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Rheology of liquid metals and alloys

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ABSTRACT

The flow behavior and viscosity of liquid Zn, Sn, Cd, Bi-42 wt%Sn, Zn-7 wt%Al, and Sn-3 wt%Ag-0.5 wt%Cu were characterized and quantified with rotational rheometry experiments. Evidences from this study shows these liquid systems uniquely exhibit a shear thinning and time-independent (non-thixotropic) flow behavior in all the evaluated shear rate regimes. We have attempted to offer a physical explanation from prior-art for the observed unique flow behavior of the liquid metal systems. The strong short range atomic order in these metals significantly contribute to their flow behavior and at any shear rate the viscosity obeys the standard Arrhenius energy equation for temperature dependence.

1. Introduction

Quantified flow behavior of liquid metals and metallic alloys is imperative to many scientists who are trying to understand and simulate processes such as casting, welding, galvanizing and thermal coating. Extensive experiments and physical modeling efforts have been dedicated to evaluate viscosity of liquid metals and metallic alloys during the second half of the last century [1,2]. However, these efforts are still continuing because of a lack of valid physical or empirical model to quantify the flow behavior. An underlying universal assumption in most past experimental efforts had been that the liquid metal systems exhibit a Newtonian flow behavior, which assumes that at any given temperature there exists a linear dependency of the applied shear stress to the shear rate experienced by the liquid and the slope of this dependency would represent the constant shear viscosity [3].

An underlying universal assumption in most of the past experimental efforts (Oscillating Vessel Viscometer and Co-axial Cylinder Rheometry) [4–10] had been that the liquid metal systems exhibit a Newtonian flow behavior [3]. However, recent efforts [11–14] have suggested that the liquid metal systems show tendencies of non-Newtonian flow behavior. Sun et al. [15] and Way et al. [16], evaluated the viscosity of liquid Sn and Bi [15], and metallic glass above the re-crystallization temperature [16], respectively, using rotational rheometry with a coaxial cylinder measuring geometry, to suggest that these systems are non-Newtonian shear thinning systems. However, the measuring geometry used by them containing the liquid metal and such a system would have a range of shear between the stagnant liquid at the stator wall and maximum angular velocity (shear rate) at the rotor wall of the measuring geometry. Hence, these results, though present qualitative evidence that liquid metals may be non-Newtonian and shear thinning in flow behavior, one could not derive quantitative flow behavior from these results. A unique value of shear rate would have to be maintained in the entire sheared liquid to ascertain the validity of the Newtonian flow behavior in liquids and this could only be possible with few measuring geometries such as the cone and plate and double concentric cylinder (DCC) [17]. In the previous article [17] we have explained the difficulties in the measurement of the liquid metal viscosity and the geometry which is more suitable to use in this measurements. Further, in Malik et al. [17], it was shown that the liquid metals may exhibit a non-Newtonian and shear thinning flow behavior. The flow behavior and viscosity of the various liquid metals and alloys have been quantified for a wide range of shear rate regime and a physical expression for the flow behavior of the liquid metals has been hypothesized in this publication.

2. Materials, apparatus and procedure

An AR-2000[1]-rheometer was used to evaluate the liquid study and the schematic of the experimental arrangement and the geometry details are shown in Fig. 1. Rotational rheometry experiments

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[1] Advanced Rheometer, TA Instruments, New Castle, DE, USA http://www.tainstruments.com.

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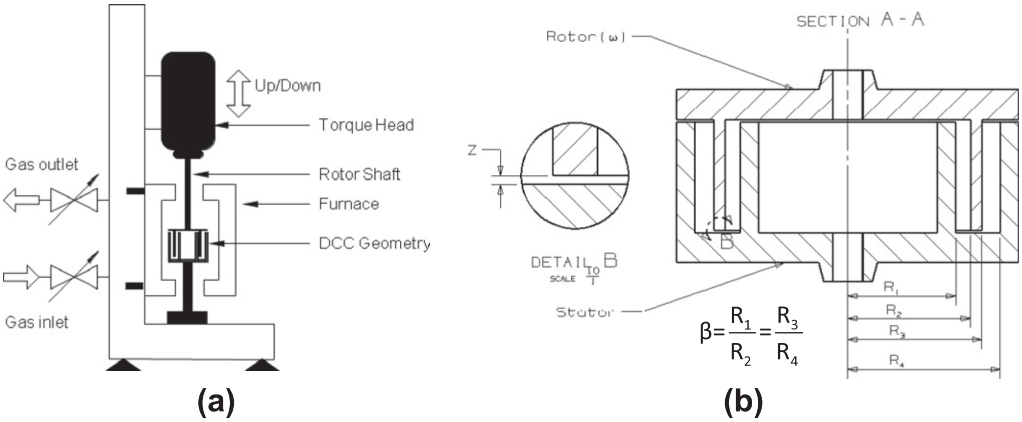

Fig. 1. Schematic of experiment apparatus. (a) Rotational rheometer (b) cross-section of double concentric cylinder (DCC) measurement geometry used in the present experiment study. R_1 = 14.5 mm, R_2 = 16.5 mm, R_3 = 18 mm, R_4 = 20.5 mm, H = 15 mm and, z = 0.30 mm. T is the torque experienced by rotor.

with the double cocentric cylinder (DCC) containment geometry [17] enclosed in a high temperature environment controlled furnace was used to evaluate the flow behavior of three pure metal liquids; Zn (99.99% purity), Sn (99.8% purity) and Cd (99.9% purity); two binary metallic alloys (Bi-42 wt%Sn (commercial alloy purity) and Zn-7 wt%Al (99.999% pure Al was added to 99.99% pure Zn)) and one ternary metallic alloy (Sn-3 wt%Ag-0.5 wt%Cu (commercial alloy purity)), which is a popular lead free soldering alloy. Ultra high purity Argon gas was continuously purged into the furnace chamber at a flow rate of about 10 L min^{-1} to minimize oxidation of the liquid and the temperature within the furnace was maintained at an accuracy of ±0.5 K. The measuring geometry and rheometer has been elaborately described in Malik et al. [17] and were critically calibrated with standard liquids such as S2000 [18], S60 [18] and silicone [19] and verified with de-ionized water. Several rotational rheometry experiments were carried out for each metal system as shown in Table 1. The four experiment temperatures of each metal system are shown along with the two types of rotational rheometry experiments, namely the ramp-up and ramp down cycles, and the shear rate peak hold experiments. Liquid Zn has been used as an example system to show the rheometry experiment conditions for the two types of experiment in Table 1, and except the temperature, these conditions were identical for the other five liquid metals and alloys in this study, as well. Each liquid metal system at a specific temperature above the respective liquidus temperature was rotated in three different shear rate conditions: (i) a ramp up of shear rate from 0 to 300 s^{-1}, (ii) a ramp down from 300 to 0 s^{-1}, and (iii) 31 peak hold experiments at various specific shear rates between 1 and 300 s^{-1} for a period of 10 s each. The ramp up–ramp down cycles of shear rate were carried out for five total cycle time periods of 60 s once, 120 s twice, and 180 s twice, respectively wherein one data point was collected every second. The ramp-up and ramp-down cycles at various cycle time intervals enabled a thorough study of the thixotropic behavior of the sheared liquid. A total of 41 rotational rheometry experiment conditions were carried out and repeated three times, totaling 123 experiment conditions for each

Table 1
Experiment conditions of rotation rheometry for each metal and alloy system showing the four respective experiment temperatures and the rotational experiment conditions for each temperature for each metal system using the ramp-up and ramp-down cycles, and peak hold cycles using Zn as an example system at one temperature.

Material	Liquidus temperature (K)	Experiment temperatures (K)			
Experiment temperatures					
Cd	594	598, 623, 648, 673			
Sn	505	510, 535, 560, 585			
Zn	693	698, 723, 748, 773			
Bi-42Sn	411	423, 448, 473, 498			
Zn-7Al	663	668, 693, 718, 743			
Sn-3Ag-0.5Cu	493	498, 523, 548, 573			
Material	Temperature (K)	Shear rate range (s^{-1})	Total time of cycle (s)	Rate of data acquisition (s^{-1})	Repetitions
Ramp up and ramp down cycle experiments (Zn as example)					
Zn	748	0–300	60 120 120 180 180	1	3
		300–0	60 120 120 180 180		
Material	Temperature (K)	Shear rate (s^{-1})	Time (s) at each shear rate hold	Rate of data acquisition (s^{-1})	Repetitions
Peak hold experiment (Zn as example)					
Zn	748	0.1–300 in 31 peak hold steps	10	2	3