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Research Article

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Study of the mechanism of the direct reduction roasting of the limonite in Jiangxi

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ABSTRACT

A process with coal-based direct reduction followed by magnetic separation is presented for recovering metallic iron from a limonite in Jiangxi province, the effects of reduction temperature and reduction time on the concentrate product were investigated. The results show that, the reduction time has an important influence on the formation and growth of the metallic iron particles. When the reduction time is too short, the iron particle formed is fine; when the reduction time is too long, a large amount of fayalite produced by the solid reaction of FeO and SiO_2, which may resulted in a decrease of the iron content of the concentrate. The increase of reduction temperature has a positive impact on the gas diffusion and chemical reaction. The higher the temperature, the faster the chemical reaction which is facilitated to the agglomeration of the metallic iron; but the temperature should not be too high, too high temperature resulted in the gas diffusion velocity increased, it is difficult to provide reducing atmosphere for the reduction reaction.

Key words: limonite; direct reduction; mechanism study; low grade

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INTRODUCTION

In nature, most of limonite occurs in the form of $2Fe_2O_3$•$3H_2O$, is amorphous, aphanitic or jelly, as amorphous iron oxides and hydroxides, with goethite, mainly [1,2]. The reserves of limonite ore in Jiangxi province are very large, but because the chemical composition of the limonite is not fixed, and the iron content is not stable, moisture content changes to a large extent. Moreover, it be easy to over crushing during crushing and grinding [3-5]. Therefore, it belongs to a very refractory iron ore. At present, the rate of resource utilization of this ore has been very low since its complex characteristic. The recovery of the iron is only about 50% when the high intensity magnetic separator was employed. Direct reduction roasting-magnetic separation is one of the effective ways to recover iron from the refractory iron ore. In this paper, the process of direct reduction roasting-magnetic separation was employed to recover iron from refractory limonite ore in Jiangxi province, the effect of roasting conditions on the grade and recovery of iron concentrate were studied [6-10].

ORE PROPERTIES

The Chemical compositions of the limonite were shown in table 1.

Table.1: Chemical composition of raw ore /%

TFe	FeO	SiO_2	Al_2O_3	K_2O	Na_2O	CaO	As	Cu	Pb	Zn
38.25	0.077	27.77	1.33	0.057	0.0056	0.17	0.13	0.13	0.055	0.067

As shown in the table, the main useful content was iron, less ferrous content mean that the ore had a weak magnetic. The silica content was higher to 27.77%, in addition, the copper content was relatively high, they may have some influence on the subsequent processing. Other harmful impurities content were very low that had no effect on the iron concentrate quality.

The mineral phase analysis was carried out and the results were listed in table 2.

Table.2: Mineral phase analysis /%

Magnetite	Hematite	Limontite	Botryogen	Pyrolusite	Pyrite	Chalcopyrite	castanite	Quartz	Clay mineral	Kaolinite
1.02	5.13	38.10	10.09	trace	trace	trace	0.70	28.45	2.51	0.27

It can be seen from table 2 that the iron mainly occurred in the form of limonite with a small amount of hematite and magnetite, while the gangue minerals were mainly quartz, botryogen, and clay minerals.

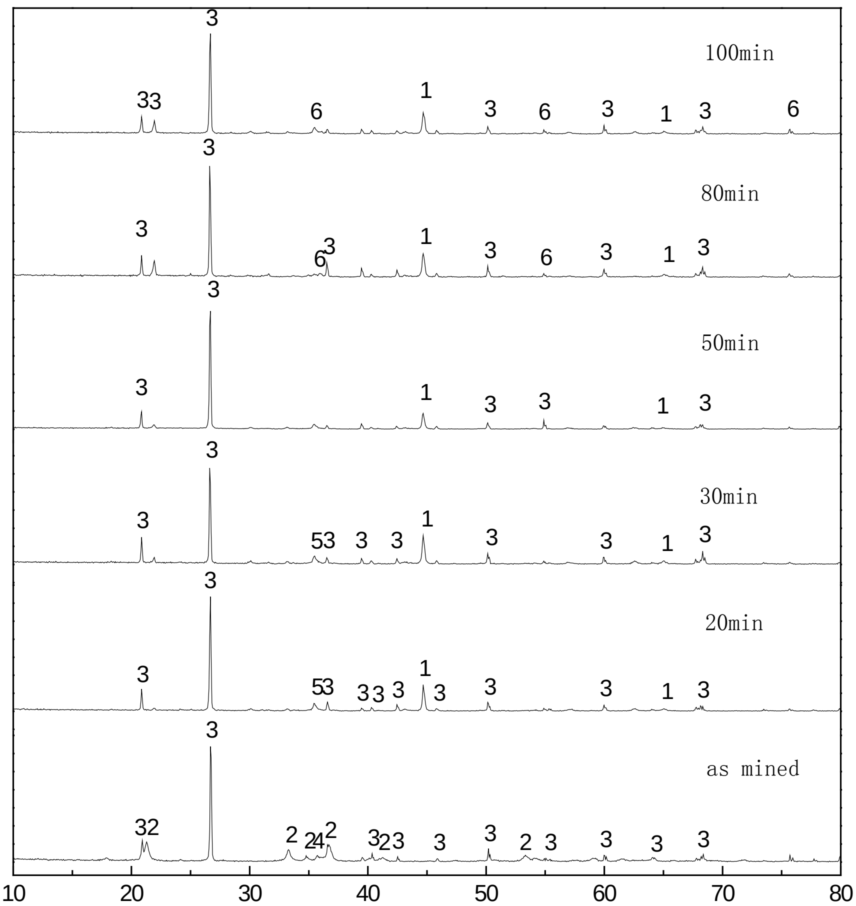
REDUCTION MECHANISM RESEARCH

In order to investigate the phase translation and the morphological changes of iron-bearing minerals at different roasting conditions. SEM with EDS and XRD were used.

1 Effect of roasting time on the reduction of limonite

The effect of roasting time on the reduction of limonite was investigated. The experiments were conducted at a condition of temperature1100°C.

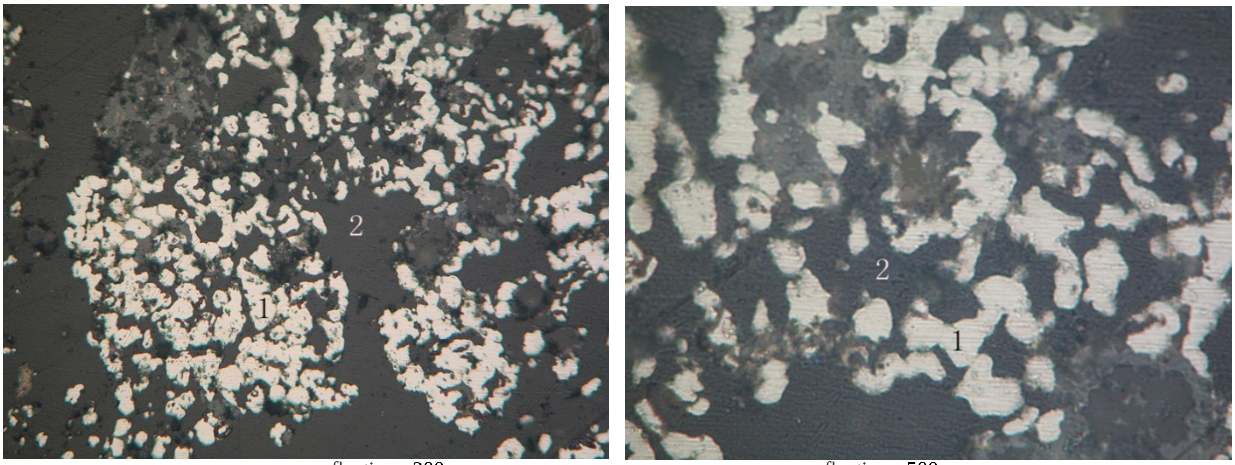
The XRD patterns of the roasted products obtained at different roasting time are shown in Fig.1.



1-metallic iron (Fe);2- goethite(FeO(OH));3- quartz(SiO_2);4- hematite(Fe_2O_3); 5- wustite(FexO);6- fayalite(Fe_2SiO_4);7- magnetite(Fe_3O_4);8- quick lime(CaO)

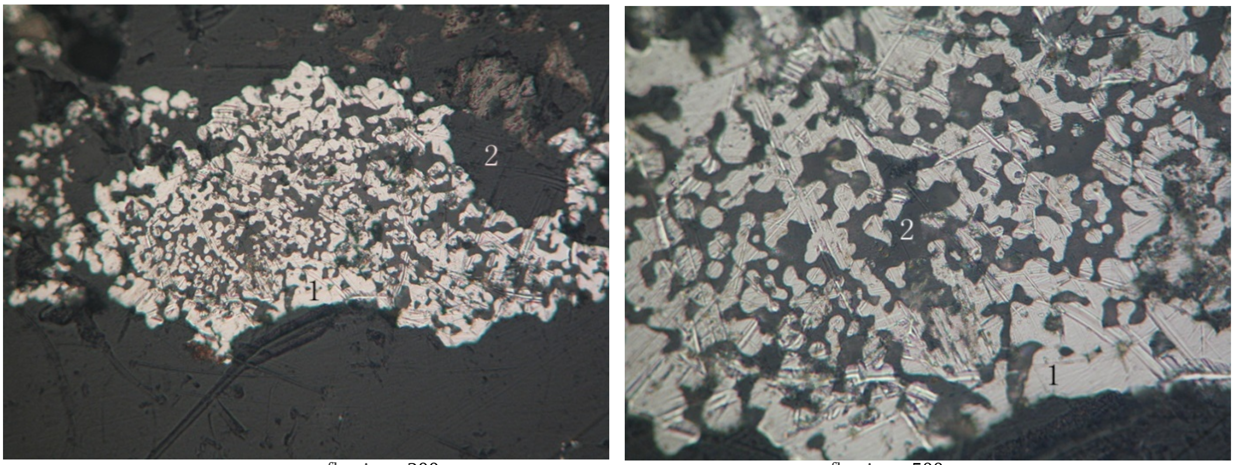
Fig.1:Analysis of calcined under different reduction time XRD

It can be seen from the Fig.1 that, when the reduction time is 20min, the peaks of limonite and hematite are all disappear, and the peak of metallic iron began to form, moreover, peak of wustite was observed. Along with the reduction time increased to 50min, the peak of FeO disappeared, and the Fe peak enhancement. Further increasing the reduction time, a peak of fayalite formed while the peak of Fe decreased, it is may contribute to the fact that the reduction atmosphere weakened as the reduction time increased which resulted in a solid reaction between metallic iron and quartz. Therefore the optimal reduction time is 50min.



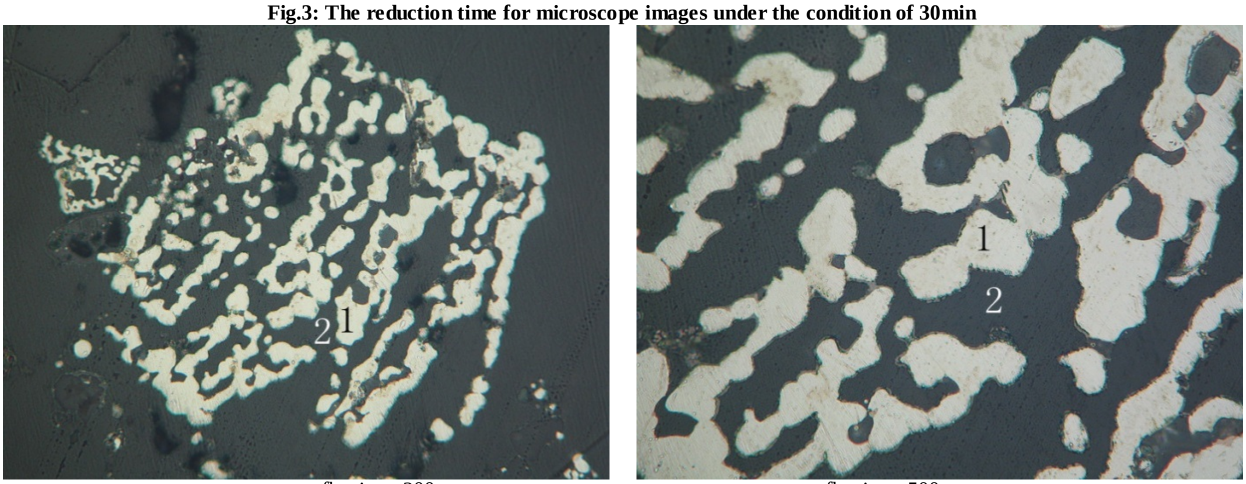
reflection,×200 reflection,×500

Fig.2: The reduction time for microscope images under the condition of 20min



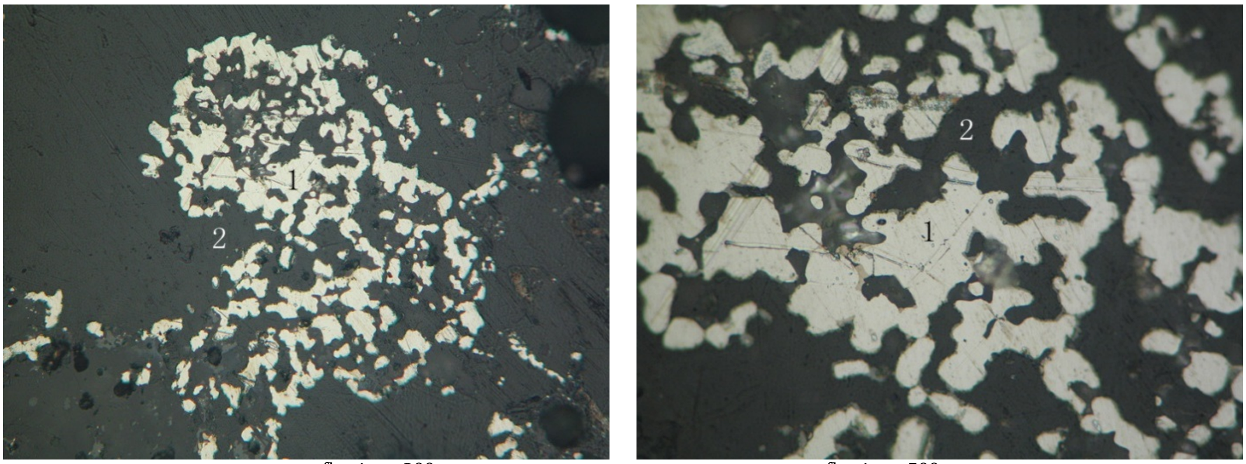
reflection,×200 reflection,×500

Fig.3: The reduction time for microscope images under the condition of 30min



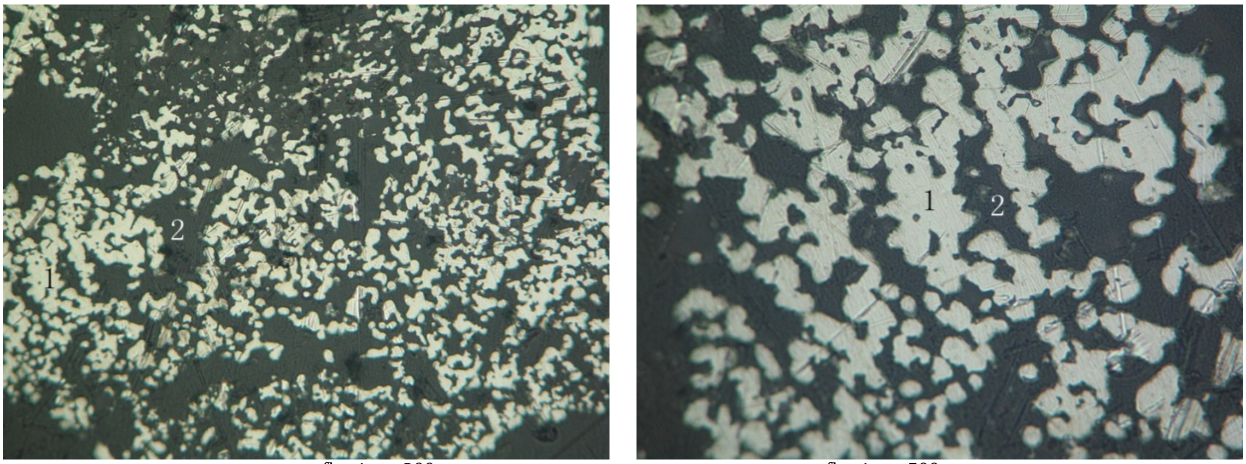
reflection,×200 reflection,×500

Fig.4: The reduction time for microscope images under the condition of 50min



reflection,×200 reflection,×500

Fig.5: The reduction time for microscope images under the condition of 80min



reflection,×200 reflection,×500

Fig.6: The reduction time for microscope images under the condition of 100min

The microscope images of reduced products roasted at different temperature are shown in Fig. 2~6, the write part and the dark part in the reduced products represent the metallic iron and silicate phase, respectively. It is clear that the increase of reduction time is facilitated to the growth and agglomeration of the metallic iron grains. When roasted for 20min, as show in Fig.2, the metallic iron grains were very fine and intimately intermixed with gangue minerals; when the reduction time increased to 30min, the iron grains are coarsened significantly as show in Fig.3; when the reduction time increased to 50min, as show in Fig.4, the metallic iron grains linked to each other, and the boundaries between metallic iron and slag are very clear. When further increasing the reduction time to 80min, however, the particle size of the metallic iron grain decreased, which could be explained that the reduction atmosphere weakened as the reduction time increased which resulted in an oxidation of metallic iron. The optimal reduction time is 50min, this result is agreed with the XRD shown in Fig.1.

2 Effect of roasting temperature on the reduction of iron limonite

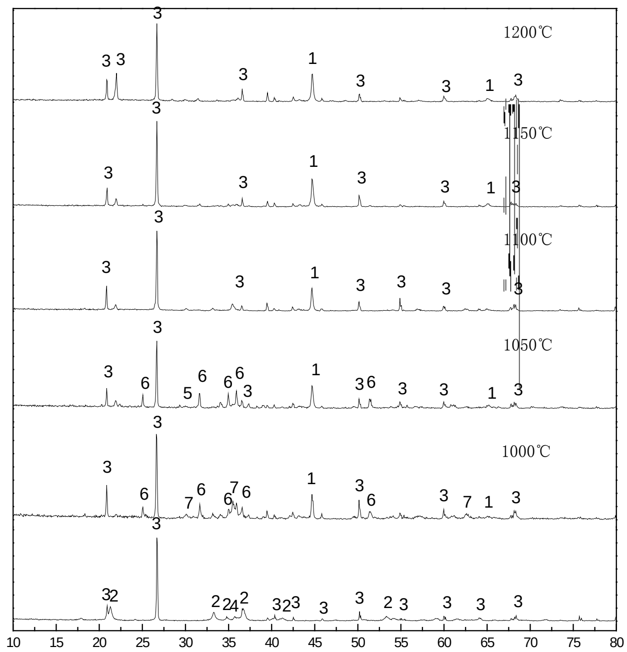
Increasing the temperature can facilitate both the diffusion of gas and the chemical reaction, particularly on the chemical reaction. Direct reduction is a strong endothermic reaction, the reaction speed can be accelerated with the increase of temperature. When the temperature is low, the rate of a chemical reaction is very low is the bottleneck of the reduction process, especially for the reduction of FeO by CO, and the diffusion speed of CO is sufficient to meet the requirements of the reduction process; raising the temperature, the chemical reaction and the reactivity of coal can be improved which in turn promote the reduction of iron oxides and the growth of the iron phase; when the temperature is too high, it is not conductive to the reduction of the iron oxides because it is difficult to provide reducing atmosphere, and iron oxides reaction with quartz to form fayalite and hercynite which has a low melting point and are very difficult to be reduced. On the one hand, these materials melt during reduction and reduces the porosity of the pellet which in turn deterioration of the reduction kinetics; on the other hand, fayalite and hercynite acts as a nucleating agent for the formation of the metallic iron particles in the reduction process, along with the reaction, a metallic iron layer formed on the surface of fayalite and hercynite, and inhibit the contact of reducing agent which in turn makes them very difficult to be reduced, and thereby reducing the iron recovery and iron grade. Therefore the reduction temperature should be controlled within the proper range of temperature, is not the higher the better.

3 XRD Analysis of the reduced products under different reduction temperature

The effect of roasting temperature on the reduction of limonite was investigated. The experiments were conducted at the reduction time of 50min.

The XRD patterns of the roasted products obtained at different roasting temperature are shown in Fig.7.

It can be seen from Fig.7 that, when the limonite was roasted at 1000°C, the peaks of iron minerals in the raw ore disappeared, and peak of fayalite increased, while some of magnetite generated; when the temperature increased to 1100°C, the magnetite and iron olivine peaks gradually disappeared, and metallic iron peak increased; when the temperature increased to 1150°C, the peaks of metallic iron increased. When further increasing the temperature increased to 1200°C, the diffraction peak of metallic iron maintains stability. So the reduction temperature at 1150°C is most appropriate.

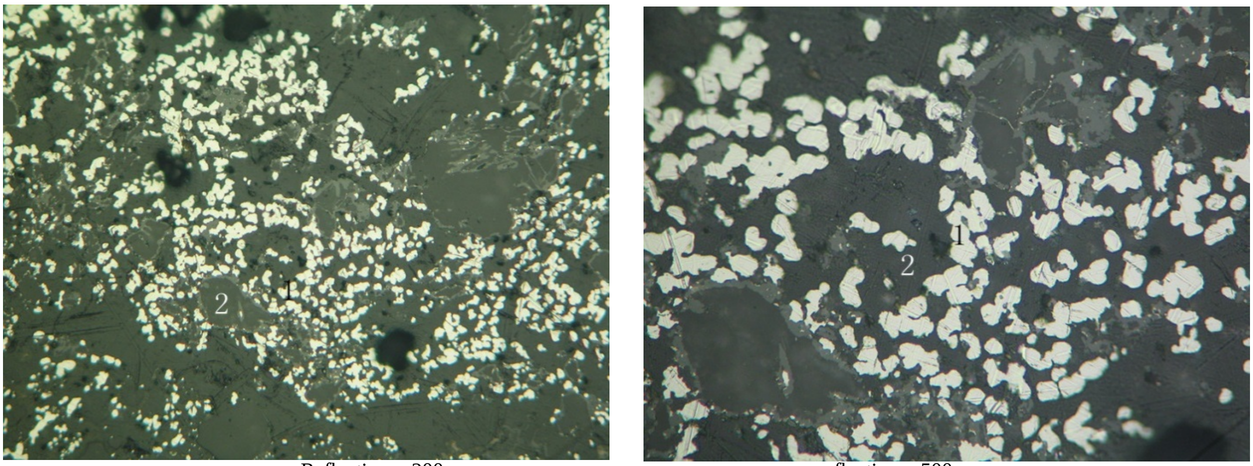


1-Fe;2- goethite(FeO(OH));3- quartz(SiO_2);4- hematite(Fe_2O_3);
5-Fu iron(Fe_xO);6- fayalite(Fe_2SiO_4);7- magnetite(Fe_3O_4);8- quick lime(CaO)

Fig.7: Analysis of calcined under different reduction temperature of XRD

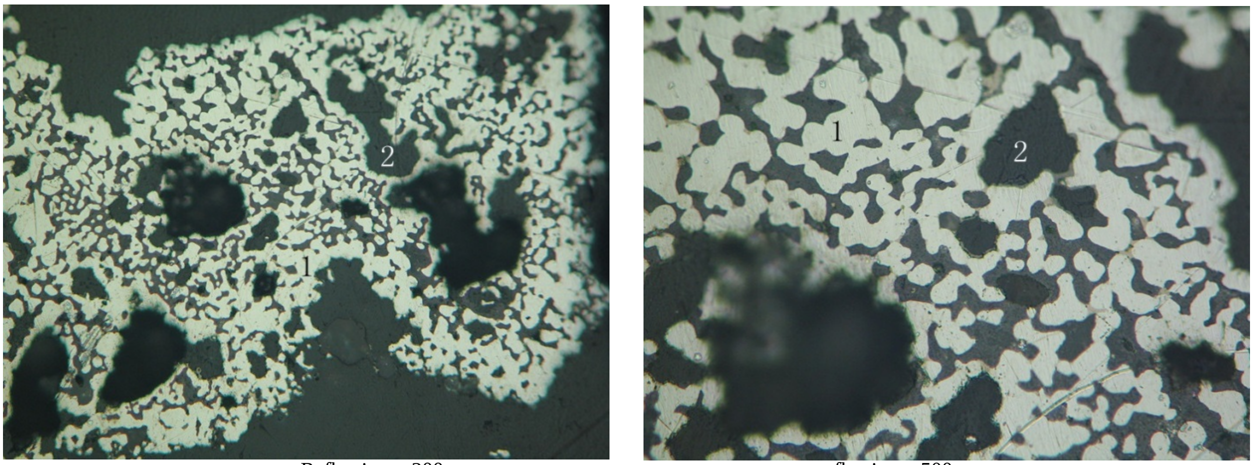
4 Microscope analysis of the reduced products under different roasting temperature

The microscope images of reduced products roasted at different temperature are shown in Fig. 8~12. As can be seen from the pictures, with the increase of temperature, fine particles of metallic iron are constantly merger and grow to form larger grains, some iron particles even connected pieces, the boundary of metallic iron and slag phase is also more obvious. When the reaction temperature is 1200°C, the size of the iron particle hasn't increased when comparing with that in 1150°C, however, but when combine with the test data of magnetic separation that the iron recovery decreased as the reaction temperature increased to 1200°C, indicating that the fayalite was formed at high temperature. So in order to obtain a good separation of metallic iron and slag, the optimal temperature should be 1150°C.



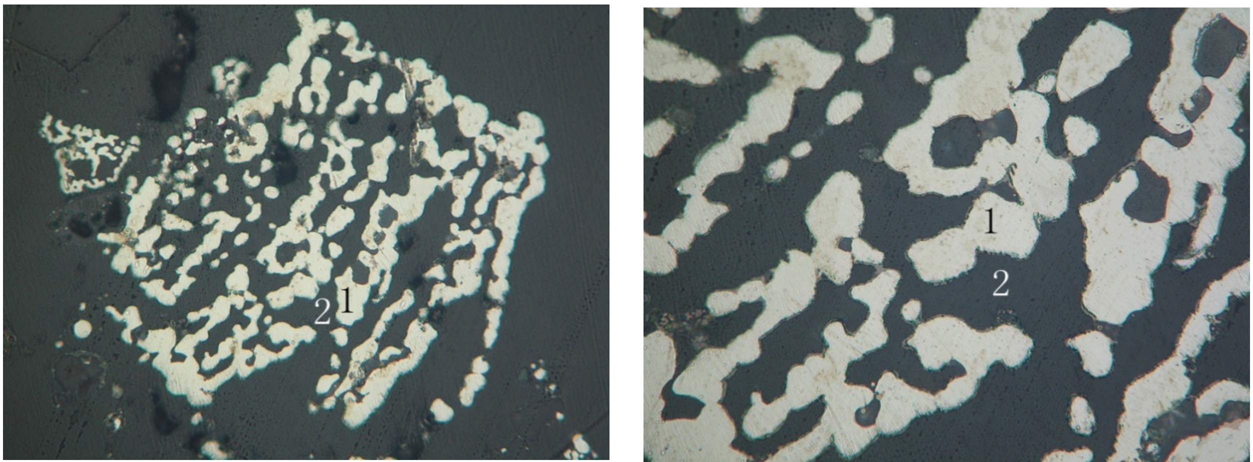
Reflection, ×200 reflection, ×500

Fig.8: Microscope image of the reduction temperature for 1000℃



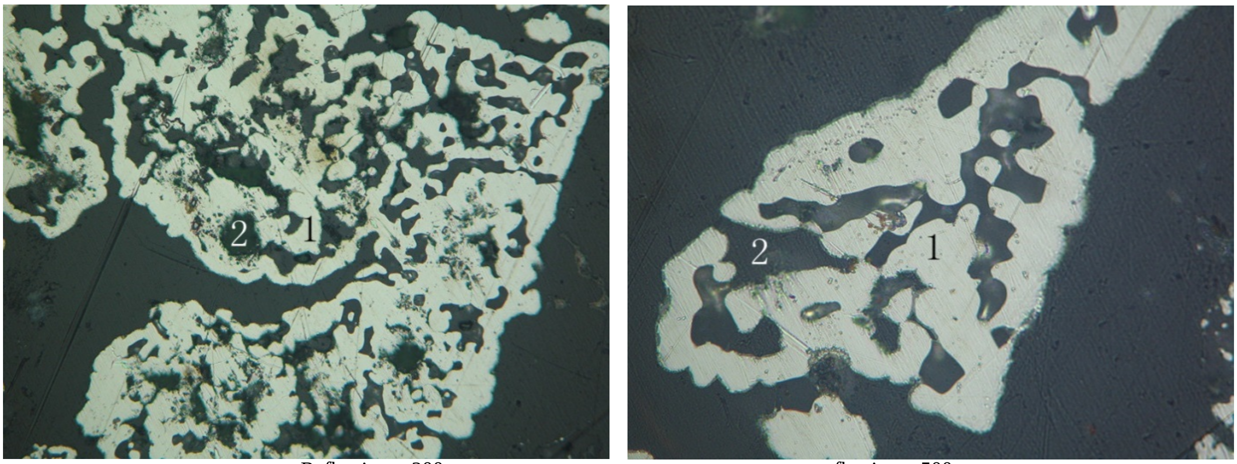
Reflection, ×200 reflection, ×500

Fig.9: Microscope image of the reduction temperature for 1050℃



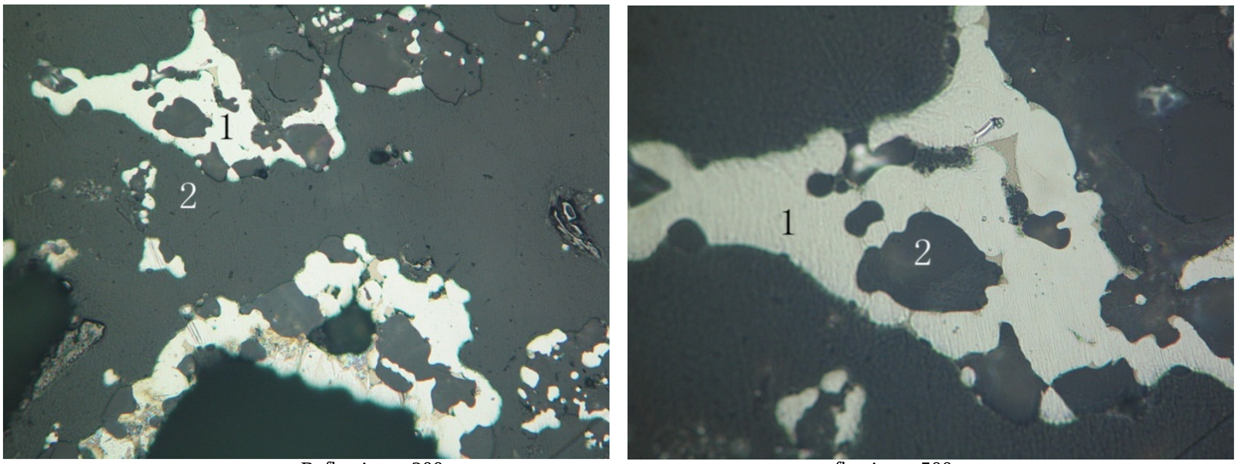
Reflection, ×200 reflection, ×500

Fig.10: Microscope image of the reduction temperature for 1100℃



Reflection, ×200 reflection, ×500

Fig.11: Microscope image of the reduction temperature for 1150℃



Reflection, ×200 reflection, ×500

Fig.12: Microscope image of the reduction temperature for 1200℃

CONCLUSION

1) The reduction time has an important influence on the formation and growth of the metallic iron grains. The reduction time is too short, the reduction of iron oxides is insufficient, and metallic iron grains are fine; the reduction time is too long, the re-oxidation of the metallic iron occurred which resulted in a decrease in the quality of the metallic iron.

2) The temperature has a positive impact on the gas diffusion and chemical reaction. The higher the temperature is, the faster the chemical reaction is. Increasing the temperature can improve reaction activity of coal and facilitate the agglomeration of iron phase; but the temperature should not be too high, too high temperature is not conductive to the reduction of the iron oxides because it is difficult to provide reducing atmosphere, and iron oxide reaction with quartz to form fayalite which has a low melting point, the fayalite melt during reduction and reduces the porosity of the pellet which in turn deterioration of the reduction kinetics. Therefore, the reduction temperature should not be too low and should not be too high.

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