Study on the Viscosity of Molten Iron*

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The viscosity of Fe-C-Si alloy (3.5%C, 1.20%Si) was measured by the oscillating crucible method to investigate the presence of hysteresis and the difference in viscosity between white and grey iron melts of the same chemical composition, and to clarify a problem on the heredity of cast iron whose properties after melting depend upon charge materials. The results obtained are summarized as follows: (1) Viscosity curves measured in the superheating process did not correspond with those in the cooling process over the temperature range of 1255° to 1480°C; a negative hysteresis was recognized as proposed by Turovskii. The higher the maximum heating temperature, the wider became the hysteresis. (2) In this temperature range the viscosity of grey cast iron was higher than that of white cast iron. (3) In case of holding the melt at 1342° and 1288°C, the change in viscosity coefficient with time decreased rapidly by 20 minutes of holding. Subsequently, the difference between the white iron and grey cast iron melts was clearly recognized, where the viscosity of the latter was higher than that of the former. (4) With increasing Si content up to 0.43 %, the hysteresis was hardly recognized, but over 1.22% Si there appeared the negative hysteresis. In both superheating and cooling processes the viscosity showed a maximum at 1.22 %Si and a minimum at 0.43 and 2.70 %Si, respectively.

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I. Introduction

The viscosity of molten iron and steel has been thought to be one of the most important factors in the practical operation of each process of refining, ingot-making and casting. However, the data are scarcely available except for the melt of a very simple composition because of the extreme difficulty of measurements, and also the measured values differs greatly by investigators^{(1)~(5)}.

In view of such circumstances, increasing attention has recently been directed to problems on the viscosity of molten iron, steel and slag.

The present authors have considered that the viscosity is an important factor to make clear the problems on the formation, growth and floatation of primary deoxidation products, the formation of the remaining secondary deoxidation products which induce huge inclusions in the negative segregating zone of steel ingot, and the behaviour of graphite in cast iron. Therefore, a series of measurements have been carried out with a measuring apparatus devised on the basis of the oscillating crucible method. This paper deals with interesting results of the measurements of the viscosity of cast iron.

Turovskii and Lyodimov⁽⁶⁾⁽⁷⁾ experimented the viscosity and electric resistance of molten Fe-C alloy

and the influence of silicon on them, and pointed out that the viscosity of molten iron varies between the superheating and cooling processes, showing a hysteresis phenomenon. And they attributed the cause to the characteristic behavior of the viscosity of heterogeneous alloy in which microgroups increase in number with the dissolution of graphite particles into molten iron.

The authors confirmed experimentally the presence of such a hysteresis phenomenon as well as the difference in viscosity between white cast iron and grey cast iron melts, as proposed by Turovskii et al.

II. Experimental procedure

1. Samples

Three grades of samples, Fe-C, Fe-C-Si and Fe-C-Si-Al alloys, were first melted in the induction furnace using electrolytic iron, high-purity graphite, metallic silicon and aluminum. Then a half of the melt was cast into a metal mould of 3.5 mm thickness to get white iron samples of 50 g and the remaining half of the melt was cast into a sand mould of 50 mm × 80 mm preheated at 1000°C. They were then furnace cooled from 1000°C to prepare grey iron samples of 50 g in which coarse flaky graphite was precipitated.

2. Experimental method

Measurements of the viscosity were made with the apparatus devised by the authors on the basis of the crucible oscillating method⁽⁸⁾. A sample of 350 g was heated at a rate of 450°C/hr up to 1000°C, and then at a rate of 900°C/hr until a given temperature is reached. After reaching the temperature, the sample was held for 20 minutes, and its viscosity was measured four times after torsional oscillation. The average value thus obtained was taken as the viscosity

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coefficient at each given temperature.

3. Experimental groups

Measurements were made on five groups of samples as shown in Table 1.

in the superheating process and the difference is clearly recognized between white and grey cast irons even if they have the same chemical composition. Especially, the difference is greater between the processes of superheating to and cooling from 1388°C. The viscosity

Table 1 Chemical compositions of specimens (%).

Sample No.	Grade	Fracture	TC	Si	Al	Al*	Temperature range	Measurement
V-10 V-12	Difference between grey iron and white iron in Fe-C-Si alloy	White	3.54	1.22	0.212	0.425	1280°~1460°C	Heating-cooling
		Grey	3.54	1.22	0.212	0.425		
V-13 V-14		Grey	3.54	1.22	0.212	0.243	1255°~1388°C	Heating-cooling-heating
		White	3.54	1.22	0.212	0.243		
V-18 V-19		Grey	3.40	1.19	0.202	0.287	1248°~1343°C	Heating-cooling
		White	3.40	1.19	0.202	0.287		
V-16 V-17	Influence of holding time in Fe-C-Si alloy	White	3.40	1.19	0.202	0.266	1255°~1388°C	Heating-cooling after holding for 2 hrs at 1288°C
		Grey	3.40	1.19	0.202	0.256		
V-29 V-30		White	3.59	1.12	0.230	0.290	1342°C	Holding for 160 min at 1342°C
		Grey	3.59	1.12	0.230	0.277		
V-22 V-21 V-33 V-20 V-23	Influence of Si in Fe-C-Si alloy	Grey	3.43	0.42	0.209	0.533	1266°~1388°C	Heating-cooling
		Grey	3.33	1.94	0.223	0.283		
		Grey	3.47	2.14	0.240			
		Grey	3.35	2.70	0.250	0.297		
		Grey	3.40	3.49	0.236	0.304		
		Grey	3.40	3.49	0.236	0.304		
V-31 V-41	Fe-C	White	3.54	—	—	0.219	1255°~1388°C	Heating-cooling
		Grey	3.52	—	—	0.130		

Al* Al content after measurements.

(1) The viscosity coefficients of white and grey iron samples of Fe-C-Si alloy were measured at a cycle of superheating and cooling up to the maximum temperatures of 1500°, 1400° and 1350°C, respectively.

(2) Measurements were made at intervals of 2 to 20 minutes during holding of 2 to 3 hours at 1255°, 1288° and 1343°C, in order to investigate the behaviour of the viscosity with the lapse of holding time at each temperature. After the measurements the sample was superheated to 1388°C and then cooled to a given temperature, and the influence of the prolonged holding of the melt on the viscosity at this temperature was re-examined.

(3) Grey iron samples with varying silicon contents from 0 to 3.50% were used to investigate the influence of silicon on the viscosity. Measurements were made at each temperature in a cycle of superheating and cooling in the temperature range of 1255°~1388°C.

(4) The viscosity of white and grey cast irons of the Fe-C system was measured at each temperature of the superheating and cooling processes in the temperature range of 1255°~1388°C to compare with the viscosity of Fe-C-Si alloy.

III. Experimental Results

1. Difference between white iron and grey irons of the Fe-C-Si system

White iron and grey cast iron samples prepared from the same melt were superheated up to the maximum temperatures of 1460°, 1388° and 1343°C and then cooled. The results of measurements of the viscosity are shown in Figs. 1, 2 and 3.

The viscosity of grey cast iron shows higher values

Fig. 1 Viscosity curves in the superheating and cooling process by 1480°C.

curve in the superheating process does not correspond with that in the cooling process, where a negative hysteresis* is recognized as pointed out by Turovskii. The hysteresis of grey cast iron is the greatest at a maximum superheating temperature of 1480°C. The lower the maximum superheating temperature, the narrower the width of the hysteresis.

* The phenomenon in which the viscosity in a definite temperature range of the cooling process is higher than in that of the superheating process is called the *positive hysteresis*, while the adverse phenomenon is called "the *negative hysteresis*".

On the other hand, even in the white cast iron samples, the negative hysteresis is recognized by superheating and cooling except the case of the maximum superheating temperature of 1388°C; a considerable width of the hysteresis is observed even at the lowest superheating temperature (Fig. 3).

Fig. 2 Viscosity curves in the superheating and cooling process by 1388°C.

Fig. 3 Viscosity curves in the superheating and cooling process by 1343°C.

Fig. 4 Changes in viscosity by holding at 1342°C.

There is hardly recognized the difference between white and grey cast irons in the cooling process from the maximum temperature of 1480°C, but it might be thought that the structure of the molten state has a clear difference between the two, as there is a great difference below 1400°C.

2. Change of the viscosity of the melt by holding at prescribed temperatures

As there was recognized a phenomenon which might be presumed from the above results that the constitution of the melt differed by the state of samples, viscosities of white and grey cast irons were measured by holding the melt for a long time at comparatively lower temperatures. The results are shown in Figs. 4 and 5, respectively.

In the case of holding at 1342°C, the viscosity of grey cast iron decreases rapidly 20 minutes after arriving at the temperature, but it shows an almost constant value of 7.0 c. poise. On the other hand, the viscosity of the white cast iron sample shows the same tendency, but it becomes somewhat lower when it passes over 80 minutes. The viscosity of the white cast iron sample is considerably lower than that of grey cast iron up to 20 minutes of holding, but its difference will be less over 20 minutes. However, the viscosity

Fig. 5 Change in viscosity by holding at 1288°C.

of the grey cast iron sample shows a higher value than that of the white cast iron sample even though they were held for 2 hours and a half at this temperature.

When the holding temperature is lowered to 1288°C, the difference between grey and white cast irons becomes remarkable and there exists the difference of 1.0 c. poise even by holding for 2 hours. Especially, the difference is recognized to be extremely great within 10 minutes of holding. Thus the scattering of the viscosity values becomes greater with decreasing holding temperature, but the viscosity of the grey cast iron samples is still higher than that of the white one.

Fig. 6 shows the results of measuring how

the hysteresis of the viscosity curves is influenced by the state of the samples when the melt is superheated and cooled after having been held for a long time at 1288°C.

As seen from these results and Figs. 1, 2 and 3, hys-

Fig. 6 Viscosity curves in the superheating and cooling process after holding at 1288°C for a long time.

terises with the width of 1.0 to 2.0 c. poise appear in every curve. Accordingly, the width of hysteresis is proved not to be influenced greatly by the treatment of holding for a long time at lower temperature of 1288°C. However, a difference as large as 3.0 to 4.0 c. poise is recognized between the white and grey cast irons at around 1300°C, and it was confirmed that there still existed some difference between the two even though they were held for a long time.

3. Influence of silicon on the viscosity

As silicon in the cast iron gives a remarkable influence on the graphite structure and the chilling ability, it was investigated how the hysteresis of the viscosity curves was influenced by the silicon content. The results are shown in Fig. 7 (a) and (b).

It is clear from Fig. 7 (a) and (b) that hysteresis is hardly recognized up to 0.43% Si and the negative hysteresis which appears to a considerable extent at lower temperatures below 1300°C is recognized at more than 1.22% Si.

The viscosity in both superheating and cooling processes decreases to 0.50% Si with increasing silicon content, and increases over 0.50% and shows a maximum value at 1.20%. Then it decreases again in the range of 1.20 to 2.70%. At more than 3.0% it tends to increase a little in the superheating process, but it increases clearly in the cooling process. This result is in excellent agreement with Turovskii's experimental results.

4. The viscosity of Fe-C alloy

Hysteresis is hardly recognized in the viscosity curve of Fe-C alloy as shown in Fig. 8. However, the viscosity of the grey cast iron is higher than that of the white one by 1.0 c. poise.

IV. Discussion

It was confirmed from the above results that there existed a negative hysteresis in a cycle of superheating and cooling in the viscosity of molten iron and also a

Fig. 7 Influence of Si on the viscosity during the superheating and cooling process.

Fig. 8 Viscosity curves of Fe-C alloy.

great difference in the viscosity curves of white and grey cast irons. Discussion will be made on the reason of this interesting phenomenon.

The presence of graphite in the samples may be one

of the factors which causes the difference between the white and grey cast iron melts as the experimental conditions are quite the same.

Piwowsky⁽⁹⁾ reported that the dissolution velocity of graphite is extremely rapid in the molten state. Niwa et al.⁽¹⁰⁾ explained experimentally the mechanism of graphite dissolution from the diffusion film 0.03 to 0.004 cm in thickness: That is, the dissolution velocity depends upon the diffusion velocity of C from the interface to the bulk as the chemical reaction of C (solid) = C (in Fe) proceeds extremely rapidly at the interface of graphite and molten iron (bulk), actually reaching an equilibrium state.

However, from extensive basic data on graphite present in molten iron, Bunin⁽¹¹⁾ pointed out that graphite is only dissolved into molten iron, but there is the possibility of microgroup formation. Samarin⁽¹²⁾ assumed that silicon causes the precipitation of carbon from the melt to suspend in the colloidal state. Bogachef⁽¹³⁾ confirmed that the amount of graphite in molten iron increased with increasing silicon content, and obtained the data which proved the presence of a definite amount of especially stable free graphite at each temperature for a certain time.

On the other hand, the theory of the viscosity of heterogeneous alloys has not been studied quantitatively, but it is certain that the addition of a dispersed phase to the melt increases the viscosity. Shvidkovskii⁽¹⁴⁾ considered that the viscosity of a heterogeneous alloy depends upon the concentration and size of the mixed particles. Besides, the fact that the viscosity increases during the flowing of the melt in which a dispersed phase is contained can be explained by the restraints of the flowing by the phase.

From the above considerations, the hysteresis phenomenon of the viscosity in molten iron is explained here in relation to the dispersed graphite or microgroups* which exist in the melt.

1. The difference of the viscosity between white and grey cast irons

The difference of the viscosity between white and grey cast irons during the superheating process as shown in Figs. 1, 2 and 3 is thought to be caused by the fact that graphite in the grey cast iron sample exists as microgroups without complete dissolution into the melt and the amount of the microgroups is greater than that of the white cast iron sample.

At higher superheating temperatures, a greater amount of graphite forming microgroups dissolves

into the melt, and the viscosity shows almost the same value. There is little difference between white and grey cast irons during the cooling process after the same viscosity is attained at a higher temperature, which might be thought to be a state very close to the homogeneous melt. When very fine particles are in a colloidal state, the viscosity of the colloidal solution can be expressed by the following equation, according to Einstein:

$$\eta_s = \eta (1 + 2.5 \gamma).$$

Here η_s : the viscosity coefficient of the colloidal solution, η : the viscosity coefficient of the suspended medium, γ : volume of the suspended matter per unit volume of the colloidal solution. The change of γ calculated by Einstein's equation from Fig. 1 is shown in Fig. 9 (a). As the viscosity curves of white and

Fig. 9 Change in volume of the suspended matter with temperature in the case of holding the melt at 1288°C.

grey cast irons are almost the same during the cooling process after superheating up to 1450°C, calculation was made assuming these curves as a suspended medium.

It is shown from this result that even in the white cast iron sample a suspended matter of 0.24% exists at 1288°C and 0.09% at 1423°C. The reason why the white cast iron sample has also a suspended matter at comparatively lower temperatures is thought to be caused by the deposition of graphite during the heating process up to the melting point of white cast iron. It appears that such a tendency as shown in Fig. 9 (a) occurs because graphite exists in molten iron as microgroups and decreases its amount with increasing temperature. On the other hand, it is predicted that because of the already existing graphite the amount of the suspended matter, i. e., microgroups, is greater in the grey cast iron than in the white one. In the case of lower maximum temperatures below 1400°C as shown in Figs. 2 and 3, the viscosity of grey cast iron is by far higher during the cooling process than that of white

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* Lux⁽¹⁵⁾ reported that graphite passed through the intermediate step during the dissolution and precipitation process. Therefore, on the assumption that carbon molecules of C₃, C₄, C₅, and C₆ are present in addition to the graphite ring of C₆, they are called "microgroups" in all.

(15) B. Lux: Modern Castings, 45 (1964), 222.

cast iron because the microgroups do not dissolve completely into the melt. As the microgroups do not hardly dissolve into the melt at the maximum temperature of 1343°C, the viscosity of grey cast iron during the cooling process is thought to be almost the same as that during the superheating process.

2. Change of the viscosity of the melt by holding at prescribed temperatures

If the results of Figs. 4 and 5 are similarly examined from the view point of the microgroups, it can be explained that the viscosity decreases with the holding time and the difference between white and grey cast irons becomes smaller and smaller. That is to say, the amount of the microgroups dissolved at a given temperature is thought to increase with time. The change in γ of the suspended matter calculated from the result of Fig. 5 by Einstein's equation is shown in Fig. 9 (b). Both white and grey cast irons showed a rapid decrease in volume after 10 minutes of holding. Here, γ was calculated taking the viscosity at 1288°C during cooling from 1480°C (Fig. 1) for that of the non-colloidal solution. There is a suspended matter of 0.08% in grey cast iron even after 2 hours of holding, but any suspended matter does not exist in white cast iron after 70 minutes. However, as described before, it is indicated that the suspended matter does not disappear completely. This inconsistency seems to be caused by the assumption that the above-described suspended medium is a complete non-colloidal solution. Consequently, the fact that there is still a great difference in viscosity between white and grey cast irons when measured during a cycle of superheating up to 1388°C and cooling from this temperature after holding at 1288°C can be thought to be influenced by the presence of the suspended matter without complete dissolution of microgroups into the melt. Besides, the amount of the suspended matter dissolved increases with increasing holding temperature to 1342°C, and so the difference between white and grey cast irons becomes less as shown in Fig. 4.

On the other hand, the reason why the values of viscosity are scattered at the lowest holding temperature of 1255°C is that the measurement at such a low temperature is problematical from the point of view that it is doubtful whether the sample is completely in

the molten state.

3. Influence of silicon

The increase of viscosity in the range of 0.50 to 1.25 % Si in Fig. 7 (a) (b) can be thought to be caused by the increase of microgroups in the melt because of the decrease of carbon solubility induced by the increase of silicon in this range. The decrease of viscosity in the range up to 2.50% Si cannot be clearly explained, but the increase of viscosity over 2.50% Si seems to be caused by the formation of Fe_3Si .

4. The viscosity of Fe-C alloy

A very narrow hysteresis in the viscosity of Fe-C alloy seems to be caused by a comparatively rapid dissolution of microgroups into the melt because of the scarcity of graphite, but the much larger amount of graphite in the grey iron than in the white iron can be attributed to the difference shown in Fig. 8 as the amount of the microgroups is smaller in the latter than the former.

As described above, the phenomenon of hysteresis in the viscosity of the cast iron mentioned above and the great difference in viscosity between white and grey cast irons with the same chemical composition have been discussed in the light of the microgroups, based only on the viscosity of molten iron. Therefore, it is necessary to confirm the existence of microgroups by further experiments on the electric conductivity and eutectic solidification velocity of molten iron.

V. Conclusion

The viscosity of molten iron has been measured to investigate the phenomena which are related to the properties of cast iron after melting.

The results obtained confirmed the existence of hysteresis in the viscosity during a cycle of superheating and cooling, and a great difference in values of viscosity between white and grey cast irons of the same chemical composition. This was assumed to be caused by the phenomenon that graphite in the samples did not dissolve completely into the melt, but the microgroups were formed in molten iron. The microgroups seem to depend upon the tendency of graphitization during the stage of solidification.