Simulation of Plastics Pyrolysis in Fluidized Bed with a Lumped Reaction Kinetic Model

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Introduction

The production of plastics is continuously increasing, and has reached 400 Mt in 2022 alone [1]. As a result, the generation of plastic waste has also increased dramatically. According to the OECD, approximately 353 Mt of plastic waste were generated in 2019, around 70% of it has been landfilled and incinerated, while another 20% has leaked into the environment due to mismanagement, leading to serious environmental issues such as microplastics [2]. While the conventional mechanical recycling is limited to thermoplastics with high purity, chemical recycling can be used to recycle mixed and contaminated plastics. Among the current chemical recycling technologies, pyrolysis shows the best balance between investment cost, operating cost and product quality compared to other technologies such as gasification or hydrothermal liquefaction [3,4]. During the pyrolysis process, plastic waste is decomposed in a high-temperature, oxygen-free environment into chemical building blocks, which can be reused to produce new plastic products. The feasibility of pyrolysis for recycling plastic waste has been proven extensively through laboratory-scale experiments over the past decades [3]. The development of industrial-scale, high-throughput (continuous) plastics pyrolysis processes is imperative to address the increasing production of waste plastics. However, the widespread industrial application of waste plastics pyrolysis is mainly limited by the technical feasibility of reactor scale-up and the high cost of large-scale experiments.

Among the various reactor types, fluidized bed reactors are considered highly promising for large-scale applications due to their superior heat and mass transfer capabilities, as well as excellent mixing characteristics. To develop a thorough understanding of the underlying processes while minimizing experimental costs, computational fluid dynamics (CFD) simulations can serve as a cost-effective tool for process optimization and risk reduction. The primary objective of this study is to develop a CFD model for simulating waste plastic pyrolysis in fluidized beds, capturing key phenomena such as dense gas-particle flow, heat transfer, and pyrolysis reactions. A lumped kinetic model is employed to balance computational efficiency with accuracy. The simulations are first applied to batch-operated fluidized beds and subsequently extended to continuously fed systems, providing valuable insights for future process development.

Simulation Setup

A cylindrical fluidized bed with a diameter of 5 cm and a length of 60 cm was used, along with 695 g of quartz sand as bed material and methane as fluidizing agent. The sand particles were assumed to be spherical and have a particle size distribution (PSD) with a mean diameter of 0.21 mm. Spherical, monodisperse polypropylene (PP) particles with diameters d_P ranging from 1.5 to 2.5 mm were fed into the reactor at an initial temperature of 25°C. The mass load of plastics compared with the bed inventory was set to 2%. The superficial gas velocity u_G and the reactor temperature T_R were set to $u_G = 15 - 30$ cm/s and $T_R = 470 - 530$ °C.

A Eulerian-Lagrangian approach was used to model the multiphase flow, where the collisions and heat exchange between the particles was considered (4-way coupling). A lumped 5-step kinetic model (adopted

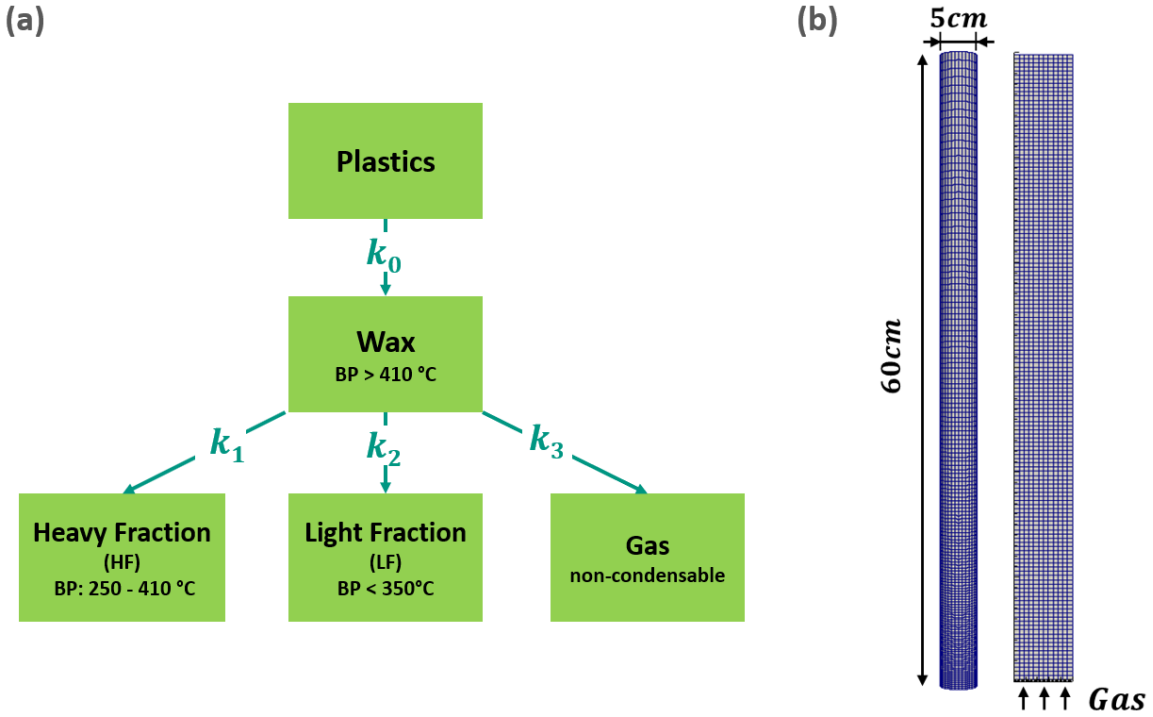

Figure 1: Illustration of the simulation setup: (a) 5-lump kinetic model (adopted from [5]) and (b) computational grid.

from [5]) was applied to characterize the pyrolysis reactions. As shown in Fig.1 (a), the degradation of polyolefins undergoes a formation of long-chained intermediates (Wax), which is followed by secondary decomposition into gaseous heavy fraction (HF), light fraction (LF) and gas (Gas). Since the boiling points of these final products are relatively lower than the reactor temperature, they are assumed to be immediately released to the gas phase after their formation. Additionally, due to the short residence time (2 - 4 s) of the gas phase, chemical reactions among the gaseous products were neglected.

Due to the long pyrolysis time (time to reach 99% conversion of plastic mass) of up to tens of minutes, the original cylindrical computational domain was simplified to a thin slice with a width of 5 cm, corresponding to the diameter of the cylindrical fluidized bed, as illustrated in Fig.1 (b). The thickness of the slice was set to 4 mm, which allows the use of particle sizes up to 2.5 mm for plastic particles. In this way, the volume of the domain was reduced to 1/10 of the original cylindrical domain of the full fluidized bed, leading to a speed-up of the computing time by a factor of 10. A modified OpenFOAM solver was used for the numerical simulations, where the heat transfer model [6] and lumped pyrolysis reaction model was implemented. The numerical setup was first validated against cold model experiments with regard to the hydrodynamics of the fluidized bed [7], which was then used for modeling plastics pyrolysis considering heat transfer and chemical reaction.

Results

Batch-wise feeding

The simulations were conducted first with batch-wise feeding of plastics. In this case, the simulation was started with an overheated fluidized bed reactor. After the formation of stable bubbling in the fluidized bed, plastic particles with 2% mass share of bed inventory were added to the reactor at once. The simulation was first conducted for T_R = 500 °C and d_p = 2.0 mm, which was treat as the reference case for further simulations. Figure 2 (a) illustrates contours of the instantaneous void fraction α_c, particle spatial distribution and temperature distribution in both gas phase and particle phase of the reference case. The bubbles can be detected by the red zones with $\alpha_c = 1$, corresponding to pure gas. The sand particles are indicated by the teal dots and plastic particles by the blue points. Due to the movement of the bubbles, the mixing of both particles was enhanced, leading to a relative uniform temperature distribution throughout the whole reactor.

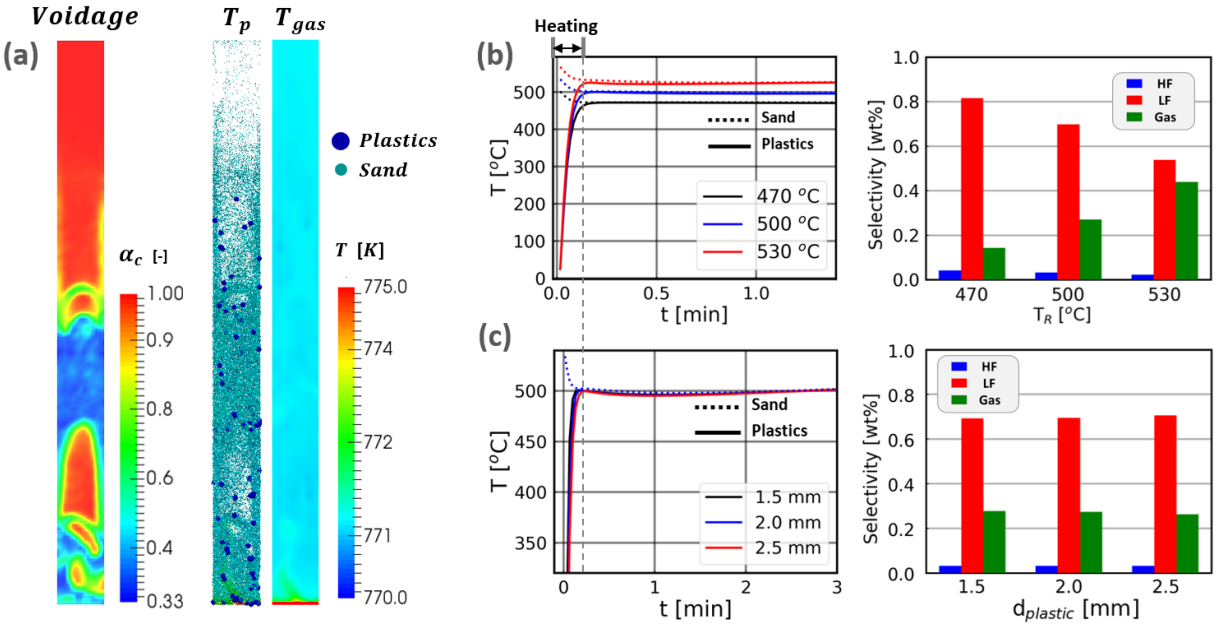

Figure 2: (a) Contours of instantaneous void fraction, particle distribution and gas phase temperature in the reference case (T_R = 500 °C, d_p = 2.0 mm); temporal evolution of temperature and product selectivity under batch-wise feeding for varying reactor temperature (b) and particle size (c).

As presented in Fig.2 (b), an increase in reactor temperature resulted in a decrease in LF's selectivity and an increase in Gas, which can be attributed to the difference in activation energy of the corresponding reactions ($E_{a,2}$ = 100 kJ/mol, $E_{a,3}$ = 249 kJ/mol). In contrast to the reactor temperature, the size of the plastic particles only have a minor influence on reaction selectivity, although smaller particles are heated up faster, as shown in Fig.2 (c). It has been found in the reference case that only approximately 1% of the particles were converted during the heating process (time to reach $T_{R,steady}$, approximately 10 s) from room temperature to reactor temperature. Therefore, despite different heating rates in the heating stage, all three sizes of particles demonstrate 1-2% discrepancy in selectivity. Moreover, the Euler-Lagrangian method treats all particles as having a homogeneous temperature, thereby further minimizing the impact of particle size. Although not shown here, the effect of plastic mass load and superficial velocity on the product selectivity were also negligible.

Continuous feeding

Under continuous feeding of plastics, the inlet gas temperature was kept constant during the simulation, which was higher than the desired reactor temperature T_R to provide the required heat for the endothermic pyrolysis process. The primary challenge in achieving continuous feeding is the establishment and maintenance of a stationary process condition at desired reactor temperature and plastic loading. In the initial stage of the simulation, the plastic particles will gradually accumulate in the reactor until the reaction rate reaches a dynamic equilibrium with the feeding rate. During this process, the boundary conditions (i.e., operating parameters) must be dynamically adjusted in order to stabilize the reactor temperature. In this study, the consumption rate of plastic under different operating conditions and the corresponding energy demand were estimated based on the given target operating conditions. These estimates were further employed in the calculations of mass and energy balance of the reactor system to determine the boundary conditions to achieve a steady state.

Figure 3 (a) demonstrates the time evolution the reactor temperature T_R starting from different initial states. With the aforementioned parameter setting strategy, the reactor can reach the steady state under all three conditions. During the startup stage, due to low mass share of plastics in the fluidized bed, the thermal energy supplied by the gas phase exceeded the energy demand, leading to energy accumulation and an increase of T_R. As the energy supplied by the hot gas and the consumption of heat for pyrolyzing

the plastics gradually reached equilibrium, the reactor temperature stabilized at a constant value. As shown in Fig.3 (b), a similar tendency in terms of product yield can be observed, where an increase of the reactor temperature led to the shift of selectivity from LF to Gas. Moreover, the size of the plastic particles d_P was found to have only a limited effect on the product selectivity during continuous feeding, although a slight decrease of T_R at smaller d_P was confirmed in the simulations.

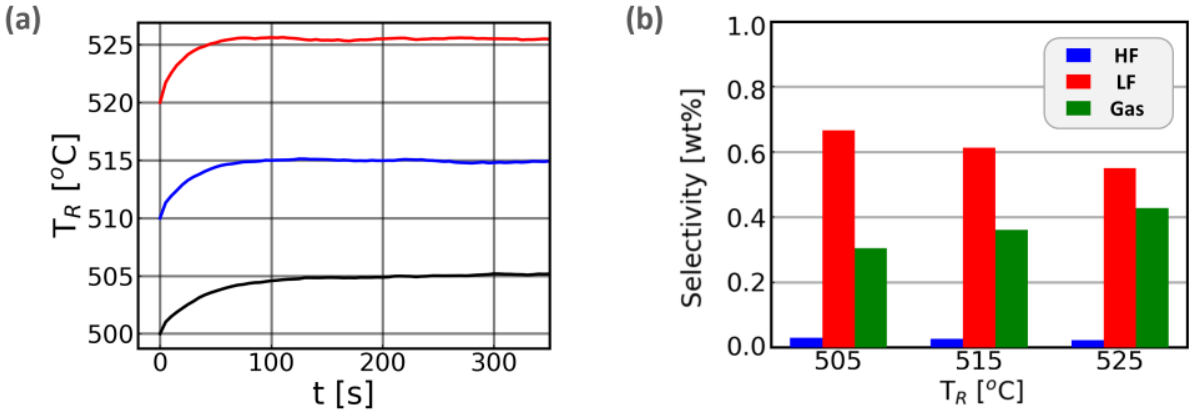

Figure 3: (a) Temporal evolution of reactor temperature under continuous feeding; (b) Selectivity of products at different reactor temperatures.

Conclusion

This study presents time-resolved Eulerian-Lagrangian simulations of plastic pyrolysis in a laboratory-scale fluidized bed, accounting for particle-gas dynamics, heat transfer, chemical reactions, and their mutual interactions. Simulations were conducted for both batch and continuous feeding operations. The promising simulation results across varying operating conditions demonstrate the potential of CFD-driven design to advance plastic pyrolysis technology on industrial scales. In future work, this modeling approach will be applied to guide the design and scale-up of plastic pyrolysis processes.

Acknowledgments

The authors gratefully acknowledge the financial support by the Helmholtz Association of German Research Centers (HGF), within the research program Materials and Technologies for the Energy Transition (MTET), topic Resource and Energy Efficiency. This work utilizes computing resources from the supercomputer HoreKa at the Scientific Centre for Computing (SCC) at KIT, Germany.

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