Research paper

Numerical Simulation of gun propellant substitute in barrel during extrusion processing assisted with SC-CO_2

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Abstract

In this work, Polyflow was applied to simulate the flow behaviors of gun propellant substitute in the extruder barrel during extrusion processing assisted with supercritical carbon dioxide (SC-CO_2). The results demonstrate that the presence of SC-CO_2 leads to the great reduction of pressure and viscosity of the fluid, which is beneficial for the safety in extrusion processing. The shear rate and viscosity appear the lowest values near the screw thread, and they increase higher in the screw channel. The high screw speed brings the high production, but it causes the high pressure distribution, enhancing the processing risk. Although both high processing temperature and solvent content are in favor of decreasing the pressure and improving the flow behaviors of CA/SC-CO_2 solution, the excessive processing temperature or solvent content weakens the plasticization of SC-CO_2 to CA solution. The pressure of CA/SC-CO_2 solution at 45 °C is closed to the pressure of CA solution at 65 °C, which means that the extrusion processing can be reduced by almost 20 °C with the assistance of SC-CO_2. Meanwhile, the solvent concentration can be reduced by SC-CO_2 while the viscosity of CA solution is kept in the same level.

Keywords: gun propellant, SC-CO_2, extrusion, Polyflow, flow behaviors

1. Introduction

The screw extrusion of gun propellants has been drawn more and more attention in the field of gun propellant manufacturing technique. Compared with the batch process, the screw extrusion is a continuous process with high yield, which means that the amount of ingredients can be processed without limits in a single run[1]. Meanwhile, the products manufactured from the screw extrusion process are consistent and compact, which guarantees less variation in the mechanical and burning properties of gun propellants. However, as the development of the weapon system, there are more and more drawbacks appearing in the extrusion processing. The inflammability, explosibility and high viscosity of gun propellants go against the safety of extrusion processing. Increasing the content of solvents can improve the fluid flow, but most of the solvents are toxic and environmentally unfriendly, and the high content of solvents leads to the serious shrink of gun propellants. What's worse, the residual solvents lead to the negative impacts on the burning behaviors and the storage stability of gun propellants. Therefore, it's an urgent issue to figure out these conflicts.

Supercritical carbon dioxide (SC-CO_2) has been applied in polymer processing as an outstanding plasticizer to decrease the high viscosity of polymer melts, such as ultra-high molecular weight polyethylene (UHWPE)[2], polystyrene (PS)[3] and polymethyl methacrylate (PMMA)[4]. Royer et al. reported that the viscosity of PS melt was lowered by as much as 80% in the presence of SC-CO_2[5]. Curia et al. found that the melting point of Poly (ε-caprolactone) (PCL) reduced by more than 20 °C and the melt viscosity lowered by 90% with the addition of SC-CO_2[6]. SC-CO_2 assisted extrusion technology provides a promising method of improving the flow behaviors of gun propellants and replacing some amount of the organic solvents, and it has prominent influence on enhancing the safety factor in the extrusion processing.

Besides of the high viscosity of the gun propellant, the cost of extruder equipment and the real-time monitoring are two other barriers to the development of the gun propellant continuous extrusion. In the debugging purposes of extruder, the assembly of screw, normal operation of extruder and measurement of equipment parameters cost a large amount of manpower, resources and financial capacities. Polyflow is a professional computer assisted software to simulate the extrusion processing of polymers. With the required parameters and data provided by Polyflow, the long design period of screw can be reduced dramatically, and the processing parameters can be optimized to improve the product quality. Besides, during the extrusion processing of gun propellants, it's difficult to accurately monitor the flow behaviors of the fluid in real time. Polyflow simulates the flow behaviors of gun propellants under various conditions and predicts the possible danger in advance. With the assistance of Polyflow, the extruder can produce different types of gun propellants to meet various demand safely. Zhou simulated the viscosity of solid propellant in the single screw extrusion process by Polyflow[7]. Zhong et al. explored the temperature, pressure and velocity distribution of GR-35 double-base propellant in the extrusion process through Polyflow[8]. However, the rheological parameters that they employed for Polyflow were measured from the off-line rheometer, and these parameters couldn't accurately describe the rheological properties in the extrusion processing.

Although a few literatures have studied the numerical simulation of energetic materials in recent years, there is no research reporting the flow behaviors of gun propellants in the barrel during extrusion processing assisted with SC-CO_2 simulated by Polyflow. Cellulose acetate (CA) is commonly selected as the substitute for nitrocellulose to safely investigate some properties of gun propellants in the previous researches, especially in the investigations of extrusion processing. In this work, the flow behaviors of CA/SC-CO_2 solution in the extruder barrel during extrusion processing were simulated under different conditions by Polyflow.

2. Numerical simulation

The screw section behind the gas injection section was selected for the numerical simulation. Figure 1 shows the finite element model of the screw and the flow channel. The finite element mesh of the screw (Figure 1(a)) is tetrahedron, while the finite element mesh of the flow channel (Figure 1(b)) is hexahedron. The diameter of the screw is 30 mm, the channel depth is 3 mm, and the lead of the screw is 26 mm. The length, inner and external diameter of the flow channel are 78 mm, 24 mm and 30 mm, respectively. The coordinate origin is located at the center of the flow inlet.

The in-line rheological parameters of CA/SC-CO_2 solution were obtained from our previous research[9]. In this work, power law is selected as the constitutive

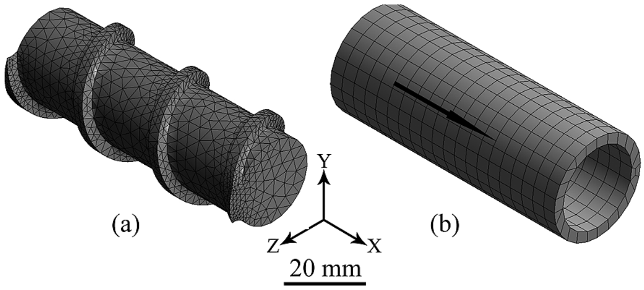

Figure 1 Finite element model of (a) single screw and (b) flow channel.

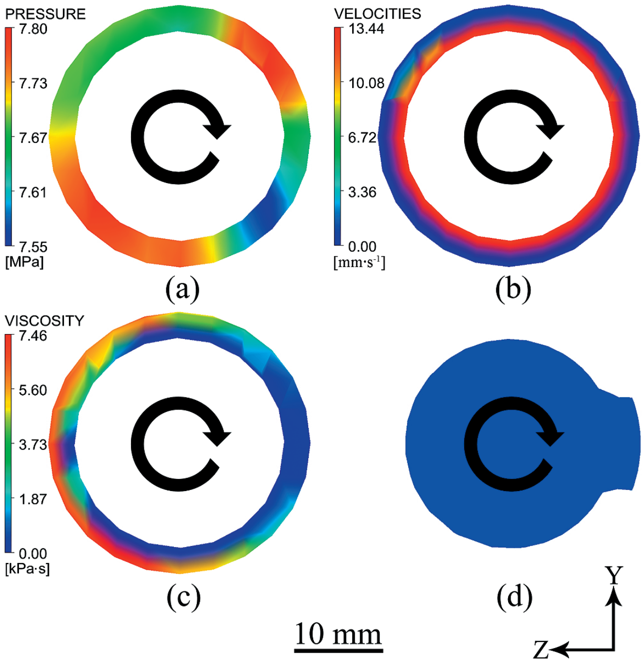

Figure 2 (a) Pressure, (b) velocity, (c) viscosity, and (d) screw distributions in YZ section. (T = 50 °C, r = 10 r·min^{-1}, c = 1.20 mL·g^{-1})

equation to express the rheological properties of CA/SC-CO_2 solution[10),11)]. The ratio of the volume of mixed solvents (V_s) to the mass of gun propellant substitute (m) was set to c (mL·g^{-1})

3. Results and discussion

3.1 Pressure, velocity and viscosity distributions

Figure 2 shows the pressure (Figure 2(a)), velocity (Figure 2(b)) and viscosity (Figure 2(c)) distributions of the CA/SC-CO_2 solution in YZ section (X = 39 mm) when the processing temperature is 50 °C, the speed rate (r) is 10 r·min^{-1}, and solvent content is 1.20 mL·g^{-1}, and Figure 2(d) is the cross section of the screw. The pressure of the section ranges from 7.55 MPa to 7.80 MPa, and the pressure value near the screw is closed to the value near the barrel. Therefore, the pressure transducer can be installed in the barrel to measure the real-time pressure of the fluid. In addition, the pressure under this condition is higher than the critical pressure of SC-CO_2 (7.38 MPa) but lower than the injection pressure of CO_2, which guarantees the strong solvation of SC-CO_2 and the continuous injection of CO_2. In the extrusion processing, the flow of fluid contains not only the flow along the extrusion direction but also the axial rotation by the

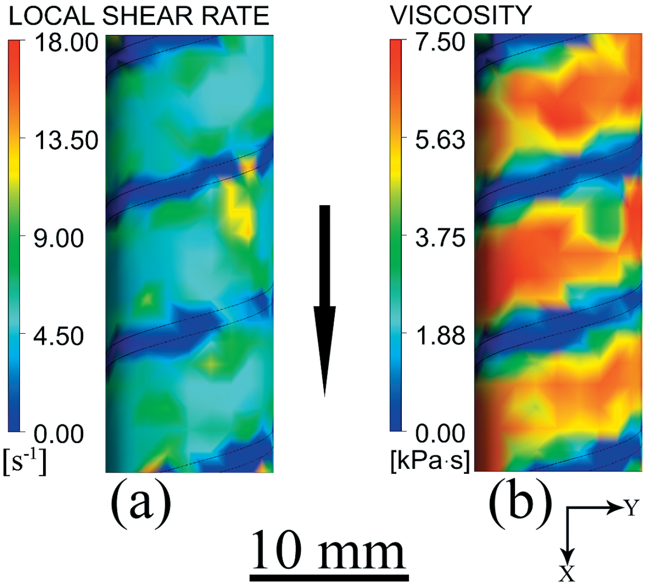

Figure 3 (a) Shear rate and viscosity (b) distributions in the surface of the fluid.
($T = 50$ °C, $r = 10$ r·min^{-1}, $c = 1.20$ mL·g^{-1})

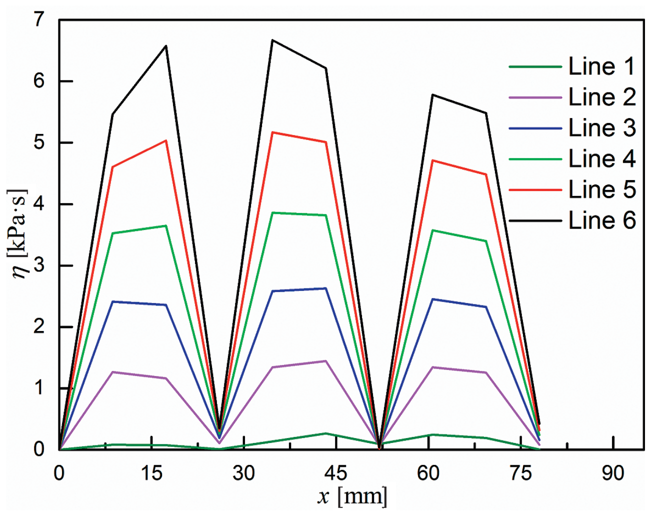

Figure 4 Viscosity distributions along the lines in XZ section.
($T = 50$ °C, $r = 10$ r·min^{-1}, $c = 1.20$ mL·g^{-1})

rotating screw. The highest velocity appears near the screw (13.44 mm·s^{-1}), and the velocity decreases along the diameter direction of the fluid smoothly. Figure 2(c) and Figure 2(d) reveal that the viscosity in the right side is much lower than that in the left side. Under this condition, the volume in the screw channel is higher than the volume near the screw thread, so the volume flow rate is higher in the screw channel, which leads to the higher viscosity in the left side.

Figure 3 displays the shear rate and viscosity distributions in the surface of the fluid at 50 °C and 10 r·min^{-1} ($c = 1.20$ mL·g^{-1}). It's obvious that the shear rate and the viscosity appear the lowest values near the screw thread, and they increase higher in the screw channel. Besides, the variation of shear rate and viscosity distributions of the fluid is almost the same among each screw lead.

Figure 4 is the viscosity distributions at 50 °C and 10 r·min^{-1} along five lines parallel to X axis in XZ section, and

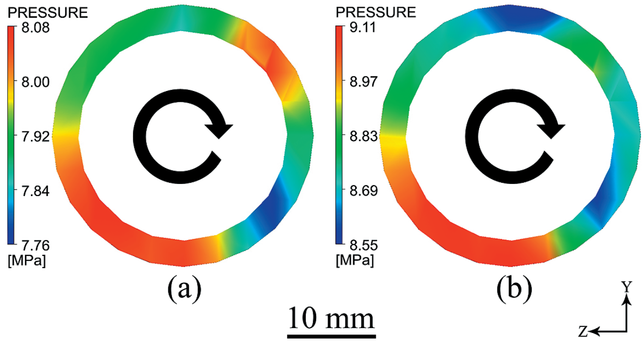

Figure 5 Pressure distributions in YZ section: (a) CA/SC-CO_2 solution, (b) CA solution.
($T = 45$ °C, $r = 10$ r·min^{-1}, $c = 1.20$ mL·g^{-1})

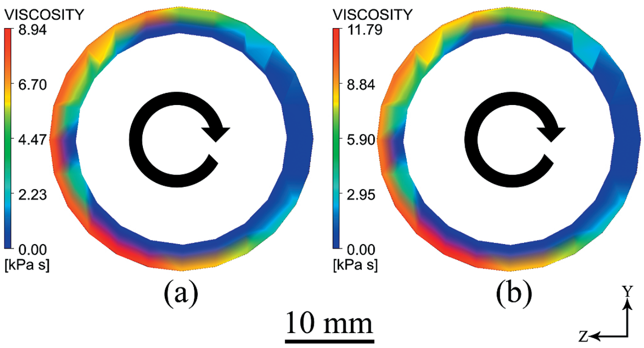

Figure 6 Viscosity distributions in YZ section: (a) CA/SC-CO_2 solution, (b) CA solution.
($T = 45$ °C, $r = 10$ r·min^{-1}, $c = 1.20$ mL·g^{-1})

the initial points in Z axis are 12.00 mm (Line 1), 12.60 mm (Line 2), 13.20 mm (Line 3), 13.80 mm (Line 4), 14.40 mm (Line 5), and 15.00 mm (Line 6). Similar to Figure 3(b), the viscosity distributions of CA/SC-CO_2 solution behave almost the same among each screw lead. The valley values appear near the screw thread, and the peak values appear in the screw channel. And the values of viscosity increase as the lines get farther away from the inwall of fluid.

3.2 Effects of SC-CO_2 on flow behaviors

In the SC-CO_2 assisted extrusion processing, the most important influence is the decrease of the pressure and viscosity of the fluid. Figure 5 and Figure 6 indicate the effects of SC-CO_2 on the pressure and viscosity in YZ section at 45 °C and 10 r·min^{-1} ($c = 1.20$ mL·g^{-1}), respectively. It can be found that the injection of SC-CO_2 decreases the highest pressure and viscosity of CA solution by 11% and 24%, respectively, which is significant for the safety of gun propellant extrusion. The average pressure and viscosity in the geometrical model are calculated by Ansys post-processing software. The average pressure of CA solution is 8.79 MPa, and the average pressure of CA/SC-CO_2 solution is 7.98 MPa. With the assistance of SC-CO_2, the viscosity of CA/SC-CO_2 solution decreases to 10.94 kPa·s while the viscosity of CA solution is 29.76 kPa·s. Therefore, SC-CO_2 is an excellent plasticizer for CA solution to decrease the pressure and viscosity in the extrusion processing.

3.3 Effects of screw speed on the flow behaviors

The screw speed is an important processing parameter

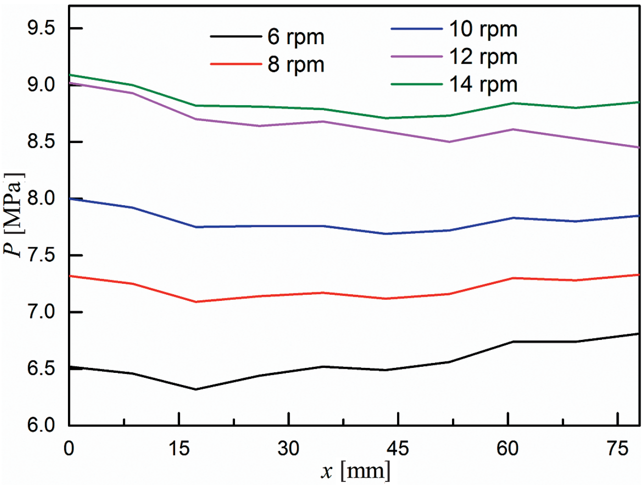

Figure 7 Effects of screw speed on pressure distributions. (T = 50 °C, c = 1.20 mL·g^{-1})

Table 1 Average pressure and velocity at different screw speeds. (T = 50 °C, c = 1.20 mL·g^{-1})

r [r·min^{-1}]	6	8	10	12	14
$\overline{P}$ [MPa]	6.48	7.14	7.73	8.56	8.77
$\overline{v}$ [mm·s^{-1}]	3.67	4.88	6.11	7.38	8.55

in the extrusion processing. Increasing the screw speed raises the production of gun propellants, but it will increase the pressure and the risk in the extrusion processing at the same time. In order to investigate the influences of the screw speed on the flow behaviors of CA/SC-CO_2 solution, the pressure along a line (Line 6) in the surface of the fluid at 50 °C (c = 1.20 mL·g^{-1}) is shown as Figure 7, and the average pressure and velocity at different screw speeds are listed in Table 1. The results reveal that increasing the screw speed increases the pressure and velocity of the fluid remarkably. The higher velocity is beneficial for improving the production efficiency of extrusion processing, but one the other hand, the higher pressure brings more risk factors to the gun propellant extrusion. Meanwhile, the pressure curves tend to fluctuate more widely when the screw speed is too low or high. And when the screw speed is 8 r·min^{-1} or 10 r·min^{-1}, the pressure curves perform steadily, which contributes to improving the quality of gun propellants. Thus, the screw speed is set at 10 r·min^{-1} in the following experiment.

3.4 Effects of temperature on the flow behaviors

In the gun propellant extrusion processing, the processing temperature is a key parameter to control the flow behaviors of the fluid and the quality of the product. Figure 8 demonstrates the effects of processing temperature on the viscosity distributions of CA/SC-CO_2 solution along Line 6 at 10 r·min^{-1} (c = 1.20 mL·g^{-1}). It's observed that the higher temperature reduces the viscosity and improves the flow behaviors of the fluid, and the decrement is obvious when the temperature is lower than 50 °C. The high temperature improves the thermal motion of CA molecular chains and increases the free

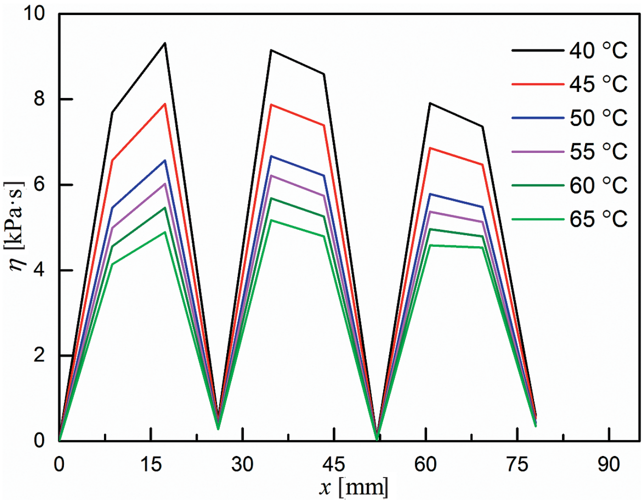

Figure 8 Effects of temperature on viscosity distributions. (r = 10 r·min^{-1}, c = 1.20 mL·g^{-1})

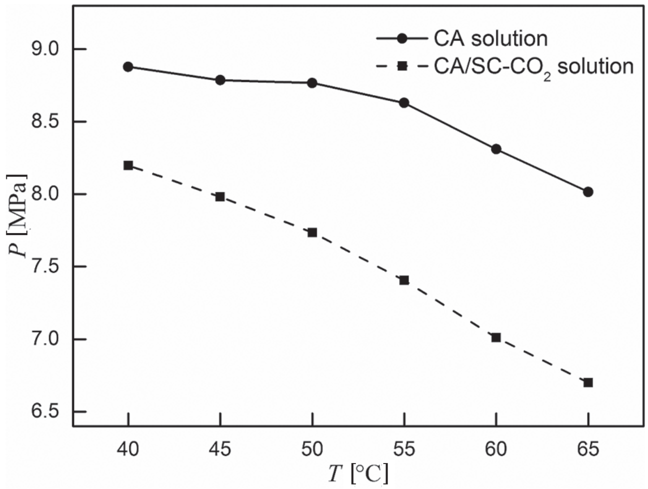

Figure 9 The average pressure at various temperatures. (r = 10 r·min^{-1}, c = 1.20 mL·g^{-1})

volume of CA/SC-CO_2 solution, which leads to the decrease of viscosity. But at the same time, the high temperature weakens the attraction between CA and SC-CO_2, and it's not in favor of the dissolution and plasticization of SC-CO_2 into CA solution. The low melt strength of CA solution is also adverse to the solubility of SC-CO_2 at high temperatures. Therefore, the effects of temperature on the viscosity become weak when the temperatures are in the range from 50 °C to 65 °C.

As indicated in Figure 9, increasing the temperature decreases the average pressures of CA/SC-CO_2 solution and CA solution at 10 r·min^{-1}. When the temperature rises from 40 °C to 65 °C, the average pressure of CA/SC-CO_2 solution decreases from 8.20 MPa to 6.70 MPa, and the decrement is 18.29%. The average pressure of CA/SC-CO_2 solution at 45 °C is 7.98 MPa, and the average pressure of CA solution at 65 °C is 8.02 MPa. In other words, the injection of SC-CO_2 is a suitable method to reduce the processing temperature by almost 20 °C in gun propellant extrusion. Furthermore, the effects of temperature on the average pressure of CA/SC-CO_2 solution are more remarkable than those of CA solution. It seems that the high temperature leads to the escape of CO_2, and the CO_2 gas accelerates the decrease of the fluid pressure.

3.5 Effects of solvent content on the flow behaviors

The solvent content is also vital to the extrusion of gun propellants. The high solvent content improves the flow of the fluid and the safety of extrusion processing. However, the excessive solvent content brings disadvantages to the driving-solvent procedure, and the residual solvents in the gun propellants have negative impacts on the mechanical property and the combustion performance of the products. The injection of SC-CO_2 can reduce the solvent concentration. Thus, it's essential to study a reasonable solvent content to the extrusion of gun propellants.

Figure 10 implies that increasing the solvent content decreases the viscosity of CA/SC-CO_2 solution at 50 °C and 10 r·min^{-1}. When the solvent content increases from 1.10 mL·g^{-1} to 1.20 mL·g^{-1} or increases from 1.30 mL·g^{-1} to 1.40 mL·g^{-1}, the viscosity shows a remarkable decrement. Because of the high viscosity with low solvent content, increasing the solvent content reduces the viscosity obviously. When the solvent content was 1.40 mL·g^{-1}, the melt strength of CA/SC-CO_2 solution was too low that the melt fracture phenomenon took place in the experiment. Therefore, 1.20 mL·g^{-1} is an appropriate solvent content for the extrusion of CA/SC-CO_2 solution.

The effects of the solvent content on the average viscosity of the fluid at 50 °C and 10 r·min^{-1} is shown in Figure 11. The increasing solvent content reduces the average viscosity. In the presence of SC-CO_2, the viscosity of CA/SC-CO_2 solution is much lower than that of CA solution, and the viscosity decreases dramatically from 88.68 kPa·s to 26.03 kPa·s when c is equal to 1.10 mL·g^{-1}. Subsequently, the plasticization of SC-CO_2 becomes weak when the solvent content is higher than 1.30 mL·g^{-1}. In addition, when c is 1.10 mL·g^{-1}, the average viscosity of CA/SC-CO_2 solution is 26.03 kPa·s, and the average viscosity of CA solution is 26.74 kPa·s while c is 1.25 mL·g^{-1}. In the SC-CO_2 assisted extrusion processing, SC-CO_2 is completely driven out from CA solution in the form of CO_2 gas in the end of extruder, so SC-CO_2 is an ideal solvent and plasticization to replace some amount of the organic solvents, and it has few effects on the mechanical and combustion properties of the final products. Therefore, with the assistance of SC-CO_2, the solvent concentration can be reduced by SC-CO_2 in the gun propellant extrusion processing, which improves the quality of gun propellants.

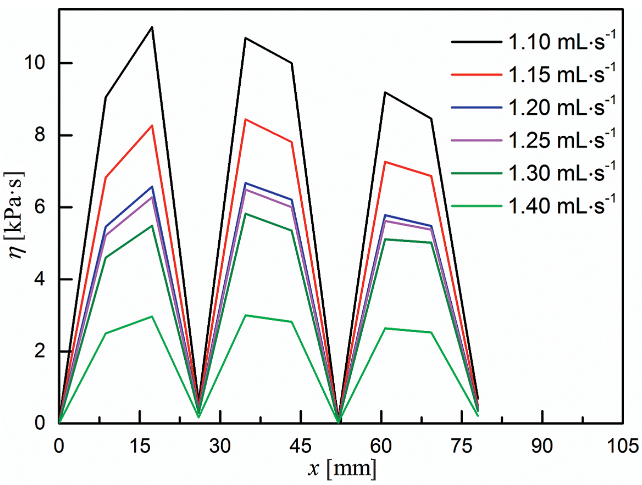

Figure10 Effects of solvent content on viscosity distributions. (T = 50 °C, r = 10 r·min^{-1})

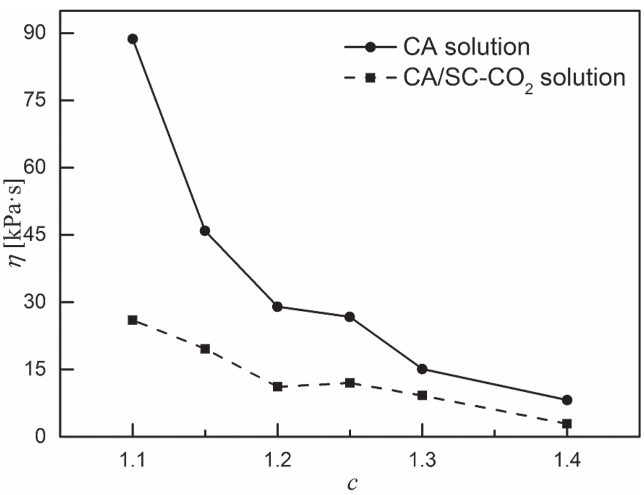

Figure11 The average viscosity with different solvent content. (T = 50 °C, r = 10 r·min^{-1})

4. Conclusions

Polyflow was applied to investigate the flow behaviors of gun propellant substitute in the extruder barrel during the extrusion processing assisted with SC-CO_2. The results demonstrate that the presence of SC-CO_2 results in great reduction of the pressure and viscosity of the fluid, which is beneficial for the extrusion processing safety. The injection of SC-CO_2 decreases the highest pressure and viscosity of CA solution by 11% and 24% at 45 °C, respectively. The shear rate and the viscosity appear the lowest values near the screw thread, and they increase higher in the screw channel. The high screw speed brings the high production, but it leads to the high pressure distributions at the same time which enhances the processing risk. Both high processing temperature and solvent content are in favor of decreasing the pressure and improving the flow behaviors of CA/SC-CO_2 solution. However, the excessive processing temperature or solvent content weakens the plasticization of SC-CO_2 and the melt strength of CA/SC-CO_2 solution. Meanwhile, the pressure of CA/SC-CO_2 solution at 45 °C is closed to the pressure of CA solution at 65 °C, which means that the extrusion processing can be reduced by almost 20 °C with the assistance of SC-CO_2. The solvent concentration can be reduced by SC-CO_2 while the viscosity of CA solution is kept in the same level.

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