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MODELING OF SEMI-SOLID BEHAVIOUR OF STEELS USING A MICROMECHANICAL APPROACH

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For non-isothermal steel semi solid processing when the dies are colder than the slug, an increase of solid fraction related to solidification due to thermal exchanges at the tool-slug interface is observed. Simulating steel semi-solid metal forming close to a real industrial forming is necessary to reduce potential run-time errors and costs. Simulating requires modeling of semi-solid behaviour. In this work a model based on micromechanical analysis with consideration the viscoplastic response along with an enhanced Herschel-Bulkley model has been proposed to predict the flow behavior of semi-solid M2 high speed steel. This model is introduced to overcome the limitations of the previously used models. The extracted model parameters from the steady state flow stress and step-change of shear rate experiments were then calculated and fitted to the experimental rheology results of continuous cooling. The model is capable of prediction of flow behavior and determination of rheological properties of semi-solid states especially for the tool steel systems in the wide range of liquid fraction.

Key words: rheology, step-change experiments, continuous cooling experiments, semi-solid steel, modelling.

1. INTRODUCTION

Semi-solid slurries are characterized by co-existence of solid and liquid phases in which spherical solid particles are suspended in a liquid matrix. The application of this technology for steels is still at the development stage due to high processing temperatures, narrow liquidus-solidus range, dies with resistance to thermal shock and temperature variations. Metal alloys in semisolid state exhibit distinctive rheological characteristics which is originated from their non-dendritic microstructures in the temperature interval between solidus and liquidus [1]. Rheological behavior of semisolid slurries is history, time, temperature and shear-rate dependent [2], which makes the study of rheological properties a challenging task. Modeling the complex flow behavior of semi-solid metals is critical to the development of die filling and successful die design for industrial processing and is difficult because semi-solid behavior is thixotropic and depends on the liquid-solid spatial distribution within the material. Atkinson [2] presented a summary of the modeling works on semi-solid processing of metallic alloys and categorized as one-phase or two-phase and as finite difference or finite element. The most reported models are capable of prediction the flow behavior in the narrow range of solid fraction. The enhancement of the available models contributes the successful simulation of the processes before the real industrial one. Various constitutive equations are used by different authors since discrepancies appear between experimental rheological data. Brown *et al.* [3–5] introduced a model based on internal variable framework. The internal variable is zero when the microstructure is fully broken and it approaches one when agglomerates form. This variable is then related to the shear stress of the material through constitutive model. This model was latterly altered by other researchers [6–8]. Recently Koeune *et al.* [9] have enhanced this internal variable frame work to overcome some limitations. The previous models could not degenerate properly to pure solid or liquid behavior nor to free solid suspensions. Modigell *et al.* [8] used Sn-Pb alloy to compare the results of experimental T-shape die filling with simulated ones. In their study, a non-isothermal two-phase model was developed and implemented into proprietary developed FEM software. Recently Pouyafar and Sadough [10] studied the flow behavior of M2 high speed steel and introduced an enhanced model based on Herschel-Bulkley model. Rouff *et al*. [11] present a novel and interesting approach. Spherical inclusions (*i.e.* particles) containing entrapped liquid are assumed to deform very little and can slip relative to each other if the restriction between them is released. They are surrounded by solid bonds and the 'not entrapped' liquid where deformation generally takes place. This 'active zone', associated with the strain localization, is gathered in a layer surrounding the inclusions. The volume solid fraction of the active zone is considered the internal variable. Favier *et al.* [12] presented an enhanced model [11] by accounting for the transient behaviour of semi-solids. This model, originally established in a viscoplastic framework, captures the transient response for shear rate jump and compression tests for solid fraction lower and higher than 0.5, respectively. The flow behavior simulated and compared with the model results qualitatively. In this work [12] a constitutive equation was proposed, accounting for the mechanical role of four phases within the material: the solid globules, the solid bonds, the entrapped liquid and the free liquid. Experimental evidence of the existence of solid interconnected skeleton was obtained by phase sensitive synchrotron of semisolid alloy structures (*e.g.* [13, 14]). Then Favier and Atkinson [15] proposed an enhanced model that is an extension from a previous model [12] to account for elastic-viscoplastic behavior.

Modeling requires experimental results in which the rheology and flow behavior of material in semisolid state are investigated. Measurement methods of rheological properties depend on the range of temperature, applied shear rate and the composition of material. Some works by Atkinson *et al.* [16] focus on thixotropy in Semisolid steel slurries under rapid compression. They studied die filling of two steel grades M2 and HP9/4/30. The results proved better die filling characteristic of M2 compared with HP9/4/30 tool steel. Two series of experiments were performed to evaluate the rheological behaviour of the material: steady state flow stress experiments to determine the equilibrium flow curves and step-change of shear rate experiments to determine the time dependent characteristics of the material.

In this work a new approach with combination of micromechanical modeling concept of the viscoplastic response and enhanced Hershel-Bulkley model [10] is proposed. This gives much more accurate prediction before the threshold of viscosity and after the formation of solid skeleton for a given shear rate in the continuous cooling process. The proposed model fitted well with the experiments in a wide range of shear rates and could predict the flow and viscosity of the M2 steel in semi-solid state. The experimental results from the previous work of the authors [17] will be used in this work for calculation of model parameters. The model was then implemented to the software and its performance to predict the flow behavior is investigated.

2. MATERIAL AND RHEOLOGICAL MEASURMENT

Material used for this study was M2 tool steel (from BÖHLER Uddeholm GmbH International). It was used in as rolled and annealed condition without any modification. In the continuous cooling experiments, the material cooled down with different cooling rate until the shear stress increased and the rotation is done hardly, while in iso-thermal experiments, the materials cooled down to the desire temperature and sheared under 65 1/s shear rate and shear rate jumps occurred to investigate the transient and steady state behavior of the material.

In this study a self-developed Searle-type rotational rheometer was designed and used for high temperature rheology tests [17]. The system was calibrated using silicon oils of known viscosity 9.81 Pa·s and 117.72 Pa·s. In the present work the modeling will be performed based on the results obtained from the experiments on M2 tool steel material. Two series of experiments was carried out: isothermal experiments and continuous cooling. The first series of experiments consist of isothermal step-change of shear rate experiments in which alloy under investigation is sheared with a constant shear rate for a specific time and then the shear rate is decreased or increased. The experiments were repeated for the low solid fractions of 10%, 25% and 37%. Continuous cooling behavior which describes the viscosity evolution during continuous cooling at constant cooling rate and shear rate gives the first insight into the effects of solid fraction, shear rate and cooling rate on rheological behavior. In particular, it is more relevant to the practical conditions set in semisolid processing techniques such as rheocasting and rheomoulding.

3. MODELING

Spherical and non-dendritic particles suspended in a liquid matrix prior to forming are necessary in the semisolid process. The required microstructures of M2 tool steel in semi-solid temperature achieved by partial remelting from as supplied material and taken after quenching. As mentioned previously, Semi-solid material involved of a two phase system composed of spheroidal solid particles of volume fraction $f^{\,s}$ suspended in a liquid matrix. A more careful observation especially on tomography of semisolid shows the presence of entrapped liquid within solid particles and presence of solid bonds between solid particles. This morphological pattern of semi-solid systems is schematically shown in [15]. It is commonly admitted that the deformation takes place in local sites such as the bonds between the solid grains and the liquid that is not entrapped in agglomerated solids [15]. In the present modeling the solid fraction in the active zone is considered as an internal parameter.

The Herschel-Bulkley model (Eq. 1) along with an internal variable framework was first used by Brown *et al*. [4] to investigate the thixotropic flow behavior semi-solid metal alloy slurries.

$$\tau = \tau_0 + k\dot{\gamma}^n. \tag{1}$$

The Herschel-Bulkley model cannot fully interpreted the experimental results. The differentiation of the above equation respect to shear rate in the steady state is zero that is far from reality. The experimental results in high shear rate shows that the thixotropic fluid tend toward an asymptotic and behaves like a Newtonian fluid. To consider the two cases mentioned, terms $k_1\dot{\gamma}$ and $\eta_\infty\dot{\gamma}$ as shown in Equation 2 has been added [10]:

$$\tau = [\tau_0 + \eta_\infty\dot{\gamma}] + [k_1\dot{\gamma} + k_2\dot{\gamma}^n], \tag{2}$$

where τ_0 is the yield stress, η_∞ is the viscosity at high shear rates, k_1, k_2 and *n* are material constants. Taking into account the above description and micromechanical modeling concepts based on Favier and Atkinson model [15, 18], the solid fraction of active zone ($f_A^{\,s}$) instead of λ is considered as internal parameter in the present modeling represents the thixotropic behavior of semisolid state.

$$\tau^C = [\tau_0 + \eta_\infty\dot{\gamma}] + [k_1\dot{\gamma} + k_2\dot{\gamma}^n]\; f_A^{\,s}. \tag{3}$$

A transient equation is used to describe the time dependent property of the structural parameter [12].

$$\frac{\mathrm{d}f_s^A}{\mathrm{d}t} = K_{ag}\, f_s (1 - f_s^A) - K_{dg}(1 - f^{\,s})\; f_s^A\, (\dot{\gamma})^n, \tag{4}$$

where K_{ag}, K_{dg}, n are the material parameters describing the agglomeration and disagglomeration mechanisms, respectively; $\dot{\gamma}$ is the overall shear rate given by $\dot{\gamma} = \sqrt{3}\dot{\varepsilon}$. For isothermal deformation and applying the strain rate, it is admitted that the kinetics of the agglomeration process are much slower than the kinetics of the disagglomeration process [19]. Consequently, we can neglect the agglomeration term.

Eq. 3 is used for modeling the suspended solid particles zone before the formation of the solid skeleton where the viscosity shows the extreme increase suddenly. The extreme variation of viscosity for M2 tool steel has been investigated in the previous work of the author [20]. This point was found from the experimental results. It can be assumed that at lower than the mentioned solid fractions, the solid skeleton is not formed. When the solid fraction increase (with decreasing temperature) solid skeleton is formed and suddenly at a narrow range of solid fraction (solid fraction threshold), shear stress increase rapidly. This threshold can be determined by continuous cooling experiments.

Substitution of $K_{ag}\; f^{\,s} = a$ and $(K_{dg})(1 - f^{\,s}) = b$ in the Eq. 3 the following equation is achieved

$$\tau^c = [\tau_0 + \eta_\infty\dot{\gamma}] + \left[\frac{k_1\, a\, \dot{\gamma}}{a + b\,(\dot{\gamma})^n} + \frac{k_2\, a\,\dot{\gamma}^n}{a + b\,(\dot{\gamma})^n}\right]. \tag{5}$$

Arranging the terms of $g = a(1 - f_s^A) - b\, f_s^A (\dot{\gamma})^n$, $f_s^A = \dfrac{a-g}{a + b\dot{\gamma}^n}$ and substitution in Eq. 5, finally the Eq. 6 is achieved,

$$\tau^c = [\tau_0 + \eta_\infty \dot{\gamma}] + [\frac{k_1 (a-g)\dot{\gamma}}{a + b\dot{\gamma}^n} + \frac{k_2 (a-g)\dot{\gamma}^n}{a + b\dot{\gamma}^n}]. \tag{6}$$

In an equilibrium state, the rate of agglomeration and dissagglomeration of the structure is the same. In this case $g = 0$ and accordingly the equilibrium flow stress equation become:

$$\tau_e^{\ c} = [\tau_0 + \eta_\infty \dot{\gamma}] + [\frac{a(k_1 \dot{\gamma} + k_2 \dot{\gamma}^n)}{a + b\dot{\gamma}^n}]. \tag{7}$$

Integrating the Eq. 7 and substituting it into the Eq. 2 will reveal a shear stress-time-shear rate equation:

$$\tau_T(t) = \tau_0 + \eta_\infty \dot{\gamma} + [k_1 \dot{\gamma}_T + k_2 \dot{\gamma}_T^n]\left\{ f_{s(eT)}^A - (f_{s(eT)}^A - f_{s0}^A)\exp[-(a + b\,\dot{\gamma}_T^n)\,t]]\right\}, \tag{8}$$

where f_{s0}^A is the initial state of structure and $f_{s(eT)}^A$ is the equilibrium value of structural parameter for a given shear rate which can be calculated as:

$$f_{s(eT)}^A = \frac{1}{1 + \frac{b}{a}\dot{\gamma}_T^n}. \tag{9}$$

Justified by the fact that the deformation is macroscopically homogeneous [15], at least at the beginning of the deformation, and both C and K branches as described above represent continuous phases point, the applied stress is dived between two branches [15, 18] with the following portion (Eq. 10)

$$\sigma = \psi\sigma^K + (1 - \psi)\sigma^C. \tag{10}$$

During deformation, solid skeleton is broken into free suspended particles which results in the change of solid skeleton fraction ψ as follows [15]:

$$\psi^{i+1} = -(1 - f_s) D_K \psi^i\, \dot{\bar{\varepsilon}}\ \bar{\varepsilon}, \tag{11}$$

where D_K is the constant value represent disagglomeration rate of the solid skeleton, $\bar{\varepsilon}$ is the equivalent strain and f^c is the threshold point of viscosity increase before that the ψ vale is to be zero. The initial value of ψ is assumed to be given by [15]:

$$\text{if } f_s < f^c,\ \psi_{init} = 0 \quad \text{and if } f^c \le f_s < 1,\ \psi_{init} = 1 - f^c / f_s. \tag{12}$$

It is assumed that the deformation behavior of K-zone (solid skeleton) compiles to a viscoplastic model and incompressible that obeys of a power-law type equation which can be written as Eq. 13:

$$\sigma^K = 2K(\sqrt{3}\,\dot{\bar{\varepsilon}})^{m-1}\,\dot{\varepsilon}^{vp_k}. \tag{13}$$

4. RESULTS AND DISCUSSION

As stated earlier, two series of experiments were performed to evaluate the flow behavior of semi-solid M2 steel: Isothermally step-change of shear rate and none isothermally continuous cooling experiments. The results of step change experiments are plotted in Fig. 2. Thixotropic shear thinning behavior can be observed for semi-solid M2 high speed steel in which the viscosity decrease in the course of time under a constant shear rate. In shear rate jump points, the viscosity increase suddenly as a result of iso-structure condition, but the viscosity decreases gradually as long as the shear applied constantly.

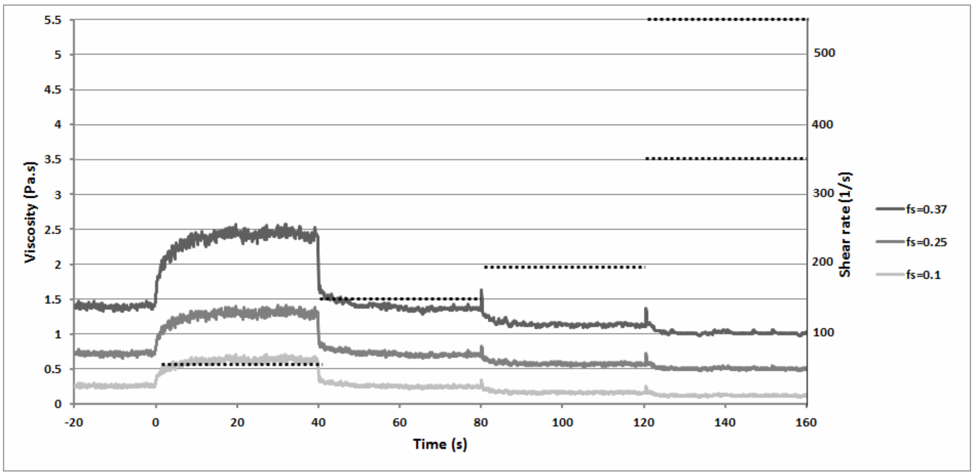


Fig. 2 – Step-change isothermal experiments for M2 tool steel; the shear rate was applied from 65 s^{-1} to 550 s^{-1}.

Table 1 shows the variation of viscosity in the shear rate increase points. For example with increasing the shear rate from 1.33 Pa·s to 1.57 Pa·s, the viscosity of semisolid metal increase in the solid fraction of 0.37. At this point the behavior of semisolid material is as same as the Newtonian fluids. In other words with increasing the shear rate the shear stress increases proportionally and this phenomenon is interpreted with Quaak's iso-structure theory [19]. With more increasing of the shear rate, the connected bonds of semisolid material decreases and active solid fraction tend to be zero. The reduction at the high shear rate is minor with respect to the first shear rate change up, since in first step a large number of the solid bonds are broken. The decreases of viscosities in next steps are only 0.25 Pa·s and 0.13 Pa·s respectively.

Table 1

The peak and steady state viscosity for step change up the shear rate from 65 s^{-1} to 550 s^{-1}

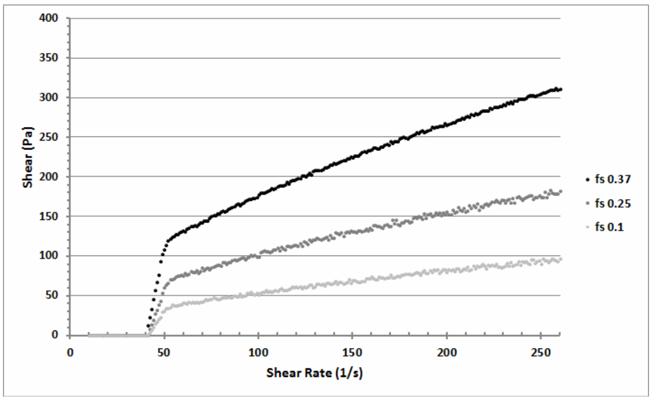
Shear Rate Range (s^{-1})	**Peak stress Viscosity, η_p (Pa·s)**			**Steady-State Viscosity, η_{ss} (Pa·s)**		
	Solid fraction			Solid fraction		
	37%	**25%**	**10%**	**37%**	**25%**	**10%**
65 to 150	2.47	1.32	0.67	1.33	0.75	0.25
150 to 195	1.57	0.82	0.7	1.15	0.56	0.16
195 to 350	1.32	0.68	0.26	1.02	0.53	0.12
350 to 550	1.21	0.64	0.19	0.95	0.47	0.07

In the modeling process, the model parameters are extracted from the transient experiments and the steady state of stress-shear rate diagram (Fig. 3a) and the provided model is adapted on the results of continuous cooling experiments. Fig. 3b shows the generalized equilibrium curve for the semisolid material that was observed in the step change experiments. The initial slope of shear stress-shear rate curve and the steady state shear rate in shear rate of zero are as follow:

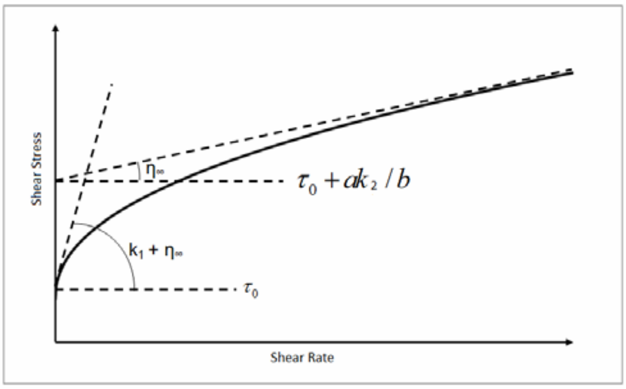
$$\left.\frac{\mathrm{d}\tau_e^{\,c}}{\mathrm{d}\dot{\gamma}}\right|_{\dot{\gamma}=0} = \eta_\infty + k_1 \;,\; \left.\tau_e\right|_{\dot{\gamma}=0} = \tau_0. \tag{14}$$

Considering $0 < n < 1$, the asymptotic of this curve is determined by taking limit tend to infinity

$$\lim_{\dot{\gamma}\to\infty} \tau_e = \left(\tau_0 + \frac{ak_2}{b}\right) + \eta_\infty \dot{\gamma}. \tag{15}$$



3(a)



3(b)

Fig. 3 – a) The flow equilibrium curve of M2 tool steel in the isothermal conditions;
b) generalized equilibrium flow curve.

The model parameters could be extracted with applying the above equation on the steady state curve and the theory of iso-structure in the point of shear rate jump in the transient state. The parameters are substituted in the Eq. 7. The diagrams of the models show a small discrepancy of maximum 7 Pa in shear stress with the experimental results. Therefor the achieved parameters are corrected with try and error method for more adaptability. The comparison between the results of model after correction and experiments is shown in Fig. 4. Figure 5 clearly indicates a shear thinning behaviour for M2 steel alloy for three different solid fractions with using the present model. As was previously observed by other researchers [21], in step-down experiments the time frame to reach an equilibrium value is larger due to the agglomeration process is naturally slower than dissagglomeration.

As explained before, the continuous cooling experiments shows the gradual variation of viscosity with different cooling rate and the constant shear rate. The model was then applied in the experimental results that are explained in details in [17]. By plotting the model and fitting to continuous cooling experimental results it can be observed that the proposed model is capable of prediction the flow behavior of semisolid material. Figure 6a shows the experimental and model results for cooling rate of 25°C/min and shear rate of 350 s^{-1}. The model is plotted and compared with experimental results for the C-zone before the formation of solid skeleton by using the extracted parameters from shear rate step change experiments. The parameters have been corrected again for more adaptability on the experimental curves. As explained earlier after the formation of solid skeleton, the viscosity increases dramatically. Therefor the Eq.16 should be used for describing the flow behavior of semisolid state of tool steel and an iteration FORTRAN code is written to extract the model results. Figure 6b shows the viscosity curve versus the solid fraction for the experimental and modeling results.

$$\sigma_{total} = \psi\sigma^{k} + (1-\psi)\sigma^{C} = \psi\, 2K(\sqrt{3}\,\dot{\bar{\varepsilon}})^{m-1}\,\dot{\varepsilon}^{\,vp} + (1-\psi)\,\sigma^{C}. \tag{16}$$

The value of D_k in the Eq. 11 is – 0.00147 for shear rate of 150 s^{-1}. In the fraction of the solid skeleton of 0.3 while the total stress is 471.66 Pa, inserted stress on the C-zone and K-zone is to be 309.70 and 161.96 respectively. Parameters associated with the M2 representation for continuous cooling process are listed in Table 2.

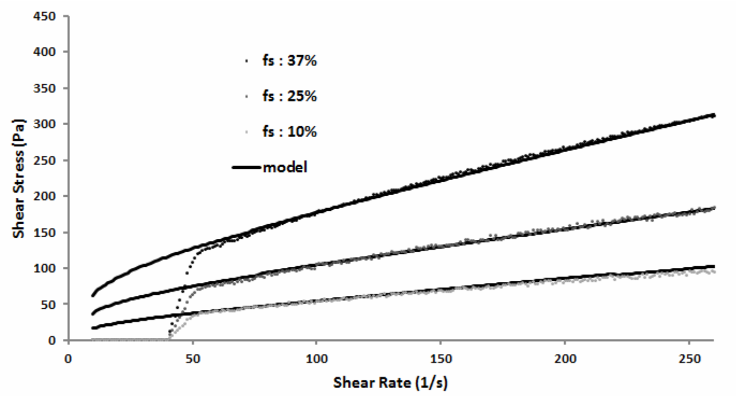


Fig. 4 – The comparison of the experimental results and the present model for solid fraction of 37%, 25% and 10% after the correction of parameters.

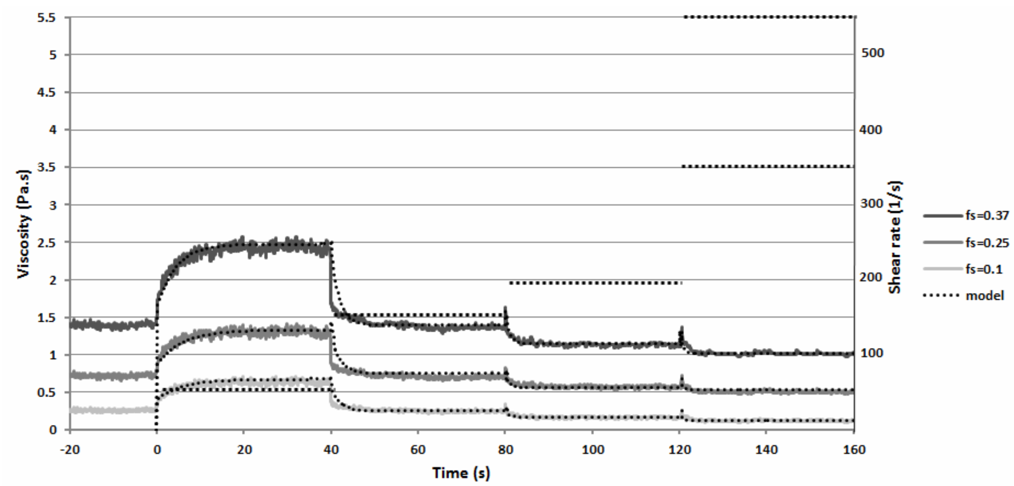
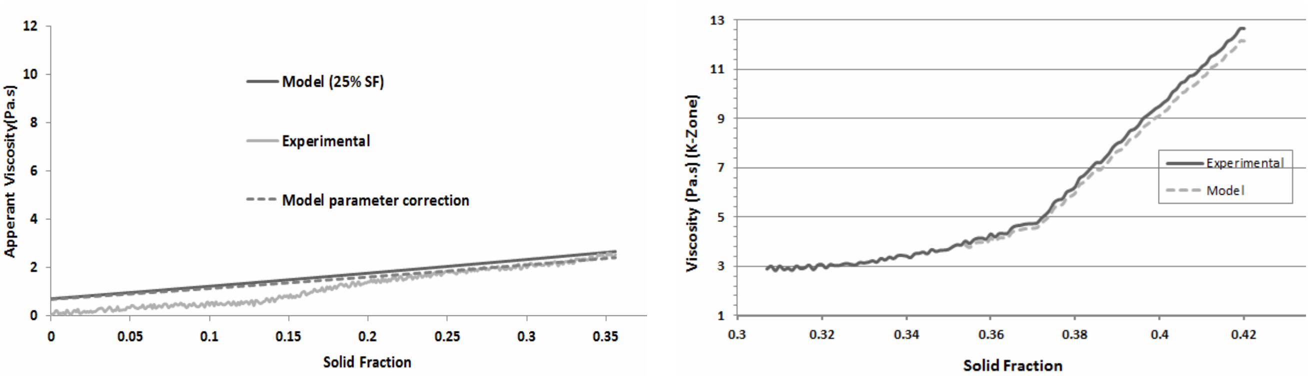


Fig. 5 – The model results in the shear rate step change up experiments.



(a) (b)

Fig. 6 – a) The plotted model comparison with experimental results for cooling rate of 25°C/min and shear rate of 350s^{-1}; b) the viscosity curve versus the solid fraction in the model after the formation of solid skeleton at shear rate of 150s^{-1}.

Table 2

The final model parameters in the continuous cooling and the shear rate of 150s^{-1}

τ_o (Pa)	η_∞ (Pa.s)	k_1	k_2	n	k_{ag}	k_{dg}	a	b	k	m
15	0.74	8.6	-0.41	0.74	1.70E-02	6.40E-04	5.18E-03	2.07E-04	96	0.25

5. CONCLUSION

A model based on micromechanical analysis with consideration the viscoplastic response along with an enhanced Herschel-Bulkley model has been proposed to predict the flow behavior and theological properties of semi-solid M2 high speed steel. Step-change experiment along with continuous cooling experiments was used to calculate the parameters of the model. The introduced model in this work is capable of prediction the flow behaviour in the wide range of solid fraction. Thixotropic shear thinning behavior can be observed for semi-solid M2 high speed steel in which the viscosity decrease in the course of time under a constant shear rate. In the shear rate jump points, the viscosity increase suddenly as a result of iso-structure condition, but the viscosity decreases gradually as long as the shear applied constantly. The reduction of viscosities at the high shear rate is minor with respect to the first shear rate change up.

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