Synthesis and characterization of a model dual-phase system using the spark plasma sintering technique

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Abstract. Samples of a model dual-phase system, consisting of copper and AISI-420 martensitic steel, have been synthesized using spark plasma sintering, with the objective of developing a microstructural analogue for dual-phase steels, in which the volume fraction and size of each phase can be controlled independently. Microstructural investigation of the samples, including fractography of samples deformed in tension until failure, show that densification is strongly temperature dependent. Samples sintered at temperatures of 900 °C or above at a pressure of 60 MPa show a density of more than 98%. The best mechanical properties, in terms of ultimate tensile strength and ductility are found in samples sintered at a temperature of 1000 °C, where a density of nearly 99% is achieved.

1. Introduction
There is much interest at present in the relationship between microstructure and mechanical properties in composite metallic systems consisting of hard and soft phases. One system that has been explored in particular detail is that of dual-phase steels, where a typical microstructure consists of ferrite, as the soft phase, and martensite as the hard phase [1-3]. These materials have been widely explored for use in automotive applications, as they show higher ultimate strength, and higher work hardening rate compared to high-strength low-alloy steels (HLSA), while also demonstrating improved formability and ductility compared to their HLSA counterparts [4].

Some important challenges nevertheless remain with regard to a detailed understanding of the relationship between microstructure and mechanical properties in such dual phase microstructures. One problem concerns the difficulty in exploring the influence of various microstructural parameters as in general it is difficult to control independently using standard thermomechanical processing the size and volume fraction of the martensite phase, and even more so to control the spatial distribution (uniformity or heterogeneity) of the two phases. Moreover, in many cases the microstructures of commercial dual phase steels are too complicated to allow the basic mechanisms controlling deformation to be easily investigated [5].

A related problem is that diffraction-based measurements, which have been used to probe the partitioning of stress and strain between the hard and soft phases, are complicated by the fact that the ferrite and martensite phases have a similar crystal structure with similar lattice parameters. As a consequence, the diffraction peaks from each phase overlap and analysis of the data requires advanced peak fitting methods where certain assumptions about the peak shapes and distribution of each phase are required [5-7].
An alternative approach to develop a model system for fundamental studies of the mechanical behaviour of dual phase systems involves the use of the spark plasma sintering (SPS) technique, sometimes also referred to as field-assisted sintering [8]. This technique is a modified hot powder pressing technique, in which a pulsed electrical current and a uniaxial pressure are applied to a powder compact at low voltage. The current passes through an electrically conductive graphite die and heats the sample. The ability of this technique to produce fully dense bulk material within a very short time and at a relatively low temperature without any secondary processing, offers many advantages compared to other powder metallurgy processes and makes it a cost-effective process, in particular for materials that are highly sensitive to temperature [9]. Although the spark plasma sintering technique was originally developed for ceramics it has also been applied for metals, such as nickel [10,11], copper [12-14], aluminium [15-17], pure iron [18], and a range of steels [3,19].

In this paper, we explore the possibility of using spark-plasma sintering for the preparation of a model dual-phase system by mixing of suitable powders. An advantage of this approach is that it allows independent control of both the size and the volume fraction of the soft and hard phases (and by control of the powder mixing process, their distribution). For this study copper is used as the soft phase, with a martensitic stainless steel as the hard phase.

2. Experimental

Spherical powders of copper (average size either 3.5, 10.0, or 15μm) and AISI420 stainless steel (average size of 10 or 15μm) were used to prepare samples (for details see table 1). The powders were mixed using a low-energy mechanical milling process. This was carried out using a horizontal ball milling system at room temperature, using stainless steel balls and a polymer vial. For this study samples of copper and AISI420 stainless steel powder were combined in weight ratio of either 50:50 or 70:30. For ball-milling the ball to powder mass ratio was set at 20:1 and filling was performed in an air atmosphere. The powder mixtures were milled for 24h at a constant rotation speed of 120 rpm.

| Cu (Fe) powder size (μm) | Cu:Fe wt. ratio | Sintering temperature (°C) | Sintering pressure (MPa) | Heating ramp parameters (Q, T₁, t) | Relative density (%) |
|--------------------------|-----------------|---------------------------|--------------------------|-----------------------------------|----------------------|
| 15 (15)                  | 50:50           | 850                       | 50                       | 80, 850, 0                        | 88.6                 |
| 10 (10)                  | 50:50           | 900                       | 50                       | 100, 850, 1                       | 95.0                 |
| 3.5 (10)                 | 70:30           | 900                       | 60                       | 100, 850, 1                       | 98.0                 |
| 3.5 (10)                 | 70:30           | 950                       | 60                       | 114, 720, 5                       | 98.1                 |
| 3.5 (10)                 | 70:30           | 1000                      | 60                       | 114, 720, 8                       | 98.9                 |

*Units of Q (°C/min), T₁ (°C), t (min) - see figure 1 for definition of these parameters.

Consolidation of the mixed powders was carried out using the SPS process (SPS-1050, Sumitomo Heavy Industries Ltd., Japan). The powders were first loaded into a graphite die (20 mm diameter). The die was separated from the mould and punches of the SPS system using graphite foils, to prevent welding from powder during the sintering process. Sintering was performed under primary dynamic vacuum and under the application of uniaxial pressure. The temperature, pressure and dwell time were varied over a wide range to investigate the influence of these SPS parameters on the samples density and microstructure. The general heating and loading cycles of the SPS process are shown in figure 1 (table 1 gives the relevant values for each sample studied). The final sintered samples were disc shaped, with a diameter of 20 mm and a height of approximately 4 mm. After sintering the compacted samples were given a heat treatment in order to control the martensitic transformation of AISI420 stainless steel phase. For this the samples were heated to 1000 °C at a heating rate of 10 °C/min and held at this temperature for 15 min before quenching in oil. The samples then were tempered in an air-furnace at 500 °C for 30 min.
temperature - 60 MPa sintering pressure (figure conditions a number of the.

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3. Results and discussion

The final sample density was determined using Archimedes’ method after mechanical grinding and polishing of all sample surfaces. The microstructure of the sintered samples was characterized using a scanning electron microscope (SEM), including elemental mapping using energy dispersive spectroscopy (EDS) to investigate the phase distribution of the samples. Mechanical characterization of the samples was carried out using Vickers hardness measurements (100 g load) and tensile tests of dog-bone shape samples (gauge length of 8 mm, width of 2 mm and thickness of 1 mm) cut from the disks using spark-erosion. Hardness measurements were obtained in the copper and AISI420 phases separately by control of the indent locations. Values are given as the average and standard deviation over at least 7 measurements in each phase. The tensile tests were carried out at room temperature under displacement control using an initial strain rate of 6.3 x 10^-4 s^-1.

Figure 1. Schematic heating and loading cycles for the SPS process. After an initial heating ramp (heating rate \( Q \) °C/min) to temperature \( T_1 \), the final sintering temperature was approached over time \( t \) (mins). The sintering pressure was applied only during a 5 min hold at the sintering temperature \( T_s \).

The measured densities of samples prepared under a selected range of conditions are given in table 1. In agreement with previous studies [14,16] the results show that the sample density increases both with increasing holding pressure and with increasing sintering temperature. At 850 °C and 50 MPa sintering pressure a resulted in density of only less than 90%. Raising the sintering temperature to 900 °C at the same pressure resulted in improvement of the density to approx. 95%. With additional increase of pressure to 60 MPa at this temperature a density of > 98% was achieved. Further increase of temperature to 1000 °C resulted in a moderate improvement of density to almost 99%.

Figure 2(a-d) shows SEM images of samples sintered under a range of conditions. In these images, the lighter continuous phase is the copper and the darker discrete regions are the AISI420 stainless steel, as illustrated by the elemental EDS maps shown in figure 3 for the sample prepared at 900 °C and 60 MPa. The results of the SEM observations provide microstructural support for the density measurements. As seen in figure 2a, large-scale porosity is present in the sample sintered at 850 °C and under a pressure of 50 MPa. Additionally, extensive fine-scale porosity is present, both between original iron particles, and at the interface with the copper phase. Increasing the temperature to 900 °C at the same sintering pressure (figure 2b) results in a sample without large-scale porosity, although extensive fine scale porosity remains, in particular at the interface region between the AISI420 stainless steel phase and the copper phase. Under these sintering conditions it is found that most of the AISI420 stainless steel particles retain their original round shape after the combined ball-milling and sintering processes. Under SPS conditions of 900 °C and 60 MPa sintering pressure (figure 2c) the interfacial porosity between the copper and AISI420 stainless steel phases is significantly reduced, although a small amount of porosity is still seen in the samples. Furthermore, under these sintering conditions a number of the AISI420 stainless steel particles are deformed and no longer maintain their round shape.
Figure 2. Example SEM images of samples sintered at different temperatures (T) and pressures (P): (a) T = 850 °C, P = 50 MPa; (b) T = 900 °C, P = 50 MPa; (c,d) T = 900 °C, P = 60 MPa.

Figure 3. Elemental EDS maps of the sample sintered at 900 °C and 60 MPa showing AISI420 stainless steel particles embedded in a continuous copper phase.

The results of the hardness measurements of the sintered samples are presented in table 2. The data show a non-systematic variation with sintering temperature. It can be seen that with increasing the sintering temperature from 850 °C up to 900 °C, the hardness of the both copper and AISI420 stainless steel phase increase, with a large increase in the hardness of the AISI420 stainless steel phase. Similarly, the specimen sintered at 900 °C and 60 MPa shows a somewhat higher hardness in both phases than for the sample sintered at the same temperature but a lower pressure of 50 MPa. With increasing the
sintering temperature up to 1000 °C the hardness of the AISI420 stainless steel phase decreases, although the reason for this decrease is at present unknown (further microstructural investigations, including determination of the grain size within both phases is currently underway).

| SPS temperature (°C) | SPS pressure (MPa) | Hardness (HV) | Ultimate tensile strength (MPa) |
|----------------------|-------------------|---------------|-------------------------------|
| 850                  | 50                | 138 ± 6       | 409 ± 56                      |
| 900                  | 50                | 126 ± 2       | 528 ± 45                      |
| 900                  | 60                | 159 ± 13      | 537 ± 64                      |
| 950                  | 60                | 154 ± 7       | 271 ± 45                      |
| 1000                 | 60                | 159 ± 14      | 278 ± 36                      |

Table 2 also shows the ultimate tensile strength (UTS) achieved during tensile testing of the samples. This is seen to increase both with sintering temperature and with sintering pressure. The increase in UTS seen above 900 °C, despite the lower hardness of the AISI420 phase, is due to the increased ductility of these samples, resulting presumably from the improved density and lack of interfacial porosity. The best ductility obtained is in the sample sintered at 1000 °C, where a total elongation of 15% was achieved. Support for this is seen from the SEM micrographs of fracture surfaces after tensile-deformation until failure. In the sample sintered at 850 °C the shape of individual particles can be clearly discerned, resulting from the lack of interfacial bonding between the copper and stainless steel phases in this sample. In the samples sintered at 900 °C (both at 50 MPa and 60 MPa pressure) extensive areas showing a dimpled morphology are seen, although in some places spherical features that can be identified with
the initial powder particles can still be observed in places. In contrast, the fracture surface of the specimen sintered at 1000 °C shows a fully dimpled fracture indicative of an effective consolidation of the material and good bonding at the interphase interfaces.

4. Summary and conclusions
Spark plasma sintering has been used to explore the synthesis of a model dual-phase system using powders of copper and AISI-420 martensitic stainless steel. Densification of the samples, in particular at the interface between the two phases is strongly dependent on temperature, and can also be improved by increased sintering pressure. The best mechanical properties, in terms of ultimate tensile strength and ductility is found in samples sintered at a temperature of 1000 °C, where a density of nearly 99% is achieved. Samples sintered at 900 °C and 60 MPa exhibit a wider difference in hardness between the copper and AISI-420 phases, but although these samples have a density greater than 98%, the tensile fracture surfaces show some evidence of interfacial weakness. Further studies on the post-sintering heat-treatment are underway to allow controlled synthesis of fully dense samples with a range of mechanical strength in each phase.

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