The counter-rotating twin screw extruder as a polymerization reactor
Ganzeveld, Klaassien

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
1992

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

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Summary.

Extruders are used in various parts of the polymer and food industry. In the food industry, they are applied in the production of ready to eat cereals, snacks, pet foods, pastas and confectionery products. The polymer industry uses extruders preferably to melt, mix, mould and compound polymers. However, over the last years, the use of extruders for polymerization reactions or for the modification of polymers increases considerably in industry.

Various types of extruders exist, each type of extruder having its specific properties and application areas. Nevertheless, generally all extruders are based on the same principle. They all consist of one or more screws, surrounded by a barrel which can be heated. Material is fed to the extruder by a hopper or a pump. In the extruder, the material is transported by the movement of the screws towards the die where it leaves the extruder. During the transport various actions can be performed in the extruder, such as, the melting of solid materials, the mixing of components or the performance of different polymerization reactions.

The interest in the use of the extruder as a new type of bulk polymerization reactor has increased lately due to the specific properties of the extruder. The main advantages are:
- No or only a small amount of solvent has to be used in the process. The polymerization process becomes much more energy efficient and environment friendly.
- The process is continuous.
- The extruder is a stable pump for highly viscous media which guarantees a constant throughput, during the polymerization process.
- The extruder has a relatively large heat exchanging surface and the thermal homogenization of the product is reasonable.

However, there are also some restrictions to the use of extruders as reactors. The need for fast reaction kinetics, a controllable temperature rise, caused by the heat of reaction, and a high viscosity
of the reaction product limits the number of reactions which can be performed in the extruder.

Basically two important extruder classes can be distinguished which can be used as reactors: the single screw and the twin screw extruders. The last category can be subdivided into three main groups, the non-intermeshing twin screw extruders, the closely-intermeshing twin screw extruders and the self wiping co-rotating twin screw extruders.

However, not all these types of extruders are equally fit for reactive extrusion, as the use of an extruder as reactor places special constraints on its design. The most important constraint is that the functioning of the extruder has to be insensitive to the rheological changes, that occur during the reaction. Based upon this condition, closely intermeshing twin screw extruders seem to be the most likely candidate for reactive extrusion. Therefore, in this thesis the use of a counter-rotating closely intermeshing twin screw extruder as a reactor was investigated.

The counter-rotating twin screw extruder consists of two screws which each rotate in a different direction, as the name already suggested. Due to the close intermesh of the screws separate spaces, the so called C-shaped chambers, exist on each screw. In these C-shaped chambers material is transported from the hopper to the die. However, due to mechanical clearances between the screws, and the screws and the barrel, leakage gaps between the chambers exist and material is able to flow backwards in the extruder. This flow of material is called the leakage flow and causes an interaction between the chambers (chapter 2).

The goal of the research was to examine the possibilities of this type of extruder as a polymerization reactor, and to develop models of the extruder reactor which accurately describe the reaction progress in the extruder.

The reactions which are performed in this extruder can, from a technological point of view, be divided into two categories: namely the single component and the multi component reactions (chapter 1).
The single component reactions occur throughout the bulk of the material. For this group of reactions macro mixing over the length of the extruder and the temperature of the reacting mixture play an important role. Both parameters determine the progress of the reaction in the extruder.

Multi component reactions, on the other hand, are also strongly influenced by micro mixing as the reaction proceeds at the interface between the components. Normally this kind of reactions becomes diffusion limited as the reaction progresses, due to the build-up of reaction product. To overcome the diffusion limitation extensive micro mixing is necessary.

By performing both types of reactions in the extruder, the functioning of the extruder as a reactor could be studied.

The single component reaction, which has been performed, is the bulk polymerization of butylmethacrylate (chapter 5). To study the multi component reactions two reactions have been examined: the polymerization of urethanes and the grafting of maleic anhydride on high density polyethylene (chapters 6 and 7). In both situations the extruder reactor operated stable without the occurrence of extensive temperature rises.

However, to understand the reaction progress of both types of reactions in the extruder, theoretical and numerical modeling is necessary.

For theoretical modeling a reactive extrusion interaction diagram is a very useful tool to analyse the extrusion process. The diagram clarifies the interactions between the extruder and reaction parameters, resulting in a clear overall picture of the process. Therefore, reactive extrusion diagrams are able to elicit the general tendencies of the reaction process and to analyse the extruder reactor performance.

With accurate numerical models, predictions can be made about the extruder behaviour, scale-up effects of the process can be studied elaborately and expensive experiments can partly be omitted.

The numerical model, used for the single component reaction, approximates the C-shaped chambers of the counter-rotating twin screw extruder as continuous stirred tank reactors on a conveyor band, interacting with each other through the leakage gaps. The model is based
upon the observation of the changes in a single chamber, moving through the extruder. To calculate the reaction progress in the chamber also equations for the fully filled length, the rheology of the reacting material, the heat transfer and the reaction kinetics have to be known (chapter 3).

The model is compared with experimental results to check its validity (5.5). From the results it can be concluded that, the model describes reality well, especially at high rotation rates. At low rotation rates of the screws, a difference between the model and the experimental results is visible, probably due to the formation of a polymer barrier at the barrel wall. At the moment, this effect cannot be incorporated in the program, as the physical background of the phenomenon is not completely clarified. Nevertheless, the program is very useful for large scale processes as industrial extruders operate mainly in the area of high rotational rates of the screws. In this area our model agrees well with the experimental findings.

To model the multi component reactions, the mixing mechanism of the extruder has to be understood. In a counter-rotating twin screw extruder micro mixing occurs mainly in the leakage gaps. The extent of the micro mixing achieved, is dependent on the number of gap passages, the striation thickness achieved and the diffusion coefficient of the components. The micro mixing is also influenced by the viscosity and quantity ratio of the components.

A study of the last two effects leads to the conclusion that, for mixing material introduced through a side feed, fast mixing is achieved in the extruder if the side feed consists of low viscous material and is present in considerable amounts.

The effect of diffusion is limited for the mixing of highly viscous materials, as present in the extruder. However, it can influence the mixing time if the striation thickness achieved in the gaps is in the same order of magnitude as the penetration depth.

From the point of view of the mixing parameters in the extruder:

- The mixing of a fluid element not leave the chamber.
- The mixing element returns to the zone of mixing, which also leaves the extruder.

Both the mixing element and the reactor describe the same physical phenomenon. Nevertheless, the program is very useful for large scale processes as industrial extruders operate mainly in the area of high rotational rates of the screws. In this area our model agrees well with the experimental findings.

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To verify the model, a comparison with experimental results is done to check its validity. From the results it can be concluded that, the model describes reality well, especially at high rotation rates. At low rotation rates of the screws, a difference between the model and the experimental results is visible, probably due to the formation of a polymer barrier at the barrel wall. At the moment, this effect cannot be incorporated in the program, as the physical background of the phenomenon is not completely clarified. Nevertheless, the program is very useful for large scale processes as industrial extruders operate mainly in the area of high rotational rates of the screws. In this area our model agrees well with the experimental findings.

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From the principle of distributive mixing stated above, two new relevant mixing parameters are developed for the counter-rotating twin screw extruder:

- The mixing efficiency being the average number of gap passages of a fluid element in the reaction zone.
- The mixing deficiency which equals the fraction of material that does not leave the C-shaped chamber in the reaction zone.

Both the mixing efficiency and deficiency are influenced by the screw speed, the throughput of the extruder and the die resistance.

To verify the mixing model, the experiments performed with the two multi component reactions were compared to the results of the model. The model describes the progress of both reactions well. From the experiments it could also be concluded that, besides the micro mixing, the mean residence time is a second crucial parameter in the performance of multi component reactions.

To transfer the knowledge obtained on small scale laboratory equipment to large scale production units scale-up rules are necessary. Therefore, in this thesis scale-up rules are proposed for counter-rotating twin screw extruders which are extended to the use of this type of extruder as a chemical reactor (chapter 8).

From the scale rules it can be concluded that, the fully filled length and the leakage flows are important factors when scaling up the extruder.

If the extruder is used as a chemical reactor, the Damköhler IV number plays an important role. For small Damköhler IV numbers no consistent scale rules can be found. However, when the heat of reaction is large compared to the heat transferred through the wall (Da IV >> 1), the throughput can be proportional to the diameter cube. This indicates that scaling up the process is economically feasible.

To indicate whether or not large thermal inhomogeneities can be expected a special dimensionless number can be defined. This is often important for process development, when a prospected process is scaled down to lab experiments.
Resuming the thesis, it can be concluded that:

- Many reactions can be performed in the counter-rotating closely intermeshing twin screw extruder.
- The reaction models describe the progress of the different types of reactions in the extruder well.
- The use of the extruder reactor in production processes is feasible.