Research paper

Numerical simulation of gun propellant substitute in die during extrusion processing assisted with SC-CO_2

Yajun Ding*, Rongjun Wei*, and Sanjiu Ying*†

*School of Chemical Engineering, Nanjing University of Science and Technology, Xiaolingwei 200#, Nanjing 210094, PEOPLE REPUBLIC OF CHINA
Phone: +86−25−84315138
†Corresponding author: yingsanjiu@126.com

4
1
5

Received: October 13, 2016 Accepted: June 13, 2017

Abstract

To simulate the flow behaviors of gun propellants in extrusion processing assisted with supercritical carbon dioxide (SC-CO_2), a numerical software Polyflow was applied to investigate the pressure, viscosity and velocity distributions of CA/SC-CO_2 mixture under various processing conditions. The results demonstrate that the highest pressure of the die appears at the entrance of the die, and the pressure decreases smoothly to the atmospheric pressure. The injection of SC-CO_2 reduces the viscosity and the pressure of the flow significantly. Increasing the length of the die raises the inlet pressure, but it has few effects on the viscosity distribution. Increasing both the processing temperature and the solvent content can decrease the pressure in the die. The inlet pressure reduces by about 30%, when the temperature rises from 45°C (9.66 MPa) to 55°C (6.77 MPa). Besides, too high solvent content leads to the structural damage of the products. The numerical simulation of CA/CO_2 mixture provides the fundamental and significant experimental data for the extrusion of gun propellants assisted with SC-CO_2.

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

1. Introduction

Gun propellants play an important role in the conventional weapons. As the highly viscous, combustible and explosive materials, the batch process is the common method to manufacture gun propellant products. However, with the development of the weapon system, the product quality and production efficiency of the batch process technology can't meet the military requirements today. In recent decades, the screw extrusion has been drawn more and more attention to the continuous processing of gun propellants[1].

In the gun propellant extrusion, the rheological behaviors of the fluids have significant effects on the product quality and the processing safety in the closed barrels. Increasing the processing temperature and the solvent content are the general ways to reduce the high viscosity and pressure of gun propellants in the extruder, but it leads to the negative impacts on the mechanical and burning properties of the products.

Supercritical carbon dioxide (SC-CO_2) has been attracted much interest in the polymer extrusion processing as an outstanding solvent and plasticizer. Royer found that the addition of SC-CO_2 lowered the viscosity of polystyrene (PS) melt as much as 80−90%[2]. Iguchi et al.[3] measured the viscosity of polyethylene glycol (PEG) with SC-CO_2, and the results demonstrated that the viscosity reduction ratio of the CO_2-PEG solutions was up to 40 %. The introduction of SC-CO_2 can deal with many disadvantages in the gun propellant extrusion. The plasticization of SC-CO_2 decreases the viscosity of gun propellants and improves the fluid flow. In addition, the frictional heat resulting from the screw and gun propellants can be absorbed by the phase transformation of SC-CO_2 to CO_2 gas. As CO_2 gas leaves out of the gun propellants in the exit of the die, the addition of SC-CO_2 won't destroy the final products. Thus, the injection of SC-CO_2 is a promising approach to deal with the high viscosity and pressure in the gun propellant extrusion processing safely.

In the energetic material processing field, many researchers focus on the off-line rheological behaviors, and there are few literatures reported about the in-line rheological behaviors[4), 5)]. And because of the limitation of the experimental equipment, the fluid flow behaviors in each position can't be obtained except at transducer positions. At the same time, the special properties of gun propellants demand the accurate control of the fluid flow under different conditions. Therefore, it's an urgent issue to solve this conflict.

Polyflow is a professional computational fluid dynamic software, which is widely applied in the numerical simulation for polymer extrusion, blends and blow molding[6)]. With the assistance of Polyflow in gun propellant processing, both the time and cost for designing and machining die are saved, and the product quality can be improved by analyzing the fluid flow under different conditions. Zhong et al.[7)] used Polyflow to simulate the extrusion process of GR-35 double-base propellant, and analyzed the temperature, the pressure, the shear rate and the velocity at the axial direction.

However, in the current literature, there is a lack of any direct study of the numerical simulation of gun propellants assisted with SC-CO_2 in extrusion processing. Because of the inflammability of nitrocellulose, cellulose acetate (CA) is a suitable substitute to investigate the rheological behaviors of gun propellants, particularly in the gun propellant extrusion. In this work, the pressure, viscosity and velocity distributions of CA/SC-CO_2 mixture were investigated extensively by Polyflow under various processing conditions.

2. Numerical simulation

In order to gain the basic data for numerical simulation, the in-line rheological behaviors of CA/SC-CO_2 mixture was characterized by a slit die rheometer installed after the SC-CO_2 assisted extruder. With the data obtained from the rheometer, the in-line rheological behaviors of CA/SC-CO_2 mixture were analyzed as our previous research[8)]. And the basic assumptions and the governing equations were shown as this literature[6)]. Power law is selected as the constitutive equation as following :

$$\eta = K$$

where η is the viscosity of fluid (Pa·s), K is the consistency index (Pa·s^n), λ is the relaxation time (s), $\dot{\gamma}$ is the shear rate (s^{-1}) and n is the flow behavior index.

Figure 1 shows the 3D diagram of the flow passage. The geometrical model contains two parts. Subdomain A represents the fluid in the end of extruder and the die, and subdomain B is on behalf of the free subdomain out of the die. The diameter of the inlet is 15 mm, and the diameter of the calibrating section of the die is 5 mm. The length of the calibrating section of the die is set to L. The length of the free subdomain is 20 mm. As the symmetry of the flow passage, one quarter of the total 3D region was used for numerical simulation. The nodes and elements of the mesh are 10475 and 37023, respectively. The min mesh size is 1×10^{-4} m, and the max mesh size is 1×10^{-3} m. The value that expresses the mess quality is 0.23 by the Skewness module of Polyflow, while 0 means the perfect and 1 means the worst.

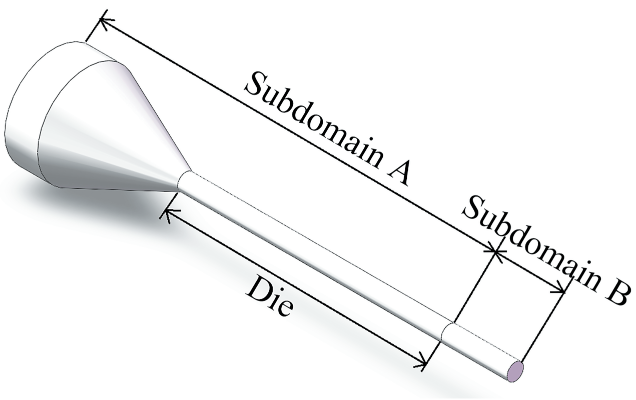

Figure 1 3D diagram of flow passage.

3. Results and discussion

3.1 Pressure, velocity and viscosity distributions

With the parameters obtained from the rheological characterization, the numerical simulation was taken under different conditions. Figure 2 shows the pressure, viscosity and velocity distributions along the extrusion direction at 50 °C ($L = 0.16$ m, $c = 1.20$, $V = 1 \times 10^{-7}$ m^3·s^{-1}), while the consistency index (K) and the flow behavior index (n) are 22020 Pa·s^n and 0.2242, respectively. 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}). And V is the volumetric flow rate (m^3·s^{-1}).

Figure 2 shows that the highest pressure appears at the inlet (7.70 MPa) and decreases smoothly to the value close to the atmospheric pressure. The values of viscosity around the inlet and outlet are high, and it is low in the die. Meanwhile, the viscosity near the wall of the die is lower than that at the center of the die. When the CA/CO_2 mixture flows into the die, the velocity increases to 30.15 mm^3·s^{-1} rapidly.

Figure 3 displays the pressure, viscosity and velocity distributions at the cross section in the middle of the die. The pressure distribution seems stable at the cross section, in the range from 3.6740 MPa to 3.6786 MPa, while the viscosity and the velocity in the center region are clearly much higher than those in other regions. Because of the effects of wall, the velocity of the fluid becomes lower along the direction from the center to the wall, resulting in the highest shear rate in the center of the die and the lowest shear rate around the wall. As CA/CO_2 flows is a kind of pseudoplastic fluids, the variation of shear rate leads to the distribution of the viscosity.

3.2 Effects of SC-CO_2

As we known, SC-CO_2 is an outstanding solvent and plasticizer, and the most intuitive expression of plasticization is the decrease of the viscosity and pressure. The average viscosity and pressure values with SC-CO_2 in the die are 10.25 kPa·s and 3.68 MPa, respectively, while the average viscosity and pressure values without SC-CO_2 are 53.05 kPa·s and 4.10 MPa, respectively.

The viscosity at the cross section of the die with and without SC-CO_2 is shown in Figure 4. It can be found that the injection of SC-CO_2 reduces the viscosity of the fluid

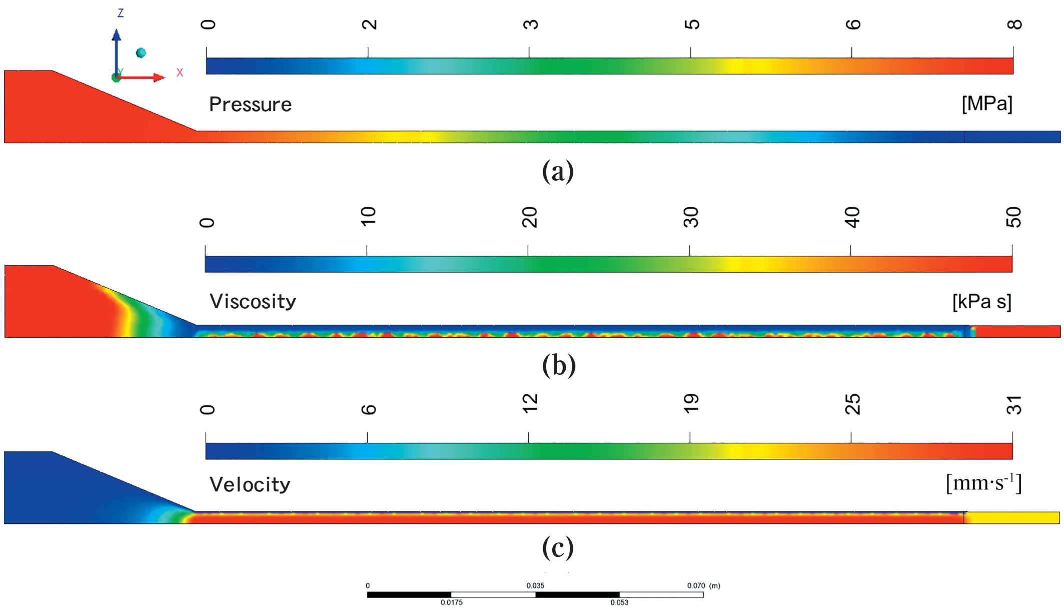

Figure 2 Nephograms of CA/SC-CO_2 mixture along the extrusion direction : (a) Pressure distributon, (b) Viscosity distribution, and (c) Velocity distribution.

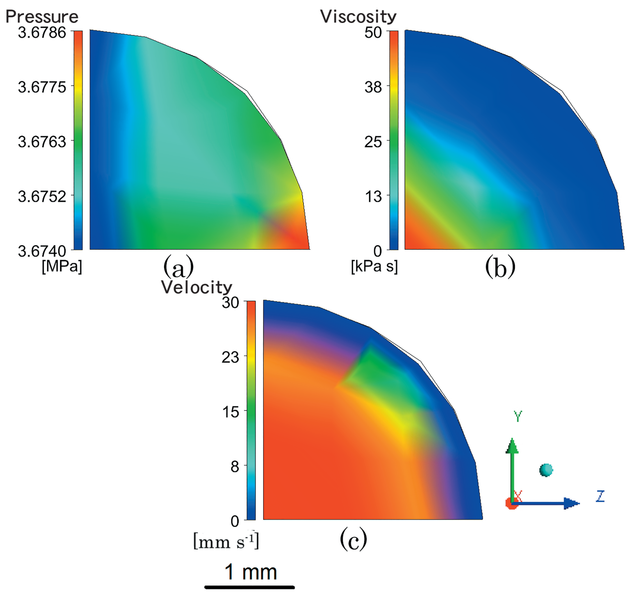

Figure 3 Nephograms of CA/SC-CO_2 mixture at the cross section in the middle of die : (a) Pressure distributon, (b) Viscosity distribution, (c) Velocity distribution.

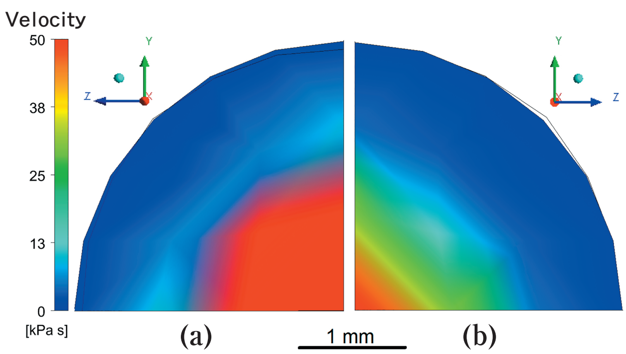

Figure 4 Viscosity distributions : (a) without SC-CO_2 , and (b) with SC-CO_2.

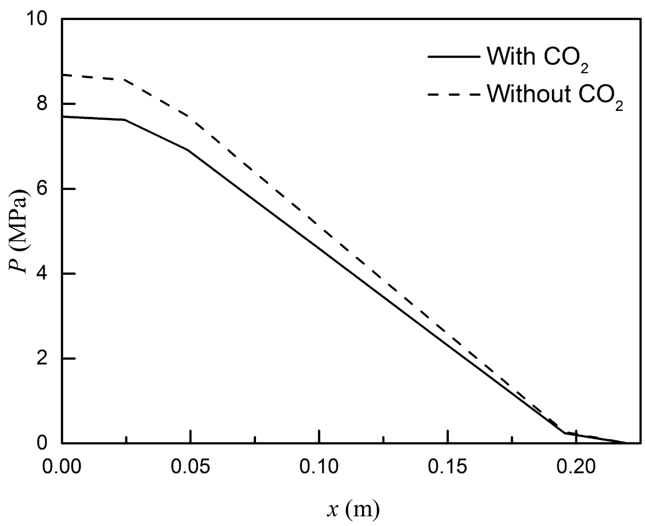

Figure 5 Effect of SC-CO_2 on pressure distribution.

significantly. The presence of SC-CO_2 enhances the free volume of CA solution, reducing the entanglement of macromolecular chains. Moreover, SC-CO_2 plays the role as a lubricating agent in the macromolecular chains. These factors contribute to decreasing the viscosity and improving the fluidity together. Therefore, the pressure distribution along the extrusion direction with SC-CO_2 is much lower than that without SC-CO_2, as plotted in Figure 5.

The pressure in the inlet without SC-CO_2 is 8.68 MPa, and the pressure with SC-CO_2 is 7.7 MPa, which means that the addition of SC-CO_2 makes a decrement of 11% to the pressure. In the field of gun propellant processing, the low processing pressure is beneficial to the safety production, so the introduction of SC-CO_2 improves the safety in gun propellant processing.

3.3 Effects of the die length

The effects of the die length on the pressure distribution are presented in Figure 6. The pressure decreases slowly

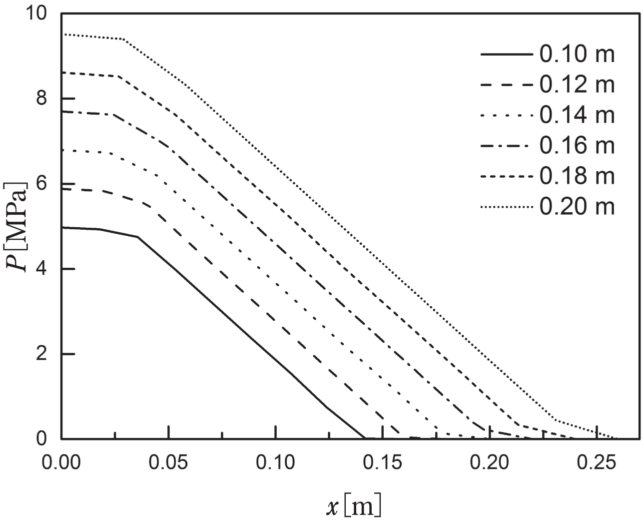

Figure 6 Effect of die length on pressure distribution. ($T = 50$ °C, $V = 1.0 \times 10^{-7}$ $m^3 \cdot s^{-1}$, $c = 1.20$ $mL \cdot g^{-1}$)

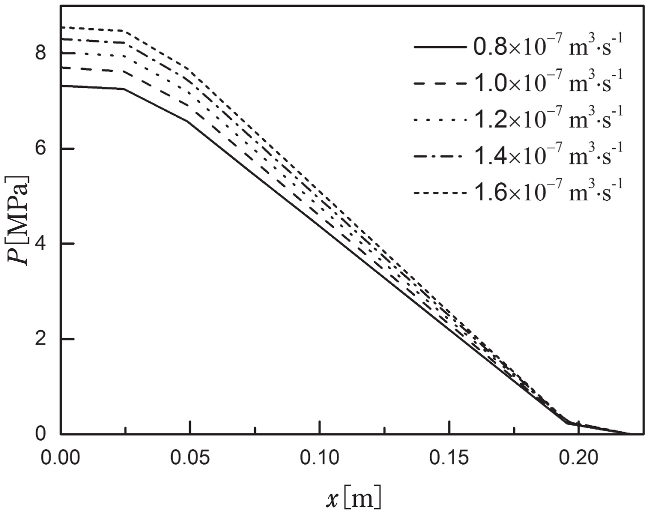

Figure 7 Effect of flow rate on pressure distribution. ($T = 50$ °C, $L = 0.16$ m, $c = 1.20$ $mL \cdot g^{-1}$)

Table 1 Effect of the length on the viscosity and pressure in the die. ($T = 50$ °C, $V = 1.0 \times 10^{-7}$ $m^3 \cdot s^{-1}$, $c = 1.20$ $mL \cdot g^{-1}$)

L[mm]	100	120	140	160	180	200
$\overline{\eta}$[kPa·s]	10.19	10.68	10.42	10.25	10.30	10.56
$\overline{P}$[MPa]	2.31	2.77	3.22	3.68	4.13	4.58
ΔP[MPa·m^{-1}]	45.77	45.51	45.24	45.51	45.42	45.59

Table 2 Viscosity and pressure in the die on various flow rates. ($T = 50$ °C, $L = 0.16$ m, $c = 1.20$ $mL \cdot g^{-1}$)

$V \times 10^{-7}$[m·s^{-1}]	0.8	1.0	1.2	1.4	1.6
$\overline{\eta}$[kPa·s]	12.19	10.25	8.90	7.90	7.12
$\overline{P}$[MPa]	3.50	3.68	3.83	3.96	4.08
ΔP[MPa·m^{-1}]	43.01	45.32	47.22	48.85	50.34

in the inlet, and it becomes faster in the die. It indicates that the pressure decreases by a constant value in the die, and the ratio of the pressure drops to the distance of the die is denoted by ΔP. The average viscosity ($\overline{\eta}$), the average pressure ($\overline{P}$) and ΔP in different die length are listed in Table 1.

The longer die raises the pressure of the inlet, but $\overline{\eta}$ and ΔP have small variation. The inlet pressure of 0.20 m (9.51 MPa) is almost twice higher than that of 0.10 m (4.97 MPa). On one hand, the long die is required to maintain the same flow rate, and it is in favor of shaping the products. On the other hand, the long die increasing the pressure of the calibrating section, which rises the processing risk of gun propellants. Thus, we selected 0.16 m as the length of the die for the numerical simulation below.

3.4 Effects of the flow rate

The pressure distributions for various flow rates are compared in Figure 7, and $\overline{\eta}$, $\overline{P}$ and ΔP are listed in Table 2. The results exhibit that the increase of the flow rate makes an increase in the inlet pressure and a decrease in the viscosity of the fluid. However, the flow rate does not have much influence on the inlet pressure. When the flow rate rises from 0.8×10^{-7} $m^3 \cdot s^{-1}$ to 1.6×10^{-7} $m^3 \cdot s^{-1}$, the inlet pressure increases from 7.32 MPa to 8.55 MPa. Besides, the die viscosity decreases by 42%, from 12.19 kPa·s to 7.12 kPa·s. As a non-Newtonian pseudoplastic fluid, the high flow rate causes the increase of the shear rate and the shear-thinning behavior of CA/SC-CO_2 mixture, resulting in the decrease of viscosity.

3.5 Effects of the processing temperature

In gun propellant processing, the high temperature improves the fluid flow, but it may lead to the combustion or even the explosion during extrusion processing. In order to investigate the flow behaviors of CA/SC-CO_2 mixture at various temperatures, the numerical simulation was taken based on the rheological parameters obtained from the in-line rheological characterization.

With reference to Figure 8 and Table 3, the results reveal that increasing the processing temperature improves the flow of CA/SC-CO_2 mixture. The high temperature leads to accelerating the thermal motion of CA molecular chains and increasing the free volume of

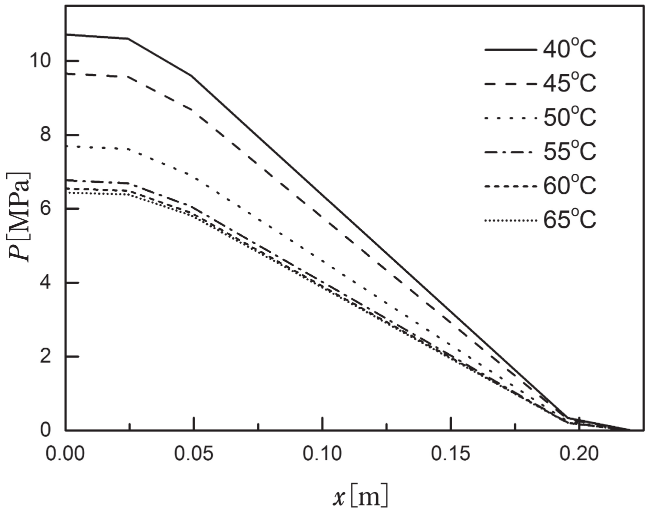

Figure 8 Effect of temperature on pressure distribution. ($L = 0.16$ m, $V = 1.0 \times 10^{-7}$ $m^3 \cdot s^{-1}$, $c = 1.20$ $mL \cdot g^{-1}$)

Table 3 Viscosity and pressure in the die at various temperatures. (L = 0.16 m, $V = 1.0 \times 10^{-7}$ $m^3 \cdot s^{-1}$, c = 1.20 $mL \cdot g^{-1}$)

T [°C]	40	45	50	55	60	65
$\bar{\eta}$[kPa·s]	17.32	12.59	10.25	9.69	7.24	4.31
$\bar{P}$[MPa]	5.11	4.62	3.68	3.22	3.13	3.10
ΔP[$MPa \cdot m^{-1}$]	62.93	56.85	45.32	39.62	38.53	38.26

Table 4 Viscosity and pressure in the die with different solvent contents. (T = 50 °C, L = 0.16 m, $V = 1.0 \times 10^{-7}$ $m^3 \cdot s^{-1}$)

c[$mL \cdot g^{-1}$]	1.10	1.15	1.20	1.25	1.30	1.40
$\bar{\eta}$[kPa·s]	29.55	22.39	10.25	11.74	8.37	2.59
$\bar{P}$[MPa]	5.82	4.31	3.67	3.41	3.07	1.89
ΔP[$MPa \cdot m^{-1}$]	71.64	53.05	45.32	42.04	37.86	23.30

CA/SC-CO_2 mixture, resulting in the decrease of pressure and viscosity. Furthermore, when the temperature rises from 45 °C to 55 °C, the inlet pressure reduces from 9.66 MPa to 6.77 MPa, decreasing by 30 %. Increasing the temperature promotes the volatilization of acetone and ethanol and the kinetic motion of SC-CO_2 dissolved in CA, and these small molecules enhance the free volume of the fluid. However, when the temperature rises from 55 °C to 65 °C (6.44 MPa), the decrement of the inlet pressure is 5%. In addition, the increasing temperature improves the kinetic motion of SC-CO_2, accelerating the dissolution of SC-CO_2. It means that the effect of temperature on the inlet pressure becomes weak when the temperature is high. Therefore, the temperature located in the range from 50°C to 55 °C is suitable for the extrusion of CA/SC-CO_2 mixture.

3.6 Effects of the solvent content

The shrinkage of gun propellant products is mainly determined by the solvent content. The high solvent content is conducive to the safety of extrusion processing, but the burning behaviors of gun propellants are unstable resulting from the residual of solvents. The ratio of the volume of mixed solvents to the mass of CA, c ($mL \cdot g^{-1}$), was applied to describe the effects of the solvent content on the flow behaviors.

Figure 9 and Table 4 point out the influence of solvent content on the flow behaviors of CA/SC-CO_2 mixture. When c is less than 1.20 $mL \cdot g^{-1}$, the increase of solvent content has remarkable effects on the inlet pressure and viscosity. In addition, it is interesting that the inlet pressure and viscosity have a great decrease when c is 1.40 $mL \cdot g^{-1}$. Too high solvent content weakens the strength of CA/SC-CO_2 mixture, and it makes some amount of CO_2 gas escape from the fluid. The escaped CO_2 gas leads to the unstable flow of CA/SC-CO_2 mixture, and it goes against to the quality of gun propellant products. Meanwhile, the low pressure in the die makes the product not compact when c is 1.40 $mL \cdot g^{-1}$. Consequently, 1.20 $mL \cdot g^{-1}$ is an appropriate solvent content for CA extrusion assisted with SC-CO_2.

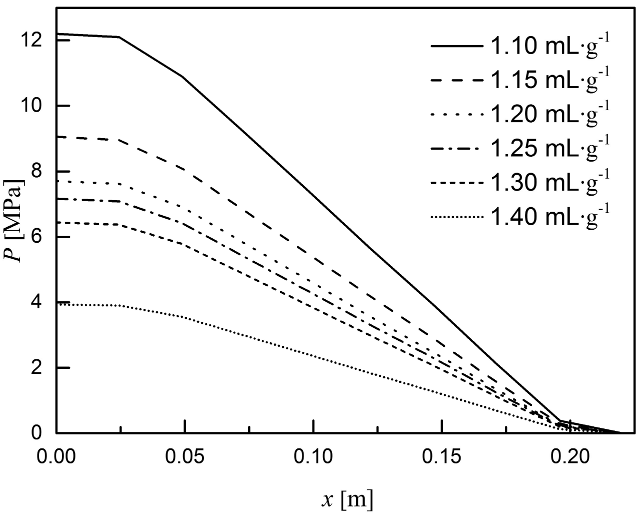

Figure 9 Effect of solvent content on pressure distribution. (T = 50 °C, L = 0.16 m, $V = 1.0 \times 10^{-7}$ $m^3 \cdot s^{-1}$)

4. Conclusions

This work simulated the flow behaviors of gun propellant substitute under various processing conditions in extrusion processing assisted with SC-CO_2 by Polyflow. The simulation results show that the pressure reaches the highest value at the inlet in the die and decreases along with the extrusion direction. With the injection of SC-CO_2, the viscosity distribution has a significant reduction. Increasing the length of the die raises the pressure of the inlet, but it has few effects on the viscosity distribution. The high processing temperature improves the flow of CA/SC-CO_2 mixture, and the inlet pressure reduces by about 30 %, when the temperature rises from 45 °C (9.66 MPa) to 55 °C (6.77 MPa). However, the effects of temperature become weak when the temperature is over than 55 °C. Increasing the solvent content reduces the pressure in the die and the viscosity of CA/SC-CO_2 mixture remarkably. Especially when c is 1.40 $mL \cdot g^{-1}$, the inlet pressure has a dramatic decrease resulting from the low structural strength of the fluid. The numerical simulation of CA/CO_2 mixture provides the fundamental and significant data for the extrusion of gun propellants assisted with SC-CO_2 safely in the future.

References

1) F. Gallant, H. Bruck, and S. Prickett, Propell. Explos. Pyrot., 31, 456–465 (2006).
2) J. Royer, Y. Gay, J. Desimone, and S. Khan, J. Pol. Sc.: Pol. Phys., 38, 3168–3180 (2000).
3) M. Iguchi, Y. Hiraga, K. Kasuya, and T. Aida, J. Supercrit. Fluids, 97, 63–73 (2015).
4) S. Rahimi, A. Peretz, and B. Natan, Propell. Explos. Pyrot., 32, 165–174 (2007).
5) B. Jyoti and S. Baek, Propell. Explos. Pyrotech., 39, 866–873 (2014).
6) Y. Ding and S. Ying, Cell. Chem. Technol., 50, 455–462 (2016).
7) T. Zhong, G. Rao, and J. Peng, Procedia Eng., 84, 920–926 (2014).
8) Y. Ding and S. Ying, Acta Armamentarii, 36 (8), 1437–1442 (2015).