Extrusion Processing of Ultra-High Molecular Weight Polyethylene

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Abstract

Ultra-high molecular weight polyethylene (UHMWPE) is a unique thermoplastic polymer with excellent performances. It has ultra-high molecular weight and extreme rheological behaviour, which make it a worldwide challenge to process UHMWPE continuously with little or without processing aids. Although the polymer processing technology has been increasingly maturated, it still cannot carry out the industrialized production efficiency by conventional processing methods and apparatus at present. In this chapter, we review the progress of extrusion processing technology for UHMWPE, including ram extrusion, single screw extrusion, twin screw extrusion and novel extrusion technology based on extensional rheology. By summarizing of these processing technologies, a basic framework of the processing principles and methods for UHMWPE is clearly presented. It is helpful for us to understand the processing characteristics and methods for such thermoplastic polymer with ultra-high molecular weight.

Keywords: ultra-high molecular weight polyethylene, extrusion processing, melting mechanism

1. Introduction

Ultra-high molecular weight polyethylene (UHMWPE) is a unique thermoplastic polymer possessing outstanding physical and mechanical performances such as good wear and corrosion resistance, low coefficient of friction, high impact strength at cryogenic temperatures, good environmental stress-cracking resistance, non-toxic and acceptable biocompatibility [1, 2]. Because of these excellent properties, UHMWPE has been widely used in many applications including mining, transportation, military industries, biomedical engineering, sports and livelihood projects [3–6].

UHMWPE is a special kind polyethylene. Except it, the polyethylene family also includes linear low density polyethylene (LLDPE), low density polyethylene (LDPE), high density polyethylene (HDPE) and cross-linked polyethylene (XLPE) [7–9]. These polyethylenes are synthesized with ethylene as monomers. By using different catalysts and synthetic process conditions, a variety of polyethylene with different structures, density and properties can be synthesized, such as LLDPE, LDPE and HDPE. In addition, cross-linked polyethylene can be obtained by the crosslinking reaction during or after the synthesis reaction process. UHMWPE is an unbranched linear polyethylene and can be synthesized with Ziegler-Natta or metallocene catalyst under the low-pressure polymerization process conditions [10, 11]. UHMWPE has extremely high molecular weight of up to several millions g/mol, which is several times larger than HDPE with a molecular weight of 200,000 g/mol. Actually, the precise molecular weight of UHMWPE is too high to be measured directly by conventional means, and it must be inferred by its intrinsic viscosity as an alternative approach.

In general, molecular weight of polymer has significant effects on its condensed structure, chemical performance, mechanical properties and processability. It is obvious that the excellent characteristics of UHMWPE are largely benefited from its extremely high molecular weight. The acceptable reasons are related to the enhanced intermolecular interactions and intensive chain entanglements. With increasing molecular weight, van der Waals force between macromolecules would be strengthened, and the ultra-long molecule chains of UHMWPE are prone to become entangled and to form intensive physical entanglements [12, 13]. These factors synthetically result in the significant improvement of mechanical strength, abrasion resistance, chemical stability, and so on.

Even if the ultra-high molecular weight has brought superior performances for UHMWPE, the extremely high molecular weight also brings great challenges for its processing and forming such as poor dissolution, extremely high melt viscosity and poor melt flowability [14–16]. The long chain movement of UHMWPE can be restricted by the strong constraint effect due to the enhanced intermolecular interactions and dense physical entanglements, resulting in improving the solvent resistance and solution or melt viscosity. This result is not a good thing for processing and forming of UHMWPE, including solution method and melting method.

For most thermoplastic polymers, there are a variety of processing and moulding methods, such as extrusion, injection, compression and casting. By these processing technologies, raw materials can be fabricated into polymer products with given profiles and appropriate properties such as large products with sufficient strength or micro/nano parts with high precision. However, it is very difficult to process nascent UHMWPE via conventional batch processing methods. UHMWPE extrusion products (pipes, sheets and bars) only can be extruded by large trust extruder with special screw structures, simultaneously adding a large amount of organic compounds as lubricants. According to the literature, even with improved upgrade screw extruder, it is almost impossible to extrude UHMWPE without any processing aids. The most straightforward reason is the extremely high melt viscosity and low melt flowability [16–19]. It is also very difficult to carry out injection moulding due to the poor fluidity of UHMWPE. Even though Huang et al. [20, 21] have obtained injection parts of UHMWPE blend containing 90 wt% commercial UHMWPE and 10 wt% ultra-low molecular weight polyethylene(ULMWPE), with a modified injection moulding technique named as oscillation shear injection moulding, pure UHMWPE parts with profiled surfaces still cannot manufactured directly by injection moulding. Thus, multistep processing method is usually used to prepare UHMWPE parts with complex profiles. For instance, in order to manufacture artificial knee joints, nascent UHMWPE powder particles must first be moulded into primary products with square or cylindrical profiles by compression moulding or plunger extrusion, and then artificial knee joints were fabricated by turning from primary products [1].

As a matter of fact, the problem of UHMWPE processing is always a worldwide challenge for material engineers and researchers from past to present. The efficient and easier processing solutions for UHMWPE could not be put forward over the past few decades. It is still a main constraint for promoting the development and application of UHMWPE at present. Actually, UHMWPE extrusion processing technique is the most likely to first achieve industrial production. The objective of this chapter is to review the progress of extrusion processing technique for UHMWPE, as a basis for understanding the processing principles and methods for such thermoplastic polymer with ultra-high molecular weight.

2. Flow characteristics of UHMWPE

2.1. Structure and thermal properties

As previously described, UHMWPE is synthesized with ethylene as monomers and has same repeat units as other common polyethylene. The single molecular chains of UHMWPE can consist of as many as hundreds of thousands repeat units. Because of the internal energy, the molecular chain could become mobile at elevated temperatures. Considering the ultra-long length and movement, a single molecular chain of UHMWPE is like a moving string over a kilometre long and tends to be tangled gradually.

As the temperature decreases, the molecular chain has a trend to rotate around the C-C bonds and create chain folds, reaching a new energy equilibrium state at a lower temperature. When cooled below the melt temperature of polymer, the activity of molecular chain is reduced, and the folded chains begin to form crystalline lamellae, which is the local ordered sheet-like regions. These lamellae gradually grow and accumulate in order to form crystalline regions of polymers. It is well known that there is almost no complete crystallization for polymer. In other words, there are always disordered regions in polymer, namely amorphous regions. Although the molecular chain of polyethylene is regular and flexible, not all segments of polyethylene chain can arrange into the ordered regions to form lamellae and crystalline regions. The lamellae are embedded within amorphous regions and may communicate with surrounding lamellae by tie molecules. Kurtz [1] has clearly observed the crystalline morphology of UHMWPE by using transmission electron microscopy (TEM), showing the composite nature of UHMWPE as an interconnected network of amorphous and crystalline regions. It can be clearly seen that UHMWPE is a typical semi-crystalline polymer as ordinary polyethylene.

Despite the close crystallinity, the morphology of nascent UHMWPE powder particles is quite different from that of HDPE. Obviously, from scanning electron microscope (SEM) images as shown in **Figure 1**, the individual nascent powder particle of UHMWPE is composed of several secondary particles. There are many orientation fibres as connections between secondary particles. Compared with other dense parts in the individual particle, the orientation fibres are loose. These fibres as weak sections in powder particles may be first melted when the temperature rises to the melting point.

The values of crystallinity degree and melting point of nascent UHMWPE with different molecular weight are listed as **Table 1**. Apparently, the crystallinity degree and melting point have a rising trend with increasing molecular weight. However, the values of crystallinity degree and melting point from the second scanning curves are all lower than that from the first scanning. The possible reason is that the crystallinity degree of polymer depends upon various factors, including molecular weight, processing conditions and environmental conditions (such as loading, flow field, ultrasonic, and so on) [22–25]. It is helpful to understand the melting mechanisms and melt flow process by realizing the melting process and crystallization process.

Thermal stability of polymer is another important factor, which must be considered in processing. In general, the decomposition temperature of HDPE or LDPE is below 400°C. But the decomposition temperature of UHMWPE usually exceeds 430°C probably because of the strong intermolecular interactions from the extremely high molecular weight. As the heating scanning curve of UHMWPE (GUR4120, Ticona) exhibited in **Figure 2**, the initial decomposition temperature is up to 450°C and the fastest degradation process occurred at about 480°C. Although it has higher decomposition temperature than other common polyethylene, the processing temperature for UHMWPE cannot be such high. Due to poor thermal conductivity, the accumulation of heat causes too high temperature in partial of polymer melt, and the thermal degradation in the melt state occurs in the processing temperature up to 300°C.

2.2. Flow characteristics

It is important to note that the melt flow characteristics of polymer usually determine the processing method and conditions. It also greatly affects the forming process and product quality. For instance, the phenomenon of melt breakup in extrusion is usually related to the critical shear rate, which is an important aspect of flow characteristics of polymer melt.

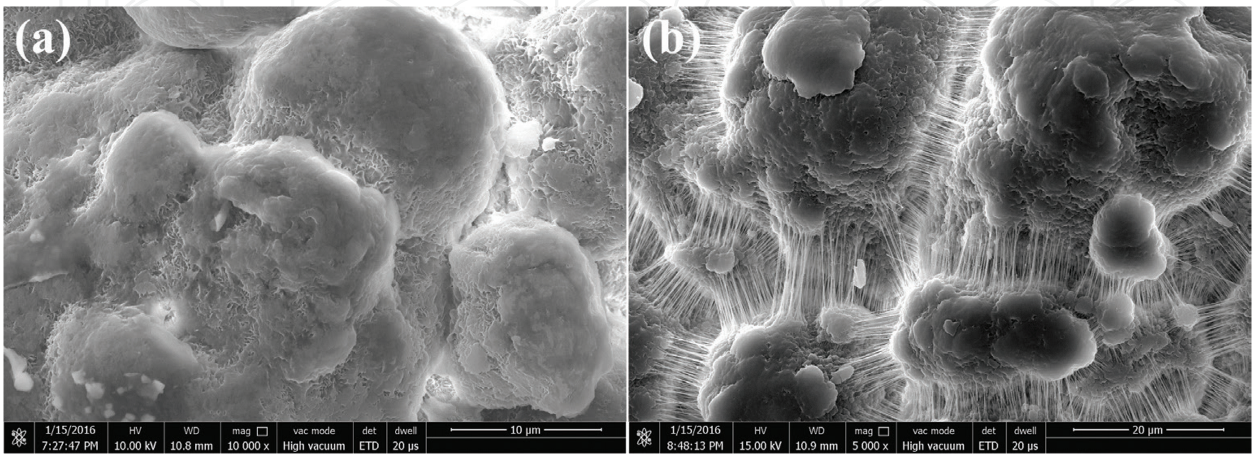

Figure 1. The morphologies of polyethylene with different molecular weight (a) HDPE with molecular weight of 600,000 g/mol, 10,000× magnification; (b) UHMWPE with molecular weight of 2,000,000 g/mol, 5000× magnification.

Samples	M_w (1×10^4 g/mol)	T_{m1} (°C)	T_{m2} (°C)	X_{c1} (%)	X_{c2} (%)
GHR8110	~60	130.2	128.5	71.1	68.3
145 M	~145	140.7	135.8	65.8	54.8
M1	150–200	143.6	136.4	69.3	54.0
M2	250–350	144.2	137.1	83.4	65.2
M3	~350	144.5	137.0	81.4	61.9
GUR4120	~500	144.5	136.4	78.5	58.9

Table 1. Melting point and crystallinity of various UHMWPE with different molecular weight from DSC curves (1—the first scanning, 2—the second scanning).

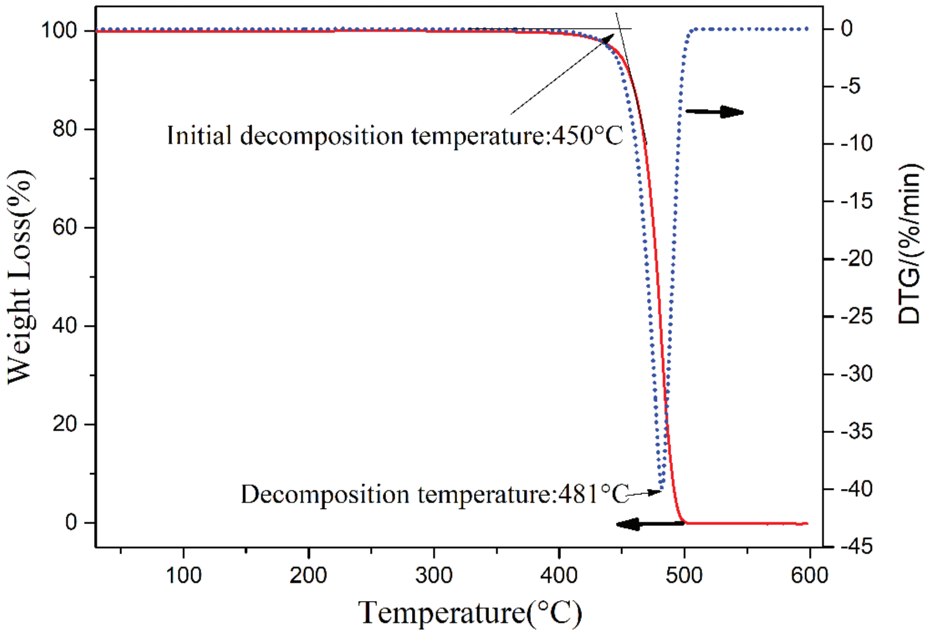

Figure 2. Thermogravimetric analysis (TGA) for UHMWPE.

For UHMWPE, its melting point is about 155°C, and the polymer powder particles can completely fuse into melt as temperature is higher than 200°C. However, the UHMWPE melt is rubberlike and has poor flowability even at the temperature much higher than melting point. Even if the temperature and load, respectively, were up to 250°C and 21.6 kg separately, the melt flow rate of UHMWPE was almost zero. Melt viscosity is an important parameter of melt flow. Actually, it is very difficult to determine directly the precise value for the melt viscosity of UHMWPE. But the trend in change of viscosity can be inferred with molecular weight by the following equations [16].

$$\eta = kM_w (M_w < M_{wc}) \quad (1)$$

$$\eta = kM_w^{3.4} (Mw > M_{wc}) \quad (2)$$

where k is a constant and η, M_w, and M_{wc} represent viscosity, average molecular weight, and critical relative average molecular weight, respectively.

Generally speaking, the molecular weight of UHMWPE should be higher than 1.5 million g/mol. Thus, the molecular weight of UHMWPE is much higher than the critical relative average molecular weight. Therefore, the viscosity can be evaluated by Eq. 2. For instance, the viscosity of UHMWPE with molecular weight of 3 million g/mol is about 2500 times higher than HDPE with molecular weight of 300,000 g/mol. According to the references, the melt viscosity of UHMWPE could be up to 10^8 Pa·s [26].

The extremely high viscosity would certainly result in the poor melt flowability and processability. For example, even if the nascent UHMWPE powder particles were processed with an internal mixer at 200°C for 20 min, the masterbatch still could not be dispersed evenly in UHMWPE melt. As displayed in **Figure 3a**, during the whole mixing process, the masterbatch only dispersed in a very narrow range along the rotation direction; however, it almost cannot disperse into the melt far away from the place where masterbatch is placed. Probably because the strong shearing effect along rotation direction makes materials exchanging significantly, and there is no obvious material exchange in the direction perpendicular to the rotation. This clearly demonstrates the limited molecular chain mobility and poor melt flowability of UHMWPE melt.

With the increase of temperature, UHMWPE melt will not enter the viscous flow state but to maintain a transparent rubberlike state. In fact, UHMWPE melt has no viscous flow state like HDPE or LDPE because its theoretical viscous flow temperature is higher than the decomposition temperature. In addition, it is easy to find out from the picture that the UHMWPE melt has ruptured and could not form uniform and continuous melt. The fundamental reason is that UHMWPE melt has a low critical shear rate of 10^{-3} s^{-1}. Therefore, the UHMWPE melt was easy to break up by strong shear effect with internal mixer. Although there exists a metastability processing window in the temperature range of 154°C and 157°C [27–29], the nascent UHMWPE cannot be processed by conventional or improved upgrade screw equipment without processing aids. Considered the complicated interfering factors of extremely high melt viscosity, low melt flow rate, low critical shear rate no viscous flow and so on, it is a huge challenge to process UHMWPE continuously and efficiently via screw equipment dominated by shear flow and methods based on viscous flow theory.

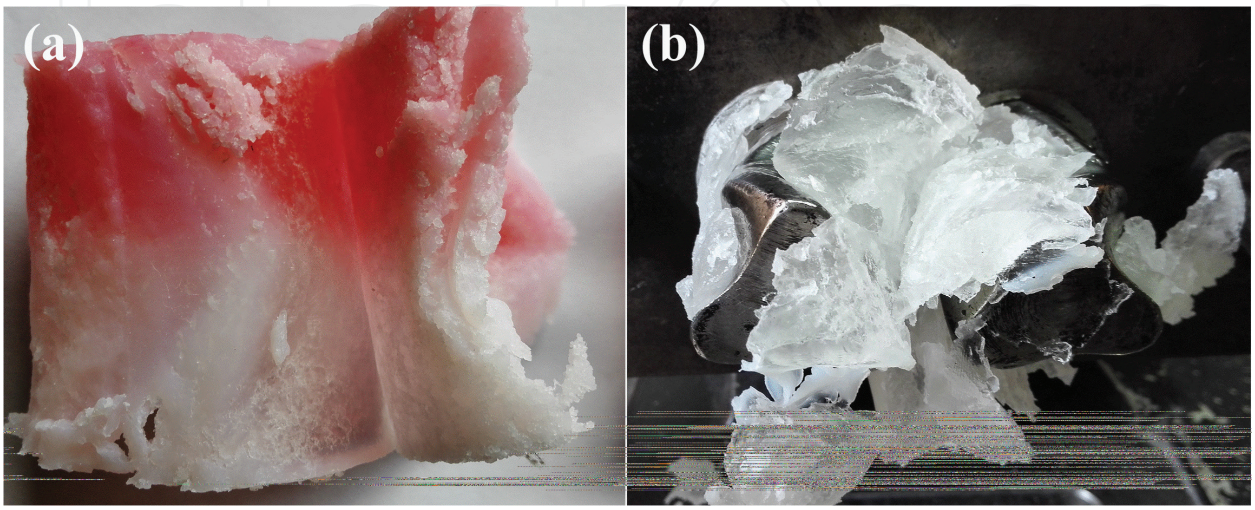

Figure 3. Melting state of UHMWPE processed with an internal mixer.

3. Extrusion processing and forming of UHMWPE

3.1. Ram extrusion of UHMWPE

The most common methods used to process UHMWPE powder particles into bulk products are compression moulding and ram extrusion [30, 31]. Compression moulding originated in Germany in the 1950s is a discontinuous process based on sintering without limitation of molecular weight and melt viscosity. Ram extrusion was developed by converters in the United States during the 1970s. It was a processing process, which can be considered as a continuous compression and sintering process. Taking into account the intermittent feeding and compaction, ram extrusion in the strict sense was a quasi-continuous process. Nevertheless, it still could consecutively produce sheets, pipes, bars and profiles, even the molecular weight of UHMWPE being up to 10 million g/mol. In contrast with single screw or twin screw extrusion, the ram extrusion only was affected slightly by molecular weight and melt viscosity.

Ram extruder consists essentially of a hopper, a feeding chamber with heating device, a horizontal reciprocating ram, a heated forming die, cooling and shaping apparatus. So it is easy to understand the ram extrusion process of feeding, compaction, melting and plasticizing, extruding, cooling, and moulding. UHMWPE powder particles are fed continuously into ram extruder and were heated at elevated temperatures. Within the extruder, the powder particles are consolidated and maintained under pressure by the ram, as well as by the back pressure from UHMWPE melt, which caused by frictional forces of the molten resin against the die wall surface. UHMWPE melt is extruded from heated forming die with specified shape. The thrust force to overcome the huge resistance originated from positive displacement movement of the reciprocating ram.

Although the ram extruder can manufacture UHMWPE products with good surface quality, the intermittent stamping process caused residual stresses inside the bulk. It is necessary for extrusion products to be annealed at elevated temperatures in order to remove residual stresses. The annealing process can also increase the crystallinity of the components, which is helpful to maintain the excellent mechanical performances of UHMWPE.

There are also some disadvantages for ram extruder to process UHWMPE, such as fluctuations in product quality, longer melt plasticization cycle, slow extrusion rate, and high energy consumption. Extruder with multiple plungers is used to reduce the pulsation frequency [32]. Several rams alternately compact the materials and push them forward. This process shortens the operation time between two compaction actions, making the whole process closer to a continuous process, which is conducive to reducing the fluctuations of processing process and product quality. Certainly, this requires more advanced control technology for the extrusion process.

3.2. Screw extrusion of UHMWPE

In fact, regardless of high energy consumption and low processing efficiency, compression and ram extrusion are the most suitable processing methods for thermoplastic polymer with extreme high molecular weight and melt viscosity. However, the cost and diversity of products must be considered in industrial production. Screw extrusion is quite popular with people in all conventional processing methods for UHWMPE because of continuous production process and well-compounded effect. The commonly used screw equipment for UHMWPE are single screw extruder and twin screw extruder.

3.2.1. Single screw extrusion

In 1939, Troester Machinery Company in Germany launched an extruder with length/diameter ratio of 10, marking the development and rise of modern single screw extruder. There are many types of single screw extruders, which are widely used in extrusion processing and moulding of polymers [33–35].

Despite various kinds of single screw extruders, they have similar functions for common thermoplastic polymers. After entering the barrel from the hopper, the material was gradually pushed to the head direction with rotation of the screw. Successively, the material passed through several functional areas of the extruder, including solid conveying section, melting section, and melt conveying section. Loose materials were compacted in solid conveying section and melted before reaching the melt conveying section, then the homogenized melt was squeezed out from heating die. For most thermoplastic polymers, they eventually became viscous fluid in the extrusion process. Various processing rheology theories and extrusion equipment are based on this fact.

However, many cases are very different for UHMWPE. Since the low friction coefficient of UHMWPE and metal, UHMWPE powder particles in the feeding section are easy to slide with the rotating screw, resulting in the difficulty for powder particles being pushed forward. On the other hand, the melt is like rubber without viscous flow, which means a poor flowability for UHMWPE melt. The extremely high resistance is easily established in compaction section and results in huge backpressure due to the extremely high melt viscosity and poor flowability. Then, the melted resins are easy to wrap in the screw and rotate with the screw, preventing UHMWPE melt to move forward. It can even cause the screw to break if the device is forced to run.

Obviously, conventional single screw extruder is almost powerless to process UHMWPE. Then, many new dedicated equipment with special internal structures were developed in order to overcome the difficulty of feeding and huge extrusion resistance [36]. For example, special single screw extruder with gradient grooves in the barrel was developed in 1971 to avoid the slippage phenomenon of UHMWPE in extrusion processing by Mitsui Petrochemical Company. Depth and width of grooves are gradually decreasing along the extrusion direction, which is favourable for establishing pressure. The principle of establishing pressure is similar to the advanced extrusion system of Institute of Plastics Processing (IKV) at RWTH Aachen University. The pressure peak of extrusion system usually appears at the end of the solid conveying section. Such extrusion system is good for improvement of the extrusion output and stability. Many other special extruders are developed to solve delivery problems for UHMWPE. However, such extruders only increase the coefficient friction between polymer and barrel and do not change the conveying mechanism, which resulting in the increases of wear of screw and barrel, drive load, and friction heat. Although the energy consumption is increasing, single screw extruder with large thrust screw and special screw structures for enhancing conveying capability is the most practical processing device for UHMWPE at present.

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As we know, in extruding process, common thermoplastic polymers successively experience solid state, viscous flow state, and high elastic state from feeding section to heating die. However, UHMWPE only experiences two physical states inside the barrel, namely solid state and high elastic state. The rotating screw consecutively grabbed UHMWPE powder particles from the feed inlet and compact them into block. The block is like a solid plug and conveys spirally forward along the rectangular tunnel by the trust of screw flight. Even the block is completely melted in metering section, it still moves forward as a whole. In other words, the flow mode of UHMWPE is plug flow, and there is almost no material exchange during the extrusion process. Therefore, the melting process of UHMWPE is similar to that of it under the static action by compression loads.

Polymer melt can be extruded out the extrusion die and forms continuous extrudates with specified cross-sectional shape as the die. During this process, polymer melt passes through the convergent channel inside extruder head and produces a shear deformation. Within the appropriate extrusion processing window, polymer melt with good flowability will maintain as a continuous melt block after flowing through the convergent channel.

As the schematic diagram shown in **Figure 4**, UHMWPE melt has very low critical shear rate, and the extrusion process will produce significant fluctuations when the shear rate is more than 10^{-2} s^{-1}. Many facts have shown that pure UHMWPE melt is easy to fracture even the extrusion speed was less than 10 r/min. The possible reason is that UHMWPE melt is fractured by shearing effect from deformation, and the fractured melt could not quickly merge to be continuous melt block again due to the poor molecular mobility of UHMWPE.

Many measures have been taken to prevent the unstable extrusion process and to improve the surface quality of extrudates, for instance, extending the length of the parallel flow path behind the convergent channel, reducing the angle of convergence, and reducing the friction resistance by using lubricants. Nevertheless, UHMWPE productivity and production efficiency have not improved significantly.

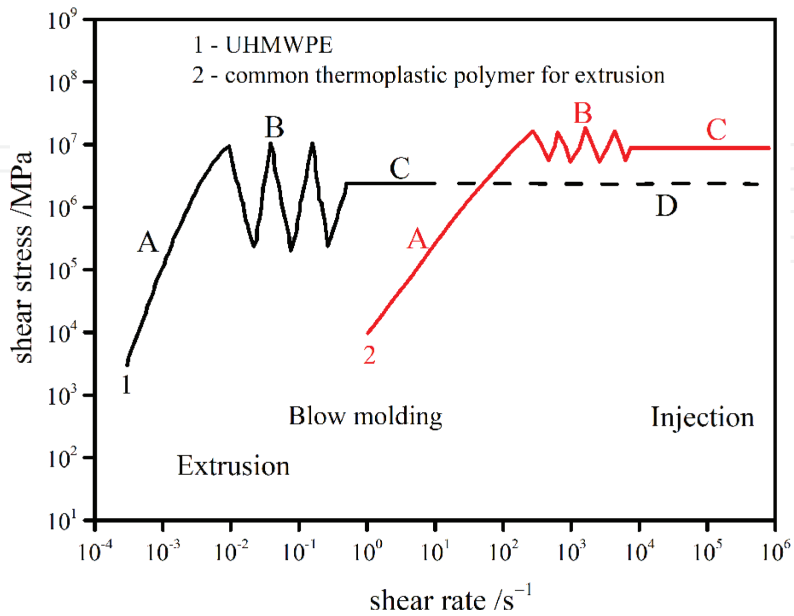

Figure 4. Schematic diagram of UHMWPE melt flow state during extrusion processing.

3.2.2. Twin screw extrusion

According to the relative rotation of two screws, twin screw extruders can be divided into counter rotating and co-rotating. The extruder of counter rotating has discontinuous channel that the spiral forward channel of screw is blocked by the screw flight of another screw with opposite rotating direction. For solid plug of UHMWPE, the continuous melt will be crushed into pieces by the strong shearing and mixing effect, and these crushed melt cannot quickly re-fuse to be a continuous melt block, causing significant instability extrusion.

Compared with counter rotating twin screw extruder, co-rotating twin screw extruder has a continuous channel to connect the feeding section and heating die, possessing good self-cleaning, and forcibly conveying capacity. The solid plug of UHMWPE can be conveyed forward along the continuous channel under positive displacement force. Such kind of extruder can effectively prevent material slippage and blockage.

At present, two types of twin-screw extruder are usually used to extrude UHMWPE in industrial production [20, 37, 38]. However, the difficult realities of low extrusion output, high energy consumption, and large driving load are still troubling people and hindering the development of UHMWPE processing.

3.2.3. Novel extrusion process of UHMWPE

As previously summarized, the nascent UHMWPE could be directly processed via compression moulding and ram extrusion. However, in most cases, processing aids should be required for screw extrusion even using an appropriative screw with special structures, and the excellent performances of UHMWPE would be damaged evidently. On the other hand, strong shearing action of screw extruder could make ultra-long molecular chains broken and even cause thermal degradation of UHMWPE.

Some new processing technologies are adopted to process UHMWPE in order to improve the extrusion output and reduce damage of material properties such as ultrasonic-assisted extrusion, gas-assisted extrusion, and near melting point extrusion process [39, 40]. Despite some progress, these extrusion processes still cannot extrude nascent UHMWPE without processing aids.

With regard to novel equipment for polymer processing, professor Qu and his research team [41–43] have independently developed novel extruders without screws such as vane extruder as shown in **Figure 5** [42] and eccentric rotor extruder as shown in **Figure 6** [43].

Compared with screw extruders, the rotor instead of screw was used in novel extruders and the rotor rotated eccentrically. The stator and rotor with special structures constitute continuous spatial path. The space volume between the rotor and stator periodically changes along the stator axial direction, which will make the volume of materials inside the spatial path change periodically along the axial direction, so that the materials were mainly subjected to stretch deformation. Put another way, the main flow field in such novel extruder was dominated by elongational flow field, and the materials were conveyed forward by positive displacement action.

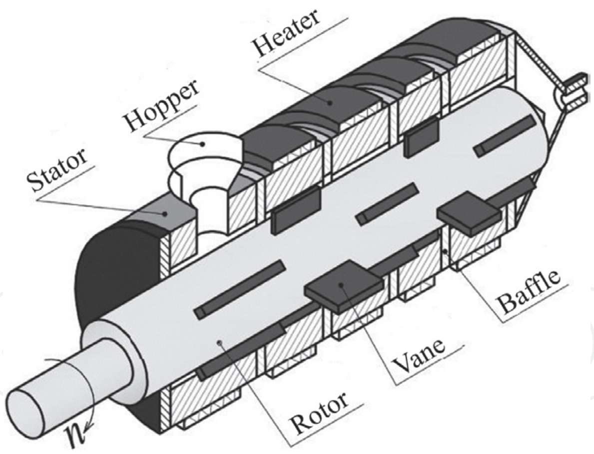

Figure 5. The schematic diagram of vane plasticizing and conveying system.

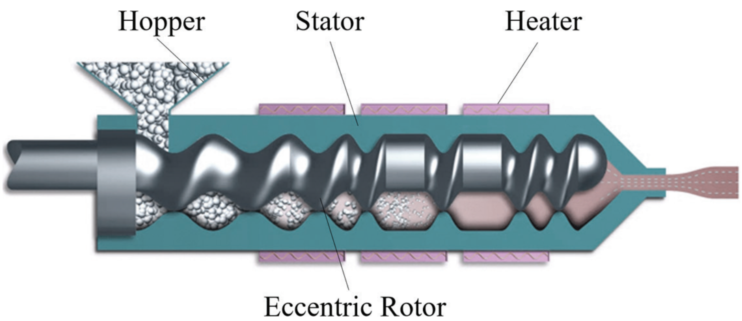

Figure 6. The schematic diagram of the eccentric rotor plasticizing and conveying system.

The eccentric rotor extruder has the same basic processing principle as vane extruder. Furthermore, eccentric rotor extruder has streamlined flow path, which is conducive to prevent thermal degradation. Nascent UHMWPE powder particles without any processing aids can be extruded directly by using an eccentric rotor extruder. The mechanical properties of the extrudates have almost been maintained as the unextruded samples. Since the material was conveyed by positive displacement force in the entire extrusion process, the energy consumption and extrusion output of UHMWPE were almost the same as that of HDPE. The melting of UHMWPE inside eccentric rotor extruder is a dynamic melting process because of the periodic variation of spatial path. However, the fluctuation of extrusion caused by the pulsating movement of eccentric rotor is one of the problems to be solved at present. Regardless of the current problems, the novel processing based on positive displacement delivery can significantly improve the production efficiency and quality of nascent UHMWPE.

4. Conclusions

The ultra-high molecular weight of UHMWPE has led to great difficulty in processing such as poor dissolution, extremely high melt viscosity, and poor melt flowability. The improved screw equipment with special structures can process UHMWPE with molecular weight of up to several hundred g/mol, but such modified upgrade equipment still cannot process the nascent UHMWPE without plasticizer and lubricants. The novel extrusion device of eccentric rotor extruder based on pulsating movement can directly process nascent UHMWPE without using any processing aids. Since the material is conveyed by positive displacement force in the entire extrusion process, the energy consumption and extrusion output of UHMWPE are almost the same as that of HDPE, showing small dependence on molecular weight. The melting process of UHMWPE inside eccentric rotor extruder is a dynamic process because of the periodic variation of spatial path.

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