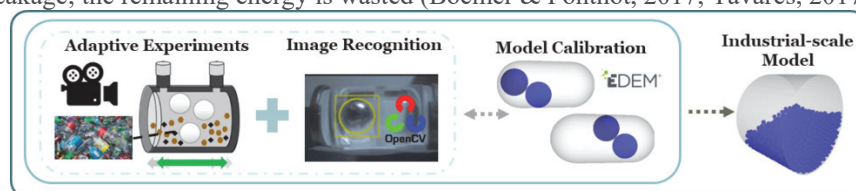


Figure 1: The main components of this study include: (a) experimental design and machine learning computer vision tools for the calibration of a lab-scale DEM model, and (b) DEM simulations of scaled-up ball mill configurations for various operating settings to estimate capital and operating costs

In this work, a DEM model was developed to investigate the depolymerization of poly(ethylene terephthalate) (PET) powder. First, high-speed video recordings of a lab-scale ball mill were obtained at operating conditions that allowed for quantitative PET depolymerization in corresponding experimental studies in the same mill (Tricker et al., 2022), and a computer vision algorithm was used to track the moving balls in the reactor. The extracted data were used to parameterize the DEM model and predict the kinematic interactions of moving entities in the mill. Once the mechanistic model parameters were optimized, the geometry of an industrial-scale ball mill reactor was created, and various operating settings were simulated. The resulting DEM simulation was used to investigate the tradeoffs between achieved PET yields and energy requirements at the industrial scale. Finally, capital, and operating costs of the process were calculated for various scenarios of waste feeds, using data obtained from US state levels of plastic waste collection.

2. Methods

2.1. High-fidelity model for the laboratory ball mill reactor

PET depolymerization experiments were performed in a Retch MM400 vibratory ball mill with stainless steel grinding bodies. PET in powder form was milled with a stoichiometric amount of sodium hydroxide (NaOH) to form ethylene glycol (EG) and disodium terephthalate salt (Na₂TPA). Details regarding experimental conditions and results have been reported in (Tricker et al., 2022). A transparent PMMA milling vessel

identical to that used in the depolymerization experiments was manufactured and used in motion-tracking experiments, but without reactants (PET + NaOH) present. The mill's operation at various conditions (ball numbers and sizes, milling frequencies) was filmed using a high-speed camera (2134 fps) to study the collision kinematics between balls and the vessel wall. A Python script based on the OpenCV computer vision library was used to analyze the video frames and track the position of the ball(s). The coordinates of the ball(s) in space and time were identified and used to calculate their velocity, which is defined as the change in position between two consecutive frames.

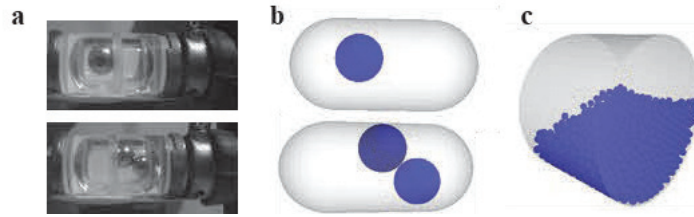


Figure 2: (a) Experimental setup including a vibratory 25mL reactor and stainless-steel balls (b) Replication of the experimental setup in DEM simulation with one and two balls (c) The geometry of the industrial-scale reactor as simulated in the DEM software

Next, the geometry of the laboratory setup was developed in Solidworks while the DEM model was developed in EDEM. The Hertz-Mindlin contact model was applied, as it is most appropriate for non-cohesive spherical shapes (Metta, Ierapetritou, & Ramachandran, 2018). Typical steel properties were used as inputs to the DEM model (density 7900 kg/m³, Poisson's ratio 0.3, coefficient of restitution 0.5, shear modulus 77 GPa). The coefficient of static and rolling friction are taken as 0.5 and 0.01 respectively based on (Metta et al., 2018). For PMMA-steel interactions, a grid search of the coefficient of restitution and the friction parameters was performed, and optimal values that match the velocity and the number of collisions between the simulation and the motion-tracking experiments were identified. The laboratory ball milling simulation was validated based on the recorded experiments under different operating conditions. Once the lab-scale simulation is validated by experiments, the identified parameters that govern ball-to-ball and ball-to-wall interactions can be used to simulate a scaled-up geometry.

2.2. Scaling up and technoeconomic analysis of ball mill reactor

A typical industrial ball milling setup comprises a rotating cylindrical vessel filled with balls and PET particles. Electric energy is used to satisfy the energy requirements necessary to drive the electric motor that rotates the mill and grinding media. To establish the operating costs, a new geometry and scale of DEM simulations was developed. A rotating cylindrical vessel was designed, and the simulation was developed (Figure 2c) using the validated material and contact parameters. The resulting torque requirements loads were extracted from the DEM simulation, and the operating costs were evaluated based on Equation (1) for 7920 hours of operation annually.

$$\text{Electricity Cost} = \text{Price} \times \text{Power} = \text{Price} \times (T_M \times \omega_M) \quad \text{Equation (1)}$$

$$C_{\text{Ball Mill}} = 64640 \times W^{0.64} \quad \text{Equation (2)}$$

$$Y_{\text{PET}}(\%) = 6.38 \text{ BPR} - 46.35 \text{ for } 10 \leq \text{BPR} \leq 23 \quad \text{Equation (3)}$$

Here, T_M is the torque associated with the rotation of the reactor vessel while ω_M is the mill's angular velocity, which in this work is equals to 15 rpm. The angular velocity was chosen such that it lies between 60-80% of the critical speed depending on the reactor

volume (Wang, Yang, & Yu, 2012). The fill level of the ball mill was kept constant at 30 %, and the radius of the balls was set to 80 mm for all cases. An electricity cost of \$0.07/kW-h was used while the purchase cost of the ball mill equipment was estimated based on the feed flowrate (W [ton/hr]) according to Equation (2) (Seider, Seader, Lewin, & Widagdo, 2004), and then adjusted to the 2022 dollar value via the CEPCI Index. According to our previous kinetics study on the PET depolymerization reaction (Tricker et al., 2022) for the lab-scale ball milling system, monomer yield follows a linear relationship (Equation (3)) with the ball-to-powder mass ratio (BPR) parameter, that is, the ratio of the ball mass to the mass of the reactants. Assuming that the same BPR values can be maintained at the industrial scale, the corresponding conversion, and costs can be calculated. Higher BPR ratios results in higher PET monomer yields, and full conversion is achieved when $BPR = 23$ with a 20 min reaction time (Tricker et al., 2022). The BPR parameter was used to scale-up the reactor and to evaluate the corresponding conversion, number of balls, and reactor volume for a certain PET waste feed.

3. Results and Discussion

3.1. Laboratory-scale model validation

The average ball velocity is compared in Figure 3a, which reveals that the values are in near-perfect agreement for the different operating conditions (vibration frequencies, diameters) tested and can thus validate the lab-scale model. Stainless-steel balls with diameters equal to 17.6 mm and 20.6 mm were used for the model validation while the milling frequency ranged from 22.5 to 30 Hz. Once the DEM parameters were estimated, the simulation can be executed for different conditions and reactor sizes, and the results can be used to further analyze the ball milling system.

3.2. Simulations at the industrial scale

DEM simulations were used to define the scaling-up procedure for PET waste depolymerization in ball mills and to evaluate the associated energy requirements. To illustrate the resulting capital and operating costs of plastic waste depolymerization at the industrial scale, two separate cases were considered.

3.2.1. Case-study 1: Influence of BPR to the operating costs

The BPR ratio significantly influenced the extent of PET depolymerization. To this end, the first case study depicts how the operating costs change with respect to the BPR value for the same feed of 1000 kg/hr PET waste (+400 kg/hr NaOH). The reactor volume and number of balls required to depolymerize the PET waste were calculated based on the BPR parameter.

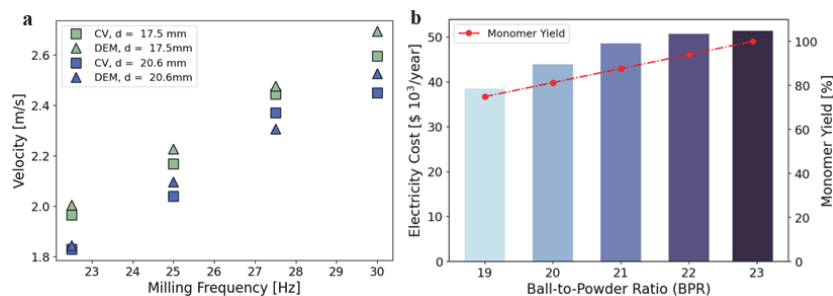


Figure 3: (a) Comparison of the average velocities of the stainless-steel ball at different vibration frequencies (22.5 – 30 Hz) extracted from the DEM simulations (triangles) and the recorded ball mill experiments (squares), (b) Comparison of annual electricity cost and achieved monomer yield for reactors processing 1000 kg/hr of PET (+400 kg/hr NaOH) at different BPR ratios.

The results of this analysis are shown in Figure 3b, which illustrates how the electricity cost changes with different BPR values. For higher BPRs, more balls are added inside the mill to grind the same amount of waste, resulting in increased power requirements for the mill rotation. More specifically, to achieve a 75% monomer (Na_2TPA) yield ($\text{BPR}=19$), \$1.62/tonne_{PET} are spent on electricity, whereas for full depolymerization ($\text{BPR}=22.9$) the cost is increased to \$2.16/tonne_{PET} for 20 min milling time (Tricker et al., 2022). The purchase cost of the ball mill vessel was estimated to be \$119,719 and was constant for all BPR combinations (assuming a negligible cost for the additional grinding balls). However, as PET conversion is reduced, subsequent unit operations will be necessary to separate and recycle unreacted PET waste which will increase capital and operating expenditures.

3.2.2. Case study 2: Influence of PET waste feedstock flowrate

The second case study illustrates the dependence of the total cost of ball milling to the mass flow rate of PET waste. Different PET waste recycling feed flow rates were evaluated, and the corresponding costs were estimated for a BPR value of 20 resulting in 81.2 % of PET converted to monomers. The PET waste for different US states were used as input to the DEM simulation based on recycling quantities for 2018 as described in the report of the National Association for PET Container Resources (*NAPCOR 2018*). The study region encompasses the South Atlantic region of the US, which in turn comprises nine states (Florida, Georgia, North Carolina, Virginia, Maryland, South Carolina, West Virginia, and District of Columbia). It is assumed that the PET recycling quantities are equally distributed to each state in the region based on the corresponding population for the same year. The resulting PET waste flowrates for each state are used as inputs to the DEM simulation using stoichiometric NaOH feed.

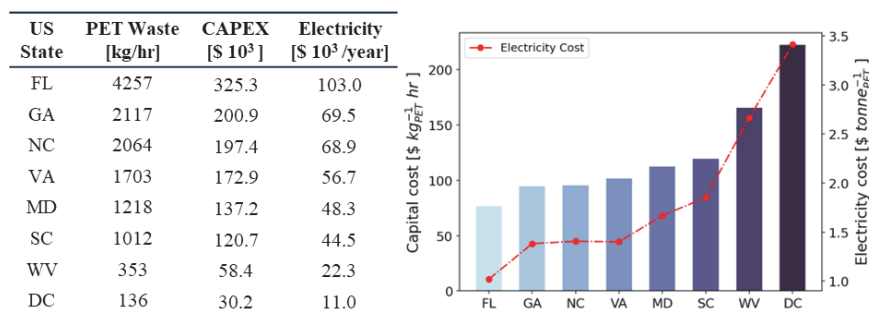


Figure 4: PET waste collected at each state in 2018 scaled based on population, and the resulting capital and operating costs. The graph illustrates the capital and operating costs per kg of PET waste fed in the ball mill reactor. The bars denote the capital cost per feed flow rate in \$ kg_{PET}⁻¹ hr, whereas the electricity cost per tonne is shown on the secondary axis for 20 min operation (red dotted line).

The results of this analysis are shown in Figure 4, which illustrates how the required expenditure per unit feed changes for different PET waste feeds. As expected, both capital and operating costs increased with feed flow rate, because maintaining the same BPR value requires larger numbers of balls and a larger reactor volume, which would require greater motor power to achieve depolymerization conditions for that entire mass of grinding bodies. In contrast, per unit costs decreases as the feed rate increases highlighting the benefits of economy of scale.

The economic potential of the mechanochemical route based solely on raw material (PET bale: \$150/tonne, NaOH: \$608/tonne, H₂SO₄: \$96/tonne) and product (vTPA: \$1143/tonne, EG: \$961/tonne) prices (Singh et al., 2021) was evaluated at \$978/tonne of

TPA, which is an indication of the upper bound of the potential profits. This value, in combination with the low electricity costs that were estimated in both case studies at \$2-3/tonne_{PET}, reveals that there is still a significant profit margin to account for mechanical pretreatment, required to grind PET waste to micro sizes, as well as downstream processes to recover the products of mechanochemical hydrolysis of PET waste that can be capital- or energy-intensive, i.e., rTPA crystallization, EG distillation, salt recovery (Singh et al., 2021).

4. Conclusions

In this work, we built a DEM simulation required to design and cost PET depolymerization via ball milling for a variety of feedstock flow rates and operating settings. Two case studies demonstrate the dependence of the operating and capital costs on the achieved PET conversion and feed flow rates. The results indicate that this can be viable process economically, especially for states that recycle more PET where there would be a benefit from economy of scale. Also, there are economic trade-offs to be considered in the future with respect to more costly depolymerization at high conversions, versus partial depolymerization and design of subsequent separation/purification steps and recycling streams. In future work, a more detailed technoeconomic analysis of plastic waste depolymerization via mechanochemical reactions will be performed to account for pre-treatment costs to shred plastic waste into powder form, as well as downstream treatment to purify the mechanocatalysis products. This analysis will be critical for comparing this technology with alternative recycling routes and aid policymakers and industry stakeholders in making informed decisions regarding its overall potential.

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