Studies on Decarburisation and Desiliconisation of Levitated Fe–C–Si Alloy Droplets

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The rate of decarburization and desiliconisation has been studied in several Fe–C–Si alloys containing 4% C and 0.37–0.71% Si using electromagnetically levitated droplets at 1 400 and 1 660°C in oxygen and helium atmospheres containing 10 and 20% oxygen. It has been found that the decarburization proceeds till a carbon content of about 0.5% is reached at a constant rate (the rates are 0.11 and 0.17%/s respectively for 10 and 20% oxygen) that is independent of temperature and silicon content. Beyond this carbon level, not only the decarburization rate has decreased but also the silicon content has started falling down. Optical microscopy studies showed typical white iron structures in the initial stages of decarburization (due to the small size of the droplet as well as rapid cooling) while low carbon acicular ferrite structure when % O₂ was 20% and medium carbon structure when % O₂ was 10%. This corresponded to the higher carbon content (~0.38%) of the latter. SEM studies did not reveal any oxide layer on the surface but EDS showed that the oxygen concentration was higher (0.02 to 0.24%) that extended to about 0.5 to 1 μm depth suggesting that oxygen gradients do occur even in the initial stages of decarburization due to the high level of turbulence within the droplet due to the electromagnetic effects in levitation. Calculations based on Thermocalc gave logarithmic decrease for decarburisation while it was found to be linear experimentally. This could be due to operating parameters other than diffusion (like convection) taking place or nonequilibrium conditions (created due to turbulence) and shorter times used for levitation.

KEY WORDS: levitation; steel droplets; decarburization; desiliconisation; Thermocalc.

1. Introduction

It is well known that the technique of levitation melting in studying the decarburization in Fe–C alloys has the advantage that it avoids complications arising from crucible side interactions with the molten metal and it also enables study of very small samples weighing about a gram. This could allow understanding of the behaviour of molten droplets splashing out of the liquid steel due to the blowing of oxygen gas in a converter where a considerable amount of decarburization takes place. Though there is a long history of using the levitation melted samples for investigations on decarburization characteristics of ferrous alloys, the total number of these investigations is not that many.

Baker et al.1) were probably the first to use the levitation melting technique for making a detailed study on the decarburization of liquid iron in an oxidizing atmosphere. They studied 0.7 g samples of Fe–0.5% C alloy heated by electromagnetic levitation in an atmosphere to 1 660°C in CO₂ using CO/He as diluents and followed the decarburization at this temperature. They observed that the rate of decarburization was constant and the rate varied substantially with the mole fraction of CO₂ (taking 20 min to 30 s for complete decarburization as the mole fraction varied from 0.01 to 1) but was independent of gas pressure. They concluded that the decarburization occurs predominantly, if not wholly, by gaseous diffusion.

The above authors2) also studied the decarburization rate of Fe–C alloys in oxygen diluted by He (with 1, 10 and 100% oxygen) and found that it was constant, independent of the initial carbon content and that oxygen was 2.5 times more effective than CO₂. They concluded that oxygen diffusion through the gas boundary layer i.e., gaseous diffusion controls decarburization wholly. However, low carbon melts (< ~1% C) became unstable when the atmosphere had 10% oxygen and when decarburized with CO₂ the decarburization rate decreased with initial carbon content. The authors have therefore concluded that carbon diffusion within the droplet was important for low carbon melts. In this case CO bubbles were found to nucleate homogeneously within the droplet.

Distin et al.3) studied decarburization of 2 g samples of Fe–4wt% C by levitation melting in various atmospheres of CO₂, O₂, O₂ + H₂O, and Ar + H₂O and found that in all cases the exothermic reaction increased the initial temperature from about 1 760°C by 10–30°C to a steady value and that the droplets got decarburized at a constant rate to about 0.5–1% C at which oxide particles formed suddenly on the surface and tended to collect at the bottom side of the droplets and caused them to boil. They found that the decarburization rate was dependent on the flow rate which made them conclude that the rate controlling factor was the
mass transfer in the gas phase. The mean rate of transfer of mass between surface of the spherical particle and gaseous fluid was found to obey Steinberger and Treybal equation that incorporates additive terms for radial diffusion, natural convection and forced convection (forced convection was important only at high flow rates). They also reported that the %C at which oxide formed varied linearly with the decarburization rate (0.1 to 0.9%) as the rate increased from ~0.02 to ~0.16%/s) and this did not depend on the oxidising medium employed by them.

Sano and Matsushita used the levitation technique to study desiliconisation of liquid iron–silicon alloy droplets containing 1.2, 1.3 and 1.6% silicon at temperatures of 1550 to 1800°C by using gas mixtures containing CO2 or O2 diluted with He. When they used samples weighing 0.7–1.4 g the temperature rose by 100°C from the initial value of 1550°C due to the initial exothermic reaction and then it was controlled at the desired value. It was observed that the silicon loss was parabolic with time, being proportional to $\sqrt{t}$ and Si loss varied from 0.02 to 0.3%/min as $P_{CO_2}$ varied from 0.026 to 0.3 at 1550°C. However, when higher Si containing alloys were heated to 1700°C, fuming was observed close to the droplet surface towards the end of desiliconisation and then deposits of solid SiO with a small amount of FeO formed on the wall of the reaction tube. This was considered equivalent to the formation of oxide in Fe–C alloys. Gaseous SiO was reported to be the reaction product at higher temperatures and lower oxygen potentials. Like in decarburisation, the mechanism of desiliconisation was also attributed to the mass transfer correlations for flow of SiO and CO2 around the droplet. At lower temperatures when oxide formed on the surface, diffusion of oxygen ion through the oxide layer was supposed to govern the removal of silicon from the melt.

El-Kadda and Robertson showed that while a gas mixture of CO and CO2 (either 1.1 or 2.15% CO2) under a high pressure of 40 atm. and at 1650°C can cause decarburisation in high carbon melts (5.0 and 5.5% C) in 1 g samples of Fe–C alloys, it can cause carburisation under the same conditions in pure iron. In either case an equilibrium level of carbon (about 1.5 and 2% respectively for 1.1 and 2.15% CO2 mixtures) was achieved in 10 to 20 min. Neither the carburisation rate nor the decarburisation rate was constant (as reported by earlier investigators) and the rate decreased as equilibrium approached. Equilibrium was attained in half the time for 2.15% CO2 mixture as compared to the 1.1% CO2 mixture. From these results, they concluded that the rate of decarburisation was controlled by diffusion in the gas phase for carbon concentrations above 3 wt% C and by mixed transport of gaseous diffusion as well as transport in the liquid phase below this level.

Lee and Rao studied the influence of 0–0.05% sulphur on the decarburisation of Fe–C melts containing 1% and 2.5 wt% C using CO–CO2 gas mixtures at 1700°C by levitation technique on samples of 5 mm diameter weighing 0.7 g. For $P_{CO_2}/P_{CO} = 1/9$, a gas flow rate of 1 L/min and 1 atm pressure, the decarburisation rate of 1% C alloy decreased from 0.035 to 0.025/s when the %S increased from 0 to about 0.05 and above this %, S did not have any noticeable effect. There was no change in the sulphur content of the steel during the process of decarburisation. They also found that the decarburization rate was the same for both Fe–C alloys studied, when other conditions remained the same. The effect of gas flow rate was also diminished in the presence of sulphur. In all the alloys studied, the decarburisation rate remained constant till the oxide appeared on the surface of the droplet and this varied from about 0.05 to 0.15% C, the lower value for alloys containing high sulphur.

Nagasaka and Fruehan studied the decarburisation kinetics in Fe–C–0.2wt%S alloys at 1773 K using CO2–Ar, CO2–H2O and CO2–Ar–H2O atmospheres and concluded that the mixed atmospheres would produce rates in an additive manner without having any synergistic effects. The sulphur loss during decarburisation was only 5–12% depending on the atmosphere used, the maximum being for CO2–30%H2O among all gas mixtures used by them. Iron oxide has formed on the surface only when the carbon content in the droplet reached a critical value that increased with lowering sulphur and increasing partial pressure of CO2 or H2O.

Sun and Pehlke studied the simultaneous oxidation characteristics of C, Si and Mn in an Fe–3.35%C, 2% Si, 0.36 Mn and 0.05% S using cube shaped samples weighing 1 g over a temperature range of 1575 to 1715°C and gas mixtures of CO2 and N2. They found that decarburisation (at a linear rate) took place first and the removal by oxidation of Si and Mn started after the carbon got lowered to ~0.4%. This took about 1 min at 1635°C when the CO2 was 20% at a flow rate of 500 mL/min. While the presence of Si and Mn as well as temperature did not alter the kinetics of decarburisation, increasing the flow rate enhanced the rate. They have also concluded that the diffusion of CO2 from the boundary layer in the gas mixture controlled most of the decarburisation period though diffusion within the metal droplet and diffusion of CO in the gas phase away from the droplet were important in the later stages.

Sun and Pehlke later developed a reaction model based on their experiments on levitation melting of Fe–C–Si–Mn alloys which reveal that for alloys containing 1.64 or 3.38% C, oxidation of Si and Mn occurred only after the cessation of the vigorous decarburization reaction at the high temperature of 1873 K (observed when oxide particles were seen at the bottom side of the droplet) but in the case of alloys containing 0.4% C removal of silicon and manganese started from the very beginning of decarburization reaction. However, at temperatures lower than 1633 K, oxide layer formed early over the entire droplet enabling removal of Si and Mn, though it lowered reaction rates considerably. They concluded that gaseous diffusion controls reaction at high temperatures while diffusion through oxide film may become the rate controlling step in lower temperatures.

Simento et al. investigated the decarburization of Fe–C–S melts containing 3.2 and 4% C and sulphur varying from 5 ppm to 0.27% by electromagnetic levitation technique. Samples weighing 0.8 g were decarburized at 1723 K using H2O vapour (nitrogen was used to dilute) and showed that the decarburization rate for the alloy containing 5 ppm S increased from ~0.02 to 0.1% as the partial pressure of water vapour increased from 0.05 to 0.2 for a gas flow rate of 0.0147 m³/min. Increasing sulphur from 5 ppm to 0.27% decreased the decarburization rate from ~0.05 to 0.02%/s. They concluded that the decarburization rate...
occurred by the mixed control of gas phase mass transfer and dissociative chemisorption of H$_2$O at the melt surface. The gas phase mass transfer obeyed Steinberger-Trebal equation that takes into account both natural convection and forced convection (this was also reported by Distin et al. $^{3}$ earlier).

Simento et al. $^{11}$ have also studied the Fe–C–S droplets using mixtures of O$_2$–CO$_2$ and O$_2$–H$_2$O diluting with N$_2$ or He at 1723 K. They have confirmed their earlier observation that S would retard decarburization even when mixtures of decarburizing gases were used. They reported that though CO$_2$ and H$_2$O would function similarly when combined with O$_2$, the presence of H$_2$O would retard decarburization as it would promote the reaction between incoming O$_2$ and outgoing CO generating CO$_2$ (that was not as effective as O$_2$).

Liu and Harris $^{12}$ have recently demonstrated that decarburization of a levitated steel droplet at the ultra low carbon levels can occur even under vacuum and reported that below about 1500 ppm, decarburization rate was proportional to the carbon content at 1550–1850°C (about 10–20 ppm carbon/s) and at vacuums of 20 and 40 Pa.

Gao et al. $^{13}$ studied carbon boil in Fe–C–S drops in a horizontal tube furnace over a temperature range 1370–1450°C and reported that though reduction in carbon should increase the density of droplets, its volume actually increased by about 50% before carbon boil (it took about 15 s for this to happen) because of the excessive formation of the subsurface gas phase within the droplet which was followed by the spitting of the droplet in an explosive manner. The time for carbon boil was not affected by sulphur when present up to about 0.2% beyond which the time for boil got reduced gradually as sulphur content increased. An important finding is that the oxygen content of the droplet increased much before the carbon boil started in both sulphur-free and sulphur-bearing alloys and the authors attributed it to the formation of FeO.

It is evident from the above that most of the investigations were confined to Fe–C alloys and studies on the decarburization in the presence of other elements (excepting sulphur) are very limited and further how the other elements get oxidized has also not drawn that attention. The present investigation has therefore been taken up to investigate the decarburization characteristics of Fe–4%C alloys containing silicon in the range 0.3 to 0.7% and also to see when the silicon in the alloy gets oxidized. The present investigation deals with a study of the influence of temperature, oxygen content of the atmosphere and silicon content of the alloy on its decarburization and desiliconisation behaviour.

### Table 1. Materials used for preparation of specimens for the experiments.

<table>
<thead>
<tr>
<th>Material</th>
<th>Pure Iron</th>
<th>Graphite</th>
<th>Silicon</th>
<th>Helium 1.8</th>
<th>Helium 5.6</th>
<th>Oxygen 5.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>Analysis</td>
<td>%C = 0.018</td>
<td>%Si = 0.007</td>
<td>%Fe = 0.002</td>
<td>%Cr = 0.100</td>
<td>%Ni = 0.070</td>
<td>%Cu = 0.051</td>
</tr>
<tr>
<td>Supplier</td>
<td>Sandvik Steel</td>
<td>Le Carbone</td>
<td>E. Merck, Darmstadt, Germany</td>
<td>AGA Gas, Stockholm, Sweden</td>
<td>AGA Gas, Stockholm, Sweden</td>
<td>AGA Gas, Stockholm, Sweden</td>
</tr>
</tbody>
</table>

### 2. Experimental

The specimens used in the experiments were prepared by melting pure iron and alloying it with high purity graphite. Part of the melt was sampled as reference by using vacuum pipettes with an inner diameter of 6 mm. After the reference samples were taken, silicon was added aiming at 0.1% Si in the melt. After allowing for the silicon to dissolve, another set of samples was taken. This procedure was repeated with a 0.1% Si step up to an aimed silicon content of 1.0%. The silicon yield was estimated to approximately 80%; thus for each 0.1% step in the silicon content an amount corresponding to 0.12% was added. The samples were cut and carefully polished to a weight of 2.00 g (±0.005 g).

For levitating, melting down and as diluting gas during decarburisation Helium 5.6 was used (purity ≥99.9996%). As reacting gas Oxygen 5.0 was used (purity ≥99.999%). Helium 1.8 (purity ≥98%) was used for cooling the samples. The gases, except the cooling gas, were purified by passing through silica gel and dehydration Mg(ClO$_4$)$_2$ to remove water and ascarite to remove carbon dioxide. Data on all materials used is provided in Table 1.

The experiments were conducted in a levitation melting apparatus shown schematically in Fig. 1. The gas supplying systems were equipped with gas purification units, flow me-
ters and magnetic valves. Two bubble bottles were used to eliminate pressure build up in the system when valves were closed. The magnetic valves were controlled by a computer equipped with a DAP801 data acquisition extension card thus enabling accurate timing for opening and closing of the valves. The software used to program the time scheme for the valves was DAP Windows version 2.1. This computer also continuously sampled temperature data from the pyrometer.

The reaction chamber consisted of a quartz tube equipped with a removable seal at the bottom and a quartz prism holder at the top. Both ends were carefully tightened with rubber rings. The loading rod was made of glass with an alumina cup attached to it. The loading rod was carefully fitted into a drilled hole through the bottom seal and tightened with a rubber ring. The induction coil was made by winding 8 mm copper tubing around a cylindrical brass mandrel with a coning section to achieve a 30 degree angle to the center axis of the mandrel (and thus to the center axis of the silica tube). The coil was designed with two lower turns and one upper reverse turn. The power was supplied by a Philips High Frequency generator with a frequency of 500 kHz. The temperature was measured by a Landmark Infrared System 3 High Temperature Ratio Thermometer. The accuracy of the temperature measurement was verified against the melting point of the reference material and was estimated to be ±10 degrees.

Initially each section of the gas supply system was flushed with its corresponding gas for 15 min. The oxygen section of the system was then flushed with high purity helium for approximately 2 min. After turning on the power, the specimen was pushed up to a position where it levitated. The computer was programmed so that the oxygen valve opened at a certain predefined temperature of the melted specimen in all the experiments except for set no. 2 where heating up to a constant temperature was allowed. The temperature curves for set no. 2 and 3 are shown in Figs. 2 and 3. The time interval for the reaction period where the oxygen valve was open was given in numbers of 1/10 of a second. After reacting with the helium/oxygen for the predetermined time, the valves for oxygen and helium were closed and the helium valve was opened simultaneously. The time delay for this operation was no more than a few tenths of a second. The specimen was then cooled in situ before the power was turned off and the non-deformed droplet fell down into the alumina cup.

In all, six experiments were conducted on steels having 4% carbon and varying amounts of silicon (0.31–0.71%) using a gas mixture comprising 80% He and 20% O₂ or 90% He and 10% O₂. The steels also were preheated to varying temperatures in the range 1400–1630°C in He gas before admitting oxygen for decarburisation. The details of these sets of experiments are listed in Table 2. Samples were analysed for carbon and silicon after fixed intervals of time up to ~40 s during which period the temperature of
the sample was also recorded. The samples were then analyzed for their carbon and silicon content examined in optical microscope to record the changes in microstructure and in SEM for any oxides that might have formed.

3. Results
3.1. Decarburisation Kinetics
3.1.1. Oxygen/Helium Ratio: 20/80
The results on the decarburisation of different steels belonging to sets #1–4 using an oxygen/helium ratio of 20/80 are shown in Fig. 4. As can be seen, there is no significant effect of silicon (in the range 0.37–0.71%) on the decarburisation rate, though a slight tendency towards a more rapid decarburisation can be inferred at higher silicon levels that may well be within the limits of experimental uncertainty. The silicon level remained constant until the carbon content was below 0.5%. Since the initial temperature varied considerably for different sets (1 400–1 630°C) it also does not seem to have any effect on the rate of decarburization that remained constant at about 0.17%/s till the carbon content reached about 0.5%. Below this level the decarburization rate decreased but continued till the carbon level was 0.2% when a vigorous reaction took place causing heavy fuming and lateral instability. This resulted in the droplet hitting the tube wall and collapsing into small pieces.

3.1.2. Oxygen Helium Ratio: 10/90
The results for sets #5 and 6 are shown in Fig. 5. These also do not show any effect of the silicon level on the rate of decarburization. The final carbon content in these sets was 0.38 and 0.33% respectively. The decarburization rate was constant, but lower than the previous case, at 0.1%/s. The results on sets #2 and #6 were superimposed in Fig. 6 to demonstrate the effect of oxygen potential on the rate of decarburization where it is clearly seen that lower oxygen concentration results in a lower rate of decarburization.

3.2. Desiliconisation Kinetics
As Figs. 4 and 5 reveal, the silicon content of the droplet has not changed till the carbon content came to a low level of about 0.6 and 0.35% respectively for the two oxygen concentrations of 20% and 10% O₂. These values were independent of the initial silicon content of the steel (which was in the range 0.37 to 0.71%). Once the silicon content has started decreasing, it took about 6 s for bringing the silicon from 0.71 to 0.45% in sets #4 and 7 and 0.37 to 0.2% in sets #1 and set #6. It must be noted that the starting carbon levels were different when desiliconisation has started.

Table 2. Conditions during decarburisation experiments. (*=temperature at which reaction commences.)

<table>
<thead>
<tr>
<th>Set no.</th>
<th>Initial temp. * (°C)</th>
<th>Flow rate (L/m)</th>
<th>%O₂/%He</th>
<th>Initial composition</th>
<th>Initial Weight (g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1450</td>
<td>5.0</td>
<td>20/80</td>
<td>4.0</td>
<td>0.37</td>
</tr>
<tr>
<td>2</td>
<td>1630</td>
<td>5.0</td>
<td>20/80</td>
<td>4.1</td>
<td>0.48</td>
</tr>
<tr>
<td>3</td>
<td>1400</td>
<td>5.0</td>
<td>20/80</td>
<td>4.0</td>
<td>0.54</td>
</tr>
<tr>
<td>4</td>
<td>1400</td>
<td>5.0</td>
<td>20/80</td>
<td>4.0</td>
<td>0.71</td>
</tr>
<tr>
<td>5</td>
<td>1400</td>
<td>5.0</td>
<td>10/90</td>
<td>4.0</td>
<td>0.54</td>
</tr>
<tr>
<td>6</td>
<td>1400</td>
<td>5.0</td>
<td>10/90</td>
<td>4.0</td>
<td>0.71</td>
</tr>
</tbody>
</table>
taking place. The oxygen concentration in the gas mixture did not have any major effect on the rate of oxidation of silicon from the melt.

3.3. Optical Microscopy

Samples for optical microscopy were taken from set #2 after decarburization for 3 s and 24 s and from set #5 after 3 and 42 s. Figures 7(a) and 7(b) and Figs. 8(a) and 8(b) show the microstructures for these conditions respectively for sets #2 and #5. The dendritic structure with fine eutectic is typical of the white iron structure and must have been caused because of the high cooling rate. Figure 7(b) shows acicular ferrite structure with small amounts of pearlite and here again the effect of fast cooling rate is evident. Figure 8(a) is similar to that in Fig. 7(a) except that the austenite dendrite content was higher because of the higher carbon (expected because of the lower rate of decarburization). Figure 8(b) shows a typical medium carbon structure with pearlite nodules along with ferrite. No visible oxide layer was seen on the surface in any of these specimens.

3.4. Scanning Electron Microscopy

Two samples each from sets 2 and 5 were examined under Scanning Electron Microscope and the regions near the surface of the samples were studied to find out whether any oxide layer existed on them. They were examined under back scattered electron mode as it would reveal atomic number contrast which can image without etching the sample. No difference in contrast was seen near the surface of any sample meaning that no distinct oxide layer has formed.

Fig. 7. Optical microstructures of samples from set #2 after decarburization for (a) 0 s and (b) 24 s.

Fig. 8. Optical microstructures of samples from set #5 after decarburization for (a) 3 s and (b) 42 s.

Fig. 9. Curves showing the oxygen content of the samples from Sets 2 and 5 as a function of distance from the surface after decarburization for (a) 3 s and (b) 6 s.
on the surface. However, chemical analysis using Energy Dispersive Spectroscopy showed a clear gradient in oxygen concentration. A very thin layer of about 0.2–0.5 μm at the surface showed 10 to 40% oxygen as shown in Figs. 9(a) and 9(b) indicating that the surface got enriched with oxygen even in the initial stages of decarburization, though no oxide layer has formed.

3.5. Results from Thermocalc Calculations

Calculations were made for three sets using Thermocalc software, developed at the Royal Institute of Technology, to calculate the temperature variation with time during decarburization taking into account the radiation losses and the results are shown for set #2 in Fig. 10. This shows good agreement with the experimentally measured values.

The calculated values of carbon contents of all the samples were much less than those observed, as seen from Figs. 11(a) and 11(b). The theoretical results were obtained by making a series of calculations using Database SSUB 3, TCFE 3 and SLAG 1 software programmes (one can get details from www.thermocalc.se) for 10 and 20% oxygen mixtures with helium gas assuming that carbon monoxide gets replaced (the CO content changes with time continuously due to decrease in carbon in the droplet) after every 3 s and also assuming that the equilibrium conditions prevail. Though this assumption cannot be right as the reactants were left to react for a limited period of time, the vast difference in the calculated and observed values gives us a
clear idea that the refining reactions are instantaneous in nature and do not follow equilibrium conditions. Though several investigators considered mass transfer as the dominant mechanism in concentrated oxidizing atmospheres, there is still debate about this and diffusion within the droplet playing an important role in the decarburization process (as discussed in the introduction).

4. Discussion
The present investigation shows that the decarburization of all the Fe–C–Si alloys studied proceeds at a constant rate for most part of the process, i.e., till the carbon level comes to about 0.5% or less, that is independent of the temperature or silicon content. After this level, there seems to be a drop in the rate of decarburization. The rate of decarburization is a strong function of the partial pressure of the oxidizing gas. For instance the rate varied from 0.11 to 0.17% C/s as O₂ in the oxygen–helium mixture increased from 10 to 20% when a gas flow rate of 5 L/min was used. The observation that the decarburization rate is constant for most part of the process is constant in agreement with several earlier investigators,1,4–6,13) though not with EL-Kaddah and Robertson5) who found that it decreases as the carbon content decreases till an equilibrium level depending on the partial pressure of CO₂ (which was 1.5% C for 1.1% CO₂ and 2.5% for 2.5% CO₂). This could be due to the very low %s of the oxidizing gas they employed. Baker et al.7) reported the decarburization rates to vary from 0.11 to 0.9% C/s as % O₂ varied from 1 to 100% (till the carbon content decreased to about 0.2%) at 1660°C when the flow rate was 5 L/min. The present results are in accordance with this as the observed rates are 0.11 and 0.17% C/s in gas mixtures containing 10% and 20% oxygen respectively. Baker et al.11) reported the rates to be 0.12 and 0.17% C/s when they used 50% and 100% CO₂ (Fig. 2) suggesting that twice the amount of O₂ or CO₂ is needed to increase the decarburization rate from 0.11 to 0.17% C/s. Distin et al.3) however, found that O₂ is twice as effective as CO₂ in decarburization (see their figures 2 and 5). Sano et al.8) also reported decarburization rate of 0.03% C/s when % oxygen was 3%.

In the case of investigations where crucible melting experiments were carried out or when Fe–C alloys containing other elements were used, the decarburization rates were much lower. Thus Nomura et al.15) as well as Sun et al.8,9) have reported lower rates when the alloy was melted in crucible. The latter authors as well as Lee and Rao6) have reported that the presence of sulphur would decrease the rate of decarburization. Sun and Pehlke,9) however, reported that the presence of silicon and manganese in the iron has not influenced the decarburization rate (they reported rates of 0.025 and 0.5% for gas mixtures containing 10% and 20% CO₂).

The present investigation reaffirms the conclusion of the earlier investigators that temperature of the molten drop has no influence on the decarburization rate. In the present investigation the decarburization rate was found to be constant either at 1400 or 1630°C. It is observed in the present investigation that the temperature of the droplet increased continuously as the decarburization proceeded due to the exothermic nature of the reaction. Thus the temperature has gone up from 1400°C to as high as 1800°C and from 1650 to 1910°C in a matter of 18 s for sets 2 and 6. These temperature increases are far higher than those reported by earlier investigators. Thus Distin et al.3) recorded only a change of 10–20°C and EL Kaddah and Robertson5) stated that the temperature once adjusted to the reaction temperature initially maintained constant during the reaction. Simento et al.10,11) maintained the temperature continuously by manipulating the current passing through the coil. In Fig. 12 these temperature changes have been plotted along with the liquidus temperature of the droplet obtained from the carbon content using the Fe–C diagram. It can be seen that the cooling of the droplet is uniformly high by ~200°C in set 6 while it is more than 400°C for set #2. It is interesting that such big changes in temperature in the range of 1400 to 1900°C have not brought any change in the decarburization rate. This is because, as most earlier investigators concluded that the mass transfer of the decarburizing (or oxidizing) gas like O₂ or CO₂ in the gas phase towards the droplet controls decarburization for most part of it (till carbon reaches a low value) and diffusion within the droplet as well as transport of CO in the gas phase away from the droplet may be important only in the last stages. This is also the main reason for getting constant rate of decarburization as the composition within the liquid droplet or its temperature does not have any control in a substantial part of the process.

The carbon content at which iron oxide forms at the surface of the droplet has also drawn considerable attention by earlier investigators. Distin et al.3) arrived at this value for a wide range of decarburization rates (Fig. 13 of their paper) and for rates of 0.1 and 0.2%/s observed in the present investigation for 10 and 20% oxygen concentrations match well their carbon contents of 0.5 and 0.9% respectively (these were the concentrations where the decarburization rate has fallen and deviated from constancy). However, Lee and Rao6) using CO/CO₂ gas mixtures where low rates of decarburization (0.03 to 0.07% C/s) were observed reported that the critical carbon where FeO would appear as very low and in the range 0.05–0.15%. Nagasaka and Fruehan7) found the critical carbon to vary from 0.1–0.4% for Fe–C alloys containing 0.1% S. The present work demonstrates that though oxide does not form as in the initial

![Fig. 12. Change in temperature for specimens of set 2, set 6, liquidus for set 2 and liquidus for set 6 with time.](image)
stages of decarburization, oxygen concentration gradients can occur within the droplet enriching the surface layers in oxygen.

It must be mentioned here that the models proposed by almost all the earlier investigators assumed uniformity in composition as well as temperature within the droplet. There have been several investigations in recent years to model the shape of the liquid drop during electromagnetic levitation as well as the conditions that prevail within the droplet. Asakuma et al. have shown that the droplet of molten silicon can change its shape to a prolate spheroidal, the extent of which depends on the electric current ratio of the heating coils to the levitation coils in a microgravity environment. Although the authors (interesting that there are 9 authors for this 3 page paper) state that their results are important while measuring the thermophysical properties using the electromagnetic levitation (EML) technique, their results can be used to keep the shape near spherical by varying the experimental parameters. The intensity of electromagnetic field on the fluid flow within the droplet and the turbulence it causes to change viscosity and velocity locally within the droplet has been the subject of investigations of several others. Berry et al. showed that the localized viscosity can change by a factor of 2 due to positioning field while the heating coils can alter by a factor of four causing considerable anisotropic distribution of flow. Hyers et al. later showed that flow inside the droplet can change from a laminar flow to viscous flow as the Reynold’s number reaches 600 (this value is about 2,300 for water flowing in a pipe) and used tracers to demonstrate that “noncoherent chaotic motion signifying emergence of turbulence inside the drop” occurs due to EML. Since viscosity controls Reynold number to a considerable extent and since it was shown that the viscosity is nonuniform within the droplet, the flow also could be complex having laminar to turbulent characteristics within the droplet. Bojarevics and Periculous have also modeled the EML droplets to show that not only intense fluid flow is generated within the droplet but the droplet can oscillate and that an additional DC field has to be used to stabilize the oscillations and turbulent velocity field. Even in the field of solidification, Medina et al. have shown recently that electromagnetic forces can alter the bulk flow structure altering the pressure distribution along the solidification front thereby altering solute gradients within a component.

5. Conclusions

(1) The rate of decarburization in several Fe–C–Si alloys is constant till the carbon content goes down to about 0.5% after which the rate decreases considerably. The rate depends mainly on the partial pressure of decarburising gas (Oxygen). When the oxygen content increased from 10 to 20% in He–O₂ mixture, the decarburisation rate increased from 0.11 to 0.17% per second. It is not influenced either by the temperature when it was in the range 1400–1700°C or by the silicon content when it was in the range 0.37–0.71%.

(2) The silicon content did not change significantly during the process of decarburisation till % C was 0.5, beyond which a decrease in % Si was observed.

(3) Optical microscopy studies showed typical white iron structures in the initial stages (due to very high rapid cooling) while low carbon acicular ferrite structure when % O₂ was 20% and medium carbon structure when % O₂ was 10%. This corresponded to the higher carbon content (~0.38%) of the latter.

(4) SEM studies did not reveal any oxide layer but EDS showed that the oxygen concentration was higher at the surface layers extending to about 1 mm depth in all cases, even in the initial stages of decarburization suggesting that oxygen gradients do occur due to high level of turbulence within the droplet due to the electromagnetic effects in levitation.

(5) Calculations based on Thermocalc gave logarithmic decrease for decarburisation while it was found to be linear experimentally. This could be due to operating parameters other than diffusion (like convection) taking place or non-equilibrium conditions (created due to turbulence) and shorter times used for levitation.

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