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Dynamic study of changes in structure and morphology during the heating and sintering of iron powder

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Abstract

Dynamic changes in structure and morphology during the heating of iron powder to high temperatures have been studied in situ in an environmental scanning electron microscope (ESEM). The results have shown that the presence of lubricants leads to the formation of ferrite needles at about 500 °C. At higher temperatures, the recrystallization of α-iron, followed by the nucleation and growth of γ-iron and finally the sintering process leading to the reduction of porosity and grain growth, were continuously monitored. This technique could be useful in the simulation and optimization of powder processes at elevated temperatures. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Iron powder; Sintering; Microstructure; Hot stage; ESEM

1. Background

Iron and steel powder metallurgical components have been commercially produced for many years and the developments in powder technology have been impressive. Today, the performance of components made from powder metallurgical methods compare favourably with those made from conventional forging techniques. Warm compaction of powders has made it easier to produce high-density, high-performance products. Progress has also been made in achieving better dimensional tolerances after sintering the compacts.

Sintering of iron powders involves a series of important steps—particle bonding at the area of contact and the growth of these bonds, grain growth and the migration of the grain boundaries formed at the bonds, spheroidization of the pores between the particles and the elimination of small pores leading to a possible increase in the growth of the larger pores. The formation of bonds is resisted by residual material from the lubricant, by impurities and surface oxides and by poor contact.

The characterization of the powders, their compaction and sintering requires the use of metallographic methods. Such studies can reveal the particle configuration, uniformity of mixing, interparticle porosity and its distribution, degree of particle bonding, the microstructure and the degree of diffusion alloying, etc. Conventional metallographic methods employ the ex situ method wherein the sample is cooled from a high temperature and examined under the microscope at room temperature. Such a method has the obvious disadvantage that while the specimen is cooled and transferred to the electron microscope, structural changes can occur and the possibility for continued reaction also exists. In other words, the microstructure seen in the microscope can be different from that under the actual experimental conditions. A good compilation of the microstructures of powder metallurgy materials obtained by the ex situ method can be found in [1].

From the preceding section, it follows that dynamic studies ('in situ') involving the direct observation of changes in structure and morphology of iron powder particles during their heat treatment could be invaluable in getting a better understanding of the processes occurring at high temperatures. Such studies are not reported in the literature.

Dynamic studies involving the use of environmental cells/heating stages in electron microscopes have been reported in literature, but they are not very extensive. The transmission electron microscope has been used to study, among others, the oxidation of copper [2], nickel and its alloy [3], and the decomposition of iron–carbon austenites [4]. Very thin samples were used for these TEM studies.

The conventional scanning electron microscope has also been used for dynamic studies using an environmental cell

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and a heating stage. Using bulk samples, Raynaud and Rapp [5] observed the formation of whiskers, pyramids and pits during the high-temperature oxidation of metals, at relatively low pressures in the sample chamber. However, the advent of the environmental scanning electron microscope (ESEM) has led to the possibility of using a variety of gaseous atmospheres in the sample chamber, at pressures of about 2500 Pa (maximum). Brown et al. [6] have reported their observations on the evolution of microstructures in compacted ceramics and composites. Yeh et al. [7] studied the combustion of boron particles in order to clarify the diffusion mechanism of reaction.

In the present work, dynamic studies have been made and the structural changes occurring during heating and sintering of iron powder particles have been monitored continuously, using an environmental scanning electron microscope equipped with a heating stage.

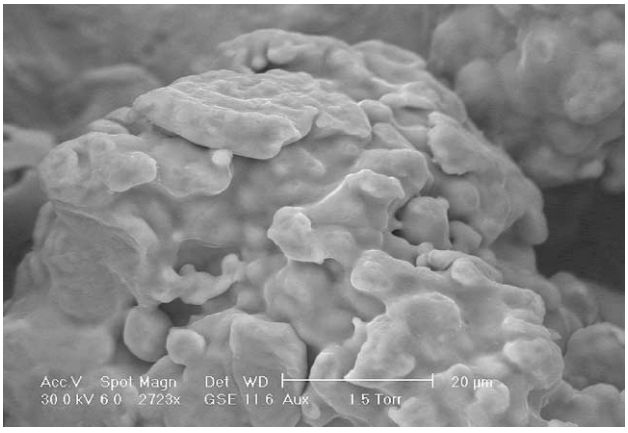

Fig. 1. SEM-image of an iron powder sample at Room Temperature.

2. Experimental

2.1. Material studied

High-purity atomized iron powder (ASC100.29) + 0.6% lubricant (wax) (supplied by Höganäs).

2.2. Microscopy

Philips XL-30 ESEM has been used in the present study. The microscope has an LaB_6 filament and an ultimate resolution of 3.5 nm. The specimen chamber is relatively large (284-mm diameter) and the use of a differential vacuum system allows the maintenance of a gaseous atmosphere in the chamber, with a maximum pressure of about 2600 Pa. Partial ionization of the gas molecules due to the electron bombardment is helpful and obviates the need for coating specimen surfaces with gold or any other conducting material. Specimen surfaces in their natural condition can thus be studied. The gaseous secondary electron detector (GSED) has been used for this purpose.

The specimen stage in the chamber is motorized for convenience in the positioning of the sample. An infrared camera allows the inspection of the chamber, if and when it becomes necessary.

2.3. Heating assembly

On the specimen stage is mounted the heating stage assembly. Thick copper plates, with criss-crossing holes for effective water cooling, form the exterior of the L-shaped assembly (57-mm long × 35-mm height × 25-mm wide). At the heart of the assembly is a spiral formed sample heater (4.6-mm diameter × 7-mm height), made from tungsten or kanthal wire. The spiral heater is supported by a hollow aluminium oxide cylinder, which in turn is covered with a hollow copper cylinder.

An alumina-sheathed, Pt–10%Rh/Pt thermocouple is centered vertically inside the spiral heater. A small platinum cup is loosely attached so that the thermocouple bead is at the center. The powder sample to be studied is placed in the platinum cup.

The spiral heater assembly is covered by an alumina lid (with a 4-mm hole), on top of which is placed a copper lid with a 3-mm central hole in it.

Table 1
Experimental details

Test date	2000-02-01
Sample	iron powder (ASC100.29) + 0.6% lubricant (wax)
Atmosphere	nitrogen–4% hydrogen
Treatment	sintering at 1120 °C

2.4. Procedure

The GSED detector with the wire-hook adapter is mounted in place. The powder sample is carefully placed in the platinum cup surrounding the thermocouple bead and the alumina and copper lids are placed in position. The heating assembly is carefully mounted on the specimen stage, ensuring that there is sufficient clearance between the copper lid and the bullet above. Water cooling is turned on and the connections to the heater as well as the thermocouple are checked. The supply of the required gas to the specimen chamber is ensured by checking the gas line. A low gas flow rate is employed and pressure build up in the gas line is avoided by providing a by-pass line.

The specimen chamber door is closed and the chamber pressure is lowered to about 100 Pa. At this stage, the chamber is flushed with the specified gas (N_2–4% H_2 mixture in this study) and the chamber pressure is allowed to rise to 1000 Pa. This pressure is once again lowered to 100 Pa and the procedure is repeated four to five times to ensure that the air in the chamber is completely replaced by the specified gas.

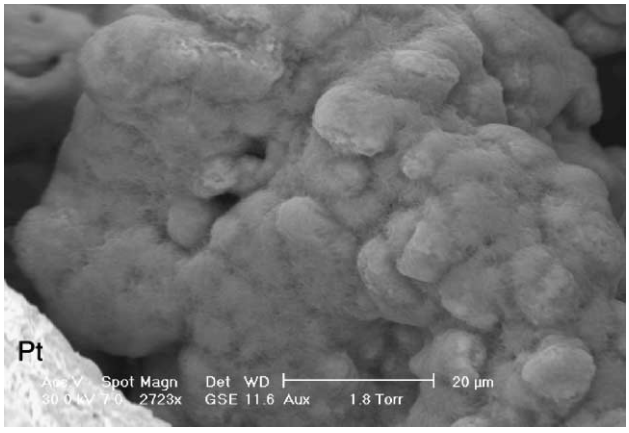

Fig. 2. SEM-image of the iron powder sample at 538 °C (30 min).

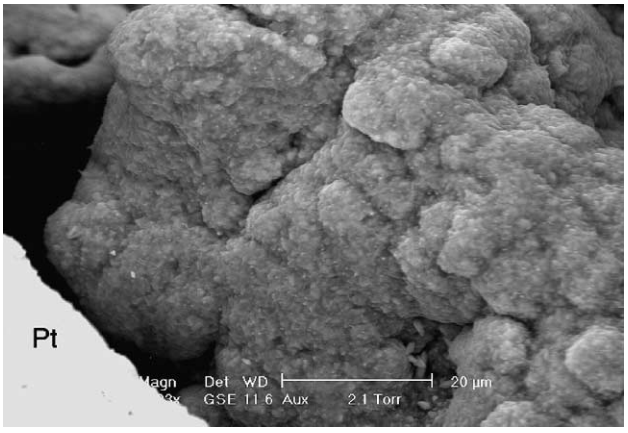

Fig. 4. SEM-image of the iron powder sample at 731 °C (45 min).

The electron beam is now switched on to get an image of the sample at low magnification. The area around the thermocouple bead is scanned with the help of the motorized stage and a particle on the bead or in contact with it is identified for study. Now the image of this particle is optimized at high magnification by finding a suitable combination of parameters. The gas pressure in the specimen chamber is also an important parameter and a value in the range of 350 to 400 Pa has been found to be suitable in the present study.

The optimized image is now saved in a digital format and represents the room temperature image. The power supply to the heater is now turned on and rate of heating is maintained at about 5 °C per minute in the first 20 min and gradually increased to over 15 °C per minute later on. The heating rates can be relatively high in temperature intervals showing no structural changes. On the other hand, in temperature ranges showing structural changes, it is important to note that while saving images at low scanning speeds, the structure could change before the scan is completed, especially for high heating rates.

During the heating process, due to thermal expansion, etc., image drift is very common and at high magnifications this could lead to losing sight of the particle being studied. So, continuous adjustments are required to keep the particle under study in the middle of the screen and get an optimized image. The increase in temperature of the sample invariably leads to a loss of contrast in the image, which could be compensated by choosing a higher spot size (if possible) and/or marginally increasing the gas pressure in the specimen chamber.

Keeping in mind what has been described in the previous sections, images are stored at different temperature and time intervals during the heating and sintering of the iron powder sample.

3. Results and discussion

The experimental details are given in Table 1.

Fig. 1 shows the untreated sample at room temperature and Figs. 2–8 show the structural changes in the sample

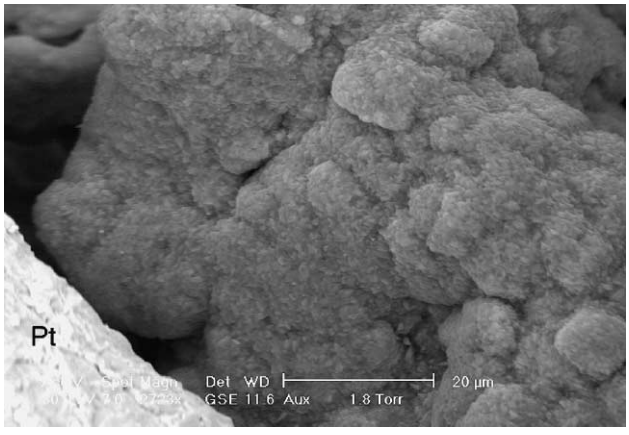

Fig. 3. SEM-image of the iron powder sample at 681 °C (37 min).

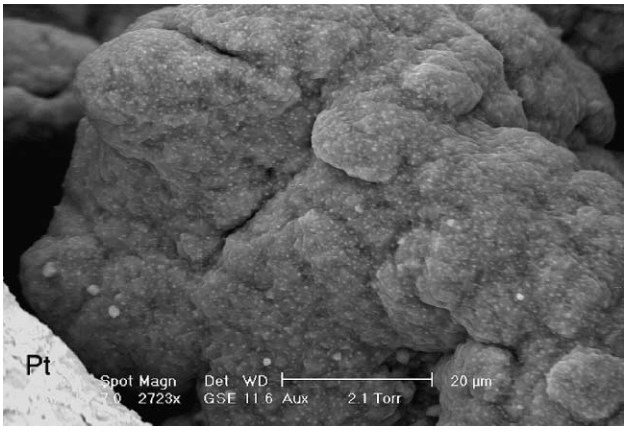

Fig. 5. SEM-image of the iron powder sample at 912 °C (58 min).

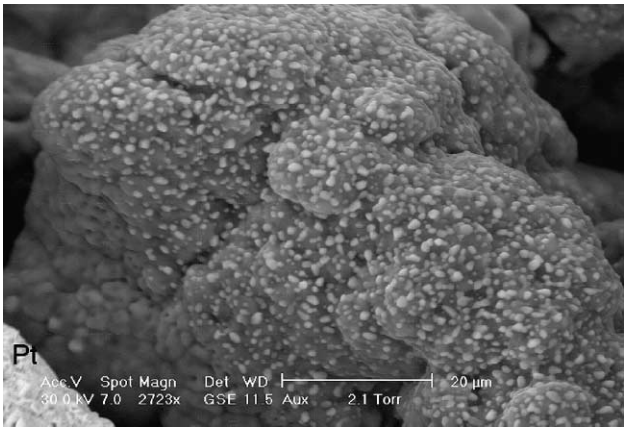

Fig. 6. SEM-image of the iron powder sample at 1050 °C (71 min).

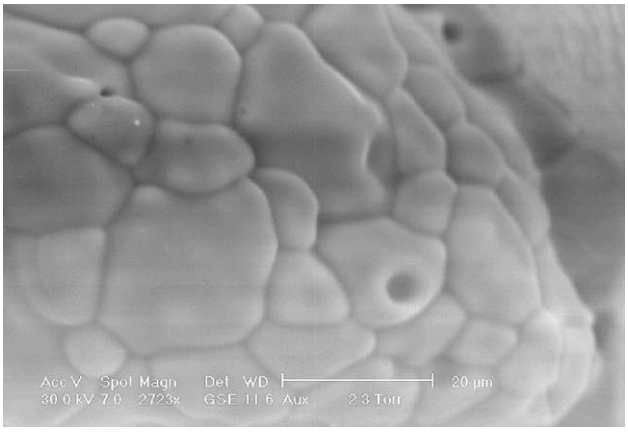

Fig. 8. SEM-image of the iron powder sample at 1123 °C (115 min) (33 min at the sintering temperature).

when it is heated to 1120 °C and subsequent sintering at that temperature.

The morphology of the iron particles shown in Fig. 1 is typical for an atomized powder.

During heating, no changes in the sample were observed up to about 500 °C. Around this temperature, a needle-like structure began to evolve. This is shown in Fig. 2. The development of this structure is probably due to a vapor phase transport process associated with the vaporization of the organic phase (lubricant) present in the powder sample. This was confirmed by experiments conducted with samples not containing the lubricant. In these samples, the needle-like structure was not observed during heating. The effect of the lubricant was similar even in the case of low alloyed iron powders.

As the temperature is further increased further, recrystallization takes place leading to the build up of a new set of small grains (Fig. 3). While these grains grow in size with increase in temperature and time (Fig. 4), the $\alpha \rightarrow \gamma$ transition temperature is approached. The image obtained at 912 °C shows the nucleation of γ-iron (Fig. 5). The recrystallization process continues with further heating and the growth of these nuclei is shown in Fig. 6 (1050 °C). Fig. 7 shows the early stages of sintering at 1122 °C. Prolonged sintering at these temperatures leads to a drastic reduction in porosity and grain growth. This is seen in Fig. 8.

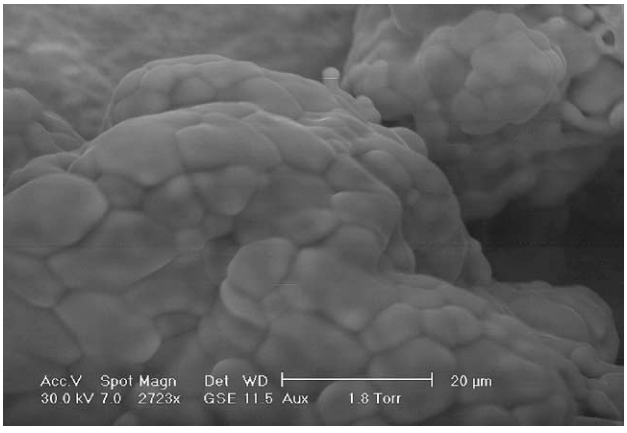

Fig. 7. SEM-image of the iron powder sample at 1122 °C (87 min) (5 min at the sintering temperature).

4. Summary and conclusions

In this study, an environmental scanning electron microscope, equipped with a heating stage, has been successfully used to study in situ structural changes associated with powder processes at high temperatures. The study has shown the formation of ferrite needles when iron powder is heated to about 500 °C. This is associated with the vapor phase transport of material caused by the escaping lubricant phase. At higher temperatures, recrystallization phenomena associated with α- and γ-iron as well as the progress of sintering at about 1120 °C could be monitored continuously. The results confirm the suitability of the technique for the simulation and optimization of high-temperature processes involving powdered materials.

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