Generation of Fe nanoparticles by He-H₂ arc plasma*

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Fe nanoparticles were generated by DC arc plasma method under atmospheric He-H₂ mixture gas with different H₂ concentration. The arc plasma was well constricted due to thermal pinching when H₂ was mixed. The generation rate of the Fe nanoparticles significantly enhanced as H₂ concentration increased to 40 %, and it was about 0.16 g/min for He-40 % H₂ arc plasma. The vaporization rate of Fe did not significantly increase any more as H₂ concentration exceeds 40 %. Numerical simulation based on magnetohydrodynamics (MHD) was performed and indicated that the Fe anode can be efficiently heated due to the H₂ induced thermal pinching. The maximum anode temperature was calculated close to boiling point of Fe when heated by He-40 % H₂ arc plasma. The specific surface area of Fe nanoparticles produced from He-40 % H₂ arc plasma was about 11.7 m²/g and its corresponding diameter was 65 nm.

Key Words: DC arc-plasma, Thermal pinching, Iron nanoparticles, Generation ratio, MHD numerical simulation

1. Introduction

The DC arc plasma method is a simple and promising route to prepare high-purity nanoparticles with high productivity¹⁻⁵⁾. In this method, anode metal can be melted and evaporated by a localized heating of the thermal plasma, and simultaneously induced plasma jet can rapidly quench the metal vapors, producing metallic nanoparticles.

Ohno and Uda have found that the vaporization rate of the anode metal, that is, the generation rate of metallic nanoparticles can be increased by mixing H₂ into an inert gas of Ar. The vaporization rate of Fe anode metal increased when H₂ concentration in Ar-H₂ mixture increased under atmospheric pressure condition. The similar relation between the generation rate of Fe nanoparticles and H₂ concentration has been widely observed^{1,5)}. We have also obtained the similar tendency⁶⁾, and observed very strong thermal pinching of arc plasma at H₂ concentration of 50 % or more. Our numerical simulation based on magnetohydrodynamics (MHD) indicated that the thermal pinching of arc plasma at relatively high H₂ concentrations can increase efficiently anode temperature to almost boiling point of Fe metal. H₂ is a molecular gas and has higher specific heat (or enthalpy) than Ar, which is the main cause for the formation of thermal pinching under MHD simulation.

In this paper, we report the influence of H₂ concentration in He-H₂ mixture gas on the generation rate of Fe nanoparticles. As well known, He arc plasma has higher maximum arc temperature than those of Ar arc plasma, and thereby He-H₂ arc plasmas will

enhance heat flux density into anode metal, with compared to Ar-H₂ system. The generation rate of metal fumes or nanoparticles in He-H₂ system may also be interested from engineering point of view.

2. Experimental procedure

Fig. 1 shows a schematic illustration of the experimental setup. The apparatus mainly consists of a DC power supply, a water-cooled vacuum chamber, a particle collector, a gas circulator and a vacuum pump. A tungsten cathode of 8.0 mm in diameter having a 60° conical tip was used. An Fe pellet (99.8 %) was placed on the water-cooled copper anode plate. After achieving a base vacuum of 200 Pa, He, H₂ or He-10, 25, 40, 50, 75 % H₂ was introduced. The arc plasma was generated under a total pressure of 101 kPa with a gas flow rate of 120 L/min. Fe nanoparticles or the generated Fe fume was transported by gas

Fig. 1 Schematic illustration of experimental apparatus for arc plasma method.

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circulation flow to the collector chamber. In this study, the arc current of 150 A and arc gap of 10 mm were carefully controlled to be constant during the experiment.

The weight of the Fe pellet was measured to estimate their vaporization rate. The vaporization rate was obtained from the weight loss of the Fe pellet per minute. It was thought that the vaporization rate was almost equal to the generation rate of Fe nanoparticles.

A MHD numerical simulation was also conducted for the present plasmas. The maximum arc temperature, the maximum anode temperature, the maximum concentration of Fe vapor on the anode, and the vaporization rate of the entire melted anode metal were calculated by using unified models which have been developed recently. The following parameters were used for this numerical simulation: an arc current of 150 A, an arc length of 10 mm, an 8.0 mm-diameter tungsten cathode, an Fe pellet anode on a water-cooled copper block, a gas flow rate of 120 L/min, atmospheric pressure. The details of the calculation are given in Ref. 7-10.

3. Results and discussion

Fig. 2 shows a set of photographs of arc plasmas in He, He-10, 25, 40 % H₂. As can be seen, the He arc plasma greatly extended in the direction of the anode. When H₂ concentration increased from 10 to 40 %, the arc column was more and more constricted. In addition, emission from ionized Fe vapor was clearly observed when H₂ was mixed, indicating that the hydrogen plays an important role in the efficient vaporization of the Fe anode.

Fig. 3 shows the relation between vaporization rate of Fe anode and H₂ concentration in He-H₂. The vaporization rate increased significantly as H₂ concentration increased to 40 %. The vaporization rate for 40 % H₂ concentration was about two orders of magnitude larger than that for 100 % He arc plasma. On the other hand, the vaporization rate remained almost constant or less for H₂ concentrations exceeding 40 %. The specific surface area of Fe nanoparticles produced from He-40 % H₂ was about 11.7 m²/g and its corresponding diameter was 65 nm.

Fig. 4 shows the two-dimensional distributions of the arc plasma temperature and concentration of Fe vapor calculated by numerical MHD simulations for He, He-25 % H₂ and He-40 % H₂. As can be seen, high temperature region got constricted with increasing H₂ concentration. Fe vapor is slightly distributed in Fe anode metal in the He arc plasma. When H₂ is mixed, Fe vapor concentration on the anode markedly increases with increasing hydrogen concentration, and Fe vapor is expanded out in the radial direction.

Here, we discuss a possible mechanism of the H₂ assisted Fe

Fig. 2 Photographs of 150A arcs in He, He-10% H₂, He-25% H₂ and He-40 % H₂.

Fig.3 Relation between vaporization rate of Fe and H₂ concentration in He-H₂.

vaporization from point of macroscopic consideration of MHD. Table 1 summarizes the maximum anode surface temperature, the maximum concentration of Fe vapor and the maximum vaporization rate of Fe obtained by numerical simulation for He, He-25 % H₂, He-40 % H₂, He-50 % H₂ arcs. The estimated maximum anode surface temperature reached the melting point (1808 K) of Fe in the He arc, and increases as H₂ concentration increases. By the He-40 % H₂ arc, the anode surface temperature almost reaches the boiling point of Fe (3160 K). This is obviously resulted from increase of the heat flux density due to hydrogen induced thermal pinching. The present simulation also shows that the

Fig.4 Two-dimensional distributions of arc temperature and mole fraction concentration of Fe vapor by calculation at He, He-25 % H₂ and He-40 % H₂.

maximum vaporization rate of Fe increases significantly as H₂ concentration increases from 0 % to 40 %, and the vaporization rate for 40 % H₂ concentration is about two orders of magnitude larger than that for 100 % He arc plasma. Thus, the calculated dependence of H₂ concentration on the vaporization rate is good agreement with the experimental observation, and it may be concluded that the vaporization from Fe anode metal could be enhanced by efficiently heated surface due to the hydrogen induced thermal pinching. The thermal pinching effect strongly depends on specific heat (or enthalpy) of gas. The specific heat of H₂ is the highest among conventionally used gaseous species, and thereby the arc plasma is mostly constricted under H₂ mixing. The arc constriction leads to increase of arc current density, and the greater ohmic heating gives rise to increase arc temperature, which enhances the heat flux density into anode, i.e., metal vaporization from anode. For the He-50 % H₂ arc, the maximum calculated temperature of the anode exceeds the boiling point of Fe which is due to the fact that latent heat of Fe vaporization is not considered in the present numerical simulation.

Fig. 5 shows the relation between the calculated vaporization rate of Fe and H₂ concentration in the He-H₂ arcs. The calculated vaporization rate well increases as H₂ concentration increases from 0 % to 40 %. The calculated vaporization rate for 40 % H₂ is

about two orders of magnitude larger than that for 100 % He arc plasma. The result of the present calculation and the experimental result (Fig.3) show similar tendencies. In case of Ar-H₂ arc plasma, the significant increase of vaporization rate was observed as H₂ concentration increased to 50 %, and its tendency was good agreement with the result of calculation⁸⁾. Also, the calculation showed that the maximum anode temperature reached almost boiling point of Fe for Ar-50 % H₂ arc plasma. Since He has larger specific heat than Ar, more efficient arc heating may be produced by H₂ mixing with the lower concentration.

Table 1 Summary of the calculated arc properties in He, He-25 % H₂, He-40 % H₂ and He-50 % H₂ arcs.

	Max. temp. of molten pool (K)	Max. conc. of Fe vapor on anode (mol%)	Max. vaporization rate (g/cm ² s)
He	2583	2.13	4.9x10 ⁻³
He-25 % H ₂	2902	6.84	7.4x10 ⁻²
He-40 % H ₂	3151	9.87	3.0x10 ⁻¹
He-50 % H ₂	3201	7.44	4.4x10 ⁻¹

Fig.5 Relation between calculated vaporization rate of Fe and H₂ concentration in He-H₂

The vaporization rate of Fe does not significantly increase any more even if the H₂ concentration exceeds 40 %. As shown in Fig.3, Fe vapor was distributed on anode for He-40 % H₂ arc plasma, and high concentrated space of Fe vapor was laterally broadened widely than the arc column. In the present MHD simulation, spatial distribution of Fe vapor on anode was taken into consideration. Base on this calculation, the maximum vaporization rate of Fe for 50 % H₂ is a little larger than that for 40 % H₂, as shown in Table 1. Therefore, the spatial distribution of concentrated Fe vapor formed on anode plays an important role on heating efficiency of anode, especially for H₂ concentration exceeding 40 %.

4. Conclusions

The vaporization rate of Fe anode metal, that is, the generation rate of Fe nanoparticles increased exponentially as H₂ concentration increased to 40 % in He-H₂ mixture gas. It increases gradually or less for H₂ concentrations exceeding 40 %. These tendencies are in agreement with the MHD numerical simulation with taking account of special distribution of Fe vapor formed on anode. According to the present MHD simulation, the enhanced vaporization from Fe anode is due to efficiently heated surface of the anode which relates to hydrogen induced thermal pinching, for He-H₂ arc plasma with H₂ concentration < 40 %. When H₂ concentration exceeds 40 %, concentrated and broadened Fe vapor formed on anode may influence the arc plasma state and its heating efficiency.

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