

ON A NATURE OF DYNAMIC VISCOSITY IN LIQUID METALS

A.B. Ibrahimli^{1,2*}

¹Azerbaijan State Pedagogical University, Baku, Azerbaijan

²Institute for Space Research of Natural Resources, Azerbaijan National Aerospace Agency, Baku, Azerbaijan

Abstract. According to electron theory of metals at melting process character of inter atomic (ion) bond doesn't change, but relative change of special volume makes $\leq 10\%$. For alkali metals the final change is especially little $\sim 2.5\%$. Besides, it was experimentally determined, that at the result of melting process only 20-25% of inter atomic bond is broken, that is to say in liquid and crystalline metals coordinative numbers are practically similar. The described structure of liquid metal has dynamic character and this allows accepting liquids as a correlated system, which consist of positive charge ions and free electrons. It is known, that free electrons play significant role in the formation of viscosity of liquid metals. Confirming the potential distribution along with the whole volume in liquid metals according to Thomas-Fermi pseudo potential, it was shown, that viscosity is defined $\eta = AT^{-2} - D$ law (A and D are value, which don't depend on temperature). On the example of alkali liquid metals satisfactory matching of this phenomenological position was determined at melting temperature till 1500K by corresponding experimental results (with $\leq 4\%$ error).

Keywords: liquid metals, Thomas-Fermi pseudo potential, viscosity.

Corresponding Author: Azerbaijan State Pedagogical University, AZ-1000, Baku, Azerbaijan; Institute for Space Research of Natural Resources, Azerbaijan National Aerospace Agency, AZ-1106, Baku, Azerbaijan, fizik-3@mail.ru

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1. Introduction

The synthesis of new materials with metallic and semi-metallic properties and the study of their various electrophysical properties is very important for modern electronics and spintronics (Rivera-Salinas *et al.*, 2020; Burkov, 2016; Demir *et al.*, 2019; Adamiak *et al.*, 2018). It is known that metal atoms allow to change different properties of compounds. The substitutions performed at low concentrations also cause fundamental changes in physical properties (Trukhanov *et al.*, 2017; Khan *et al.*, 2019; Dang *et al.*, 2016; Trukhanov *et al.*, 2016a; Dang *et al.*, 2018; Trukhanov *et al.*, 2016b). Therefore, it is very important to study the properties of metals in the different forms.

In the physics of condensed matter from scientific and practical point of view one of the important problems is a viscosity phenomena, which takes place in liquids. There are many theoretical and experimental researches, unique model or theory haven't been obtained for physical mechanism of viscosity yet (Belashchenko, 2013; De Bur, 1986; Dric & Zusman, 1998; Zajman, 1972; Kikoin, 1976). The main difficulty in the explanation of viscosity is that liquid has dynamic structure and static order. Depending on physical condition and nature of liquid particles, as in macroscopic volume the dynamic structure, and in microscopic volumes the close order change considerably, it is impossible from molecular point of view to describe change of viscosity at wide values diapason of state parameters (Belashchenko, 2013).

The above-mentioned shows that any phenomenological approach or half empiric model for all liquids at wide change interval of state parameters are very important from scientific and practical point of view. In the present research work by the help of phenomenological approach we have considered the half empiric method, which allows investigating viscosity at a wide temperature range. The main aim of the proposed approach is to use the sample liquid as Newton liquid, in other words the usefulness of Newton equation for viscosity and selection of interaction potential between liquid particles.

The given conditions correspond exactly to liquid metals. According to electron theory of metals at melting process character of inter atomic (ion) bond doesn't change, but relative change of special volume makes $\leq 10\%$. For alkali metals the final change is especially little $\sim 2.5\%$ (Prigozhin & Kondepudi, 2009). Besides, it was experimentally determined, that at the result of melting process only 20 + 25% of inter atomic bond is broken, that is to say in liquid and crystalline metals coordinative numbers are practically similar (Ubbelohde, 1965). The described structure of liquid metal has dynamic character and this allows accepting liquids as a correlated system, which consist of positive charge ions and free electrons. It is known, that free electrons play significant role in the formation of viscosity of liquid metals. So let's remember some moments concerning free electron theory of metals.

2. General approach

According to Tomas-Fermi theory in metal along with all volume the potential energy and density of electrons are distributed non-homogeneously, namely density and impulse of an electron depend on coordinate (Zajman, 1972):

$$n(r) = \frac{8\pi}{3h^3} [P_F(r)]^3 \quad (1)$$

where, r – radii-vector of electron, h – Plank constant. As it is seen density of electron depends on distance in a unique volume and is determined by corresponding change of impulse.

Change of electron density is formed at the result of phase non-homogeneously of potential. Existence of positive ion causes the change of charge distribution in electron gas. Electrons are attracted by positive ions and form local electron cloud around ions. According to potential theory the area around positive ion is like the modified Coulomb field and when distance enlarges it is reduced due to exponential law. From physical point of view the latter expresses that interaction between positive ions is screened by free electrons.

It is shown in Thomas-Fermi theory that phase distribution of free electron density around positive ion is given by the following equation:

$$n(r) = n_0 \left(1 + \frac{3eq}{2E_F} \cdot e^{-\frac{r}{\lambda}} \right) \quad (2)$$

In (2) E_F is 0 K - Fermi energy, q – electric charge of ion, n_0 – number of electrons in a unique volume in non-excited part of metal, e – electron charge.

As it is seen from (2) $r \gg \lambda$ at distances the influence of ion on electron density is less. It is natural that this has similar power with screening of ion effect. That's why λ characterizes screening effectiveness of ion effects with electrons, and is called Thomas-Fermi, or screening radii. In Thomas-Fermi theory (λ_{T-F}) is determined by following equation:

$$\lambda_{T-F} = \sqrt{\frac{\pi\hbar^2}{4m^*e^2K_F}} = \sqrt{\frac{E_F}{6\pi\cdot e^2n_0}} = \sqrt{\frac{\hbar^2}{4m^*e^2n_0^{1/3}}} \quad (3)$$

where, m^* -effective mass of electron; K_F – Fermi wave vector. For many metals λ_{T-F} is in several angstroms.

In metals as E_F and n_0 practically don't depend on temperature, the screening radii will not depend on temperature. It can be expected that due to electron-electron and ion-ion interactions addition, which Thomas-Fermi screening gave to viscosity, will not depend on temperature and according to (3) when concentration of free electrons increase, the part of viscosity, which is connected with screening, must diminish.

From the other side when concentration of free electrons increase, as screening length is reduced on $\lambda_{T-F} \sim n^{-2/3}$ law, by the rise of screening length the interaction between ions will weaken and inter ionic effective gravity will strengthen. This is equivalent to high viscosity. Viscosity of free electron gas in liquid metals was considered by us in (Eyvazov, 2012).

Using (3) it is possible to evaluate λ_{T-F} . In this law while calculating λ_{T-F} it must be considered, that Coulomb interaction in electron gas changes effective mass in alkali metals of electron till ~25% (Zajman, 1972). Values and some characteristic parameters of screening length of screening length, calculated for different alkali metals, were given in table 1.

In alkali metals as bond energy is reduced from Li to Cs (see Table 1) it can be expected that viscosity of liquids must decrease, that is $\eta_{Li} > \eta_{Na} > \eta_K > \eta_{Rb} > \eta_{Cs}$ (it is supposed that in melting process character and relative intensiveness of a bond don't change, it is like in solid phase).

Viscosities of different alkali metals are in corresponding proportion among themselves with inequalities.

Supposing the formation of viscosity of liquids due to inter molecular attraction let's consider the phenomenological approach, which has general character for Newton liquids. With this aim let's accept that viscosity (η) was given by Newton law. Then at stationary rate gradient $\left(\frac{\partial\vartheta}{\partial r} = const\right)$ field the internal friction force which influences on dS section of neighboring liquid layers, will be:

$$F_s = \eta \cdot \left(\frac{\partial\vartheta}{\partial r}\right) dS, \quad (4)$$

where η is a dynamic viscosity coefficient.

The viscosity according to equation (4) is as follows

$$\eta = F_s \left[\left(\frac{\partial\vartheta}{\partial r}\right) \cdot dS\right]^{-1}. \quad (5)$$

As it is seen at a unique rate gradient field $\left(\frac{\partial\vartheta}{\partial r} = 1\right)$ dynamic viscosity is a friction force, which influences on unique field ($dS = 1$) taken perpendicular to gradient. It is natural that this force is a cohesive force, which influences between two liquid layers near to the taken dS field (on opposite sides of the field). During laminar flow as $\left(\frac{\partial\vartheta}{\partial r}\right) = const$ it will be $\left[\left(\frac{\partial\vartheta}{\partial r}\right) \cdot dS\right]^{-1} = const = B$. From (5) we will get:

$$\eta = B \cdot F_s \quad (6)$$

So, at laminar flow dynamic viscosity of a liquid is in direct proportion with inter layer cohesive force, namely with molecular gravity. It is possible to calculate viscosity coefficient, when analytical equation of attraction is known. This forms the principle of phenomenological approach in the present research work. In this approach as viscosity is

determined by inter molecular interaction, this is not activation process and impulse between layers is carried by vibration of liquid particles.

Phenomenological expression (6) can be applied to all liquids. But F_s equation is unknown in its form for several types of liquids. Let's determine the viscosity for liquid metal.

3. Viscosity of pseudo potential liquids

It is known that in metallic crystals on electron on electrostatic type potential field formed by positive ions, the motion of free electrons is characterized by the following pseudo potential

$$\varphi = \frac{A_1}{r} \cdot e^{-B_1 r}, \quad (7)$$

where A_1 is a constant, depending on its nature (especially electrical charge), $B_1 = \lambda^{-1}$. According to Thomas-Fermi theory in metals screening radii of free electrons λ_{TF} practically doesn't depend on temperature (see (3)) and is defined by concentration of free electrons and Fermi energy of metal corresponding to 0 K of metal.

When $B_1 r < 1$ in (7) considering $e^{-B_1 r} \approx 1 - B_1 r + \frac{(B_1 r)^2}{2}$ we get

$$\varphi(r) = \frac{A_1}{r} - A_1 B_1 + \frac{A_1 B_1^2}{2} r. \quad (8)$$

From the other side since $F = -\text{grad } \varphi$ inter layer effect will be as follows

$$F = -\frac{d\varphi(r)}{dr} = \frac{A_1}{r^2} - \frac{A_1 B_1^2}{2} = \frac{A_1}{r^2} - D_1. \quad (9)$$

$D_1 = A_1 B_1^2 / 2$ is constant, which doesn't depend on temperature. Considering expression (9) in (6) we have

$$\eta = B \cdot F = \frac{B A_1}{r^2} - B D_1 = \frac{A_2}{r^2} - D_2. \quad (10)$$

In (10) $A_2 = B A_1$ and $D_2 = B D_1$. Viscosity of liquid metals can be taken as total of two limits:

$$\eta = \frac{A_2}{r^2} - D_2 = \eta_1 + \eta_{//}. \quad (11)$$

It can be supposed, that the first $\eta = \frac{A_2}{r^2}$ limit, which is in inverse proportion with squared distance of positive ion, is a viscosity, formed due to gravity. As the second limit $\eta_{//} = -D_2 = -B D_1$ is negative and doesn't depend on inter ionic distance, it points to suppose that the limit has connection with free electron gas.

In phenomenological analysis the result, obtained for viscosity of liquid metals and expressed by (10), is expectable. As liquid metal consists of ionic and electron down systems it is obvious, that in the formation of physical events these down systems will participate. We think that in (11) the first limit $-\eta_1$ inter down systems interaction (ion-free electron), but the second limit $-\eta_{//}$ is connected with electron-electron interaction. As positive charge ions are screened by free electrons, ion-ion interaction won't play decisive role in the formation of viscosity.

As widening coefficient of liquids is similar with widening coefficient of corresponding solid phase, it can be supposed that temperature dependence of inter atomic distance in liquids is given by linear law, that is $r \sim T$. In the melting process volume of metal increases ~2-5% (see table 1). Then temperature dependence of viscosity of liquid metal from (10) will be:

$$\eta(T) \approx \frac{A}{T^2} - D \quad (12)$$

Table 1. Different physical parameters for Li, Na, K, Rb, Cs liquids

Metal	Cell period, 10^{-9} m (Dang et al., 2018)	$\left(\frac{\Delta V}{V}\right) \cdot 10^3$ (Dang et al., 2018)	T_{melt} , K (Dang et al., 2018)	Coord number (liquid), (Belashchenko, 1989; De Bur, 1986)	$\left(\frac{m_e^*}{m_0}\right)$ (Trukhanov et al., 2016a)	E_{bond} · 10^{19} , C (Dang et al., 2018)	$n \cdot 10^{22}$, sm^{-3} (Dang et al., 2016)	$\lambda_{T-F} \cdot 10^{11}$, m (Trukhanov et al., 2016b)
Li	0,3502	16,5	453,7	9,5-9,8	1,19	2,26	4,7	6,07
Na	0,4250	25,0	371	7-8	1,03	1,81	2,65	6,68
K	0,5247	25,5	336,4	9-10	0,99	1,52	1,40	7,43
Rb	0,6231	25,0	312,7	9,5	0,97	1,42	1,15	7,68
Cs	0,6874	26,0	301,6	8-9	0,98	1,29	0,91	7,99

(12) is a general equation of temperature dependence of liquid metal viscosity from suggested phenomenological point of view. As it is seen in this case viscosity has connection wet ion-electron down systems interaction and due to square regularity consists of diminishing limit and the limit, which doesn't depend on temperature. As (12) regularity doesn't depend on type of liquid metal, it must have universal character. Basing to the above-mentioned let's determine the equation for temperature dependency of viscosity on samples of Li, Na and K liquids. As (12) equation has universal character, it can be applied to other liquid metals. In (12) equation if A and D numbers are defined, at any temperature the value of viscosity can be calculated. With this aim use half phenomenological method. Supposing at two arbitrary T_i and T_j temperatures the viscosity is η_i and η_j (it is supposed that $T_j > T_i$). Then after simple calculation we will get:

$$A = \frac{T_i^2 \cdot T_j^2}{T_j^2 - T_i^2} (\eta_i - \eta_j) = \frac{T_i^2 \cdot T_j^2}{T_j^2 - T_i^2} \cdot \Delta\eta_{ij},$$

$$D = \frac{T_i^2}{T_j^2 - T_i^2} (\eta_i - \eta_j) - \eta_i = \frac{T_j^2}{T_j^2 - T_i^2} \cdot \Delta\eta_{ij} - \eta_i. \quad (13)$$

In (13) $\Delta\eta_{ij} = (\eta_j - \eta_i)$ is a viscosity difference of liquid at temperatures suitable for corresponding index. Considering (13) and (12) temperature dependence of liquid viscosity is defined by following equation:

$$\eta(T) = \left[\frac{T_i^2 \cdot T_j^2}{T_j^2 - T_i^2} \cdot \Delta\eta_{ij} \right] \cdot \frac{1}{T^2} - \left[\frac{T_j^2}{T_j^2 - T_i^2} \cdot \Delta\eta_{ij} - \eta_i \right]. \quad (14)$$

When viscosity of liquid metal at two arbitrary T_i and T_j temperature is known, it is possible to calculate the viscosity and to define the form of dependency for liquid metal at a wide temperature range. According to two arbitrary experimental values of viscosity, taken from (12), the calculated values of A and D by (13) for Li, Na and K liquids were given in the Table 2.

Table 2. Experimental values of viscosity, the calculated values of A and D for Li, Na and K liquids

Liquid metal	A, $\frac{K^2 \cdot kq}{m \cdot sec}$	D · 10 ⁶ , $\frac{kq}{m \cdot sec}$	Temp. interval ΔT, K
Li	95,84	-146,8	500 ≤ T ≤ 1500
Na	81,69	-97,1	400 ≤ T ≤ 1500
K	52,7	-83	400 ≤ T ≤ 1500
Pb	621	-638	720 ≤ T ≤ 1100
Sn		-560	510 ≤ T ≤ 1100
Zn		-275	720 ≤ T ≤ 970
Hg		-742	253 ≤ T ≤ 573

As it is seen from the table both two parameters are reduced by module on Li, Na and K liquids. This is a decrease of viscosity and corresponds to $\eta(\text{Li}) > \eta(\text{Na}) > \eta(\text{K})$ inequality, which we have determined for viscosity of alkali values at $T \leq 1200$ K temperatures (see Table 3).

Table 3. η_c , η_{TF} , and η_{TF}/η_c at high temperature

T, K	500	600	700	800	900	1000	1100	1200	1300	1400	1500
$\eta_c \cdot 10^6$, $kq/(m \cdot san)^{-1}$ (Belashchenko, 1989)	531	426	358	310	275	247	225	207	192	180	163
$\eta_{TF} \cdot 10^6$, $kq/(m \cdot san)^{-1}$	530, 2	413	342, 4	296, 6	261, 1	242, 6	226	213, 3	203, 5	195, 7	189, 4
η_{TF}/η_c	0,99 8	0,97 6	0,95 6	0,95 8	0,96 4	0,98 4	1,00 4	1,03 0	1,05 9	1,08 8	1,11 8

Calculated (η_{TF}) and experimental (η_c) values of liquid Li viscosity by Thomas-Fermi potential η_c and η_{TF} are experimental and calculated values for Li liquid.

4. Conclusion

As it is seen from figure and table at temperatures higher than boiling temperatures some differences are observed between calculated (12) and experimental values of viscosity. Probably, this has connection with non-linear changer of inter ionic distance by temperature and difference of potential from pseudo potential. The present research work shows that viscosity of pseudo potential liquid metals is formed at the result of ion-electron and electron-electron interaction. The limit of viscosity $\eta \sim T^{-2}$, the second (electron-electron) limit changes by $\eta = \eta(T) = \text{const}$ regularity. In the work we have determined the half empiric analytical equation, which allows calculating viscosity at wide temperature range according to experimental value of liquid metal viscosity at two arbitrary temperatures. It was shown that calculated and experimental values of viscosity of Li, Na, K liquid metals match with small error at $T_{\text{melt}} \leq T \leq 1500$ K temperature range.

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