JOURNAL OF MATERIALS SCIENCE 30 (1995) 3883–3887

Preparation of nickel-coated alumina composite powder by an aqueous-phase reduction process

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A nickel coating on the surface of alumina particles, used as a reinforcement in metal–ceramic composite materials, substantially improves their wettability by molten metals. Nickel was deposited on alumina particles by reducing nickel ions from a solution under hydrogen pressure in the presence of ammonia as a complexing agent. It was found that the addition of Fe^{2+} ions and anthraquinone was necessary for forming an adequate nickel coating on the alumina surface. The optimum conditions were the $[NH_3]/[Ni^{2+}]$ ratio of 1.6, 2760 kPa hydrogen pressure, 175 °C, and an anthraquinone addition of $0.04\ g\ l^{-1}$

1. Introduction

Demands for aluminium alloys, which have superb physical and mechanical properties, are ever-increasing in the aeronautical and automotive industries. Composite materials with high strength and hardness, good wear resistance, and light weight can be produced by incorporating ultrafine oxide, carbide or nitride ceramic powder in an aluminium matrix [1,2]. Many production methods have been developed which include the vortex method [3], powder metallurgy [4], pressure forming [5], rheocasting [6], and compocasting [7]. Most of the reinforcements used, such as graphite, alumina, silicon carbide, silica, and zirconia, have limited interfacial reactivity and wettability [8,9]. Thus, it is important to improve the wettability between the different constituent materials when preparing metal–ceramic composite materials.

The methods for improving wettability include:

1. coating the surface of the ceramic particles with a metal [10], and
2. adding a surface-active element to the melt of the base alloy [11].

The purpose of this research was to prepare nickel–alumina composite powder by coating nickel on alumina powder to improve its wettability by liquid metals. The nickel coating was produced by reduction from a solution under hydrogen pressure in an autoclave. The effects of various reaction conditions on the rate of nickel-ion reduction, the morphology of the coated nickel layer, and the content of nickel in the composite powder, were investigated.

2. Experimental procedure

2.1. Materials

The alumina powder used in this work was of 99.9% purity with an average size of 55 ± 15 μm. Other reagents also had 99.9% purity.

2.2. Procedure

The autoclave used in this work was of 2 l volume and 20 cm height and had an impeller of 5 cm diameter. After charging the autoclave with alumina powder and a solution containing $NiSO_4$, $(NH_4)_2SO_4$, NH_4OH, and additives (Fe^{2+} ion and anthraquinone), the mixture was brought to a predetermined temperature and hydrogen pressure. The slurry was sampled at various times and the nickel content in the solution was determined. The amount of nickel coated on the alumina surface was determined by dissolving it in nitric acid. Nickel analysis was done by gravimetry and ICP. The composite powder was also subjected to X-ray analysis and light microscopy.

3. Results and discussion

The major variables in precipitating a metal from solution by hydrogen gas according to the following reaction are temperature and hydrogen pressure

$$M^{2+} + H_2 \rightarrow M + 2H^+ \qquad (1)$$

The equilibrium condition for this reaction is given by

$$K = \frac{a_{H^+}^2}{a_{M^{2+}}\, a_{H_2}} \qquad (2)$$

Thus

$$\log a_{M^{2+}} = -2\,pH - (\log K + \log a_{H_2}) \qquad (3)$$

which indicates that $\log a_{M^{2+}}$ is linearly dependent on solution pH under constant temperature and hydrogen pressure. As can be seen from Equation 1, hydrogen ions must be removed for the precipitation of metal ions to continue. This can be achieved by the addition of sulphate ions which react with hydrogen

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ions to form bisulphate ions

$$H^+ + SO_4^{2-} \rightarrow HSO_4^- \quad (4)$$

Thus, in this work, the above effect was examined using an $(NH_4)_2SO_4$ solution.

Another method for removing the hydrogen ion is to carry out the precipitation in an alkaline region. However, the hydroxide of nickel is formed at a pH above 7, which must be prevented by forming a complex ion of nickel. An optimum amount of the complexing agent should be used to prevent excessive complexation which lowers the free metal ions, thus greatly reducing the metal reduction.

When ammonia is used as the complexing agent, the metal reduction reaction is as follows

$$[M(NH_3)_n]^{2+} = nNH_3 + M^{2+} \quad (5)$$

$$M^{2+} + H_2 = M + 2H^+ \quad (6)$$

$$H^+ + NH_3 = NH_4^+ \quad (7)$$

As the ammonia concentration increases, metal precipitation by reduction increases by the neutralization of the free acid, but the concentration of the reducible M^{2+} ions decreases due to the complexing reaction.

For nickel, the hydrogen reduction reaction, when ammonia is present, is

$$Ni^{2+} + 2NH_3 + H_2 = Ni + 2NH_4^+ \quad (8)$$

It was reported that the optimum $[NH_3]/[Ni^{2+}]$ ratio was 2 [12]. According to the results of this work, as shown in Fig. 1, the optimum ratio was 1.6, which was used in all the subsequent experiments.

Fig. 2 shows the amount of nickel coated on the alumina surface as a function of the stirring speed under the conditions of 46 g Ni^{2+}/l, 2410 kPa H_2 pressure, 2 g Fe^{2+}/l, 0.12 g anthraquinone/l, 175 °C temperature, and 60 min reaction time. It can be seen that the reaction rate increases with stirring speed up to 600 r.p.m. but does not increase any further beyond it.

The effect of hydrogen pressure under otherwise the same conditions as above on the amount of nickel coated on the alumina and that of the nickel remaining

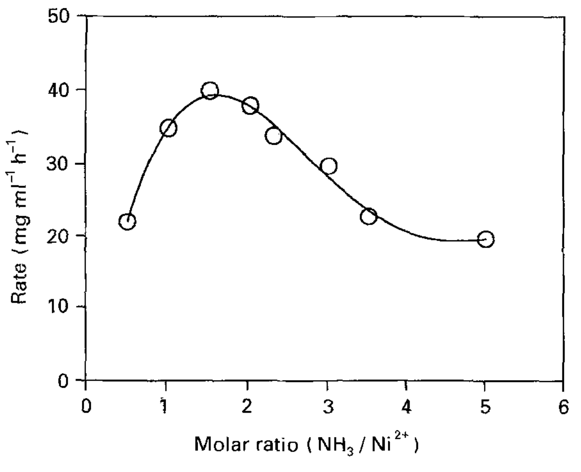

Figure 1 Effect of $[NH_3]/[Ni^{2+}]$ molar ratio on the rate of nickel precipitation.

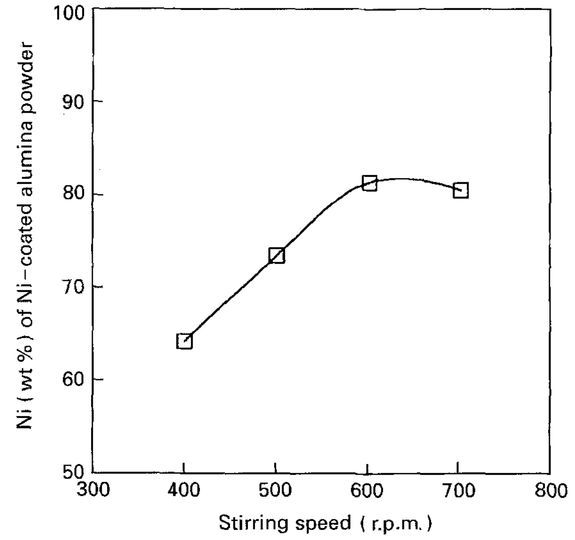

Figure 2 Effect of stirring speed on nickel deposition on alumina particles (46 g l^{-1} Ni^{2+}; p_{H_2}, 2410 kPa; 2 g l^{-1} Fe^{2+}; 0.12 g l^{-1} anthraquinone; T, 175 °C; time, 60 min).

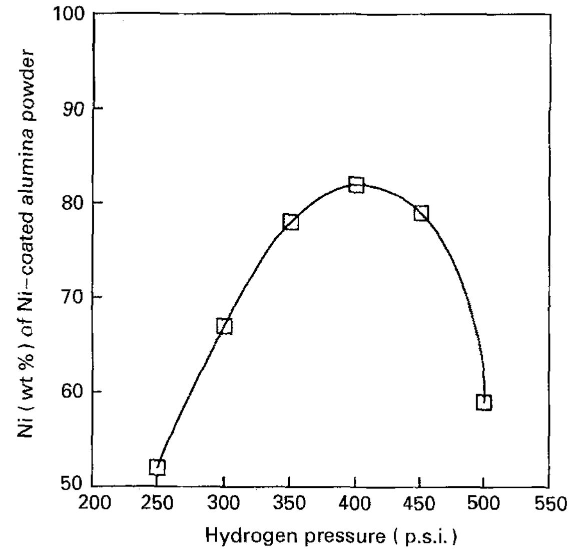

Figure 3 Effect of hydrogen pressure on nickel deposition on alumina particles (1 kPa = 0.145 p.s.i.; 600 r.p.m.; other conditions the same as for Fig. 2).

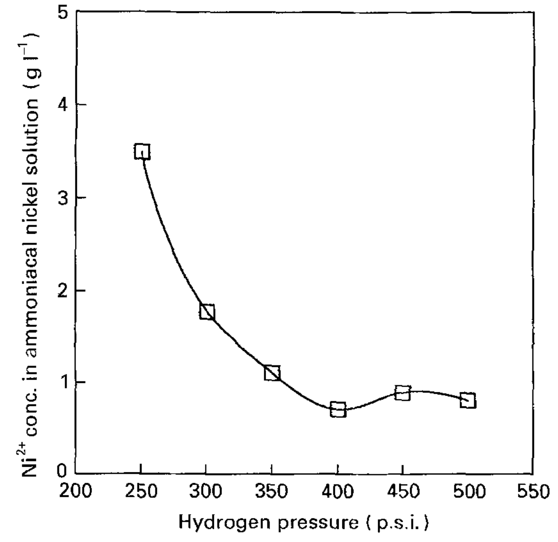

Figure 4 Effect of hydrogen pressure on nickel reduction from the solution (1 kPa = 0.145 p.s.i.; other conditions the same as for Fig. 3).

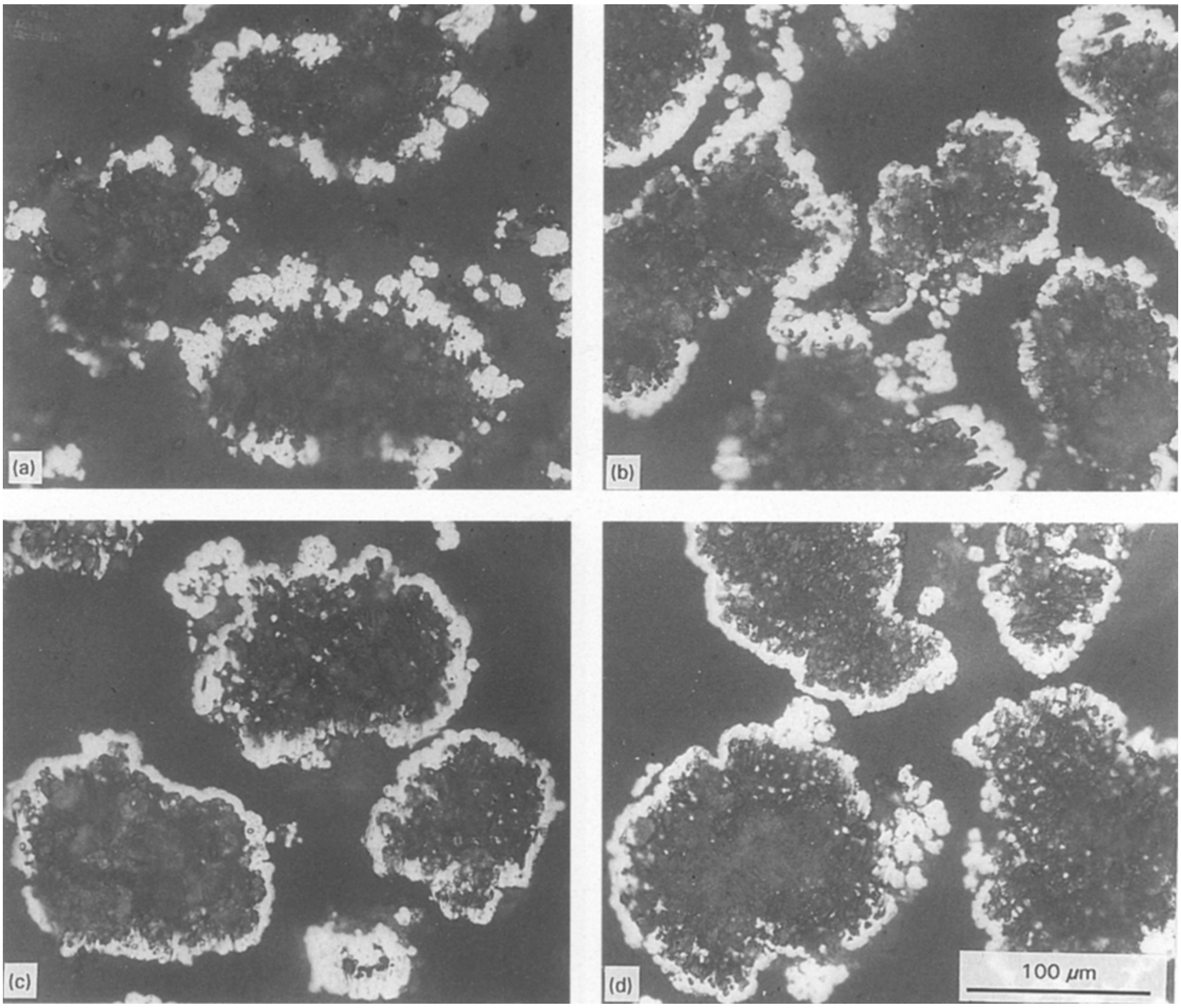

Figure 5 Cross-section of nickel-coated alumina particles produced under various hydrogen pressures: (a) 1720 kPa, (b) 2070 kPa, (c) 2410 kPa, (d) 2760 kPa.

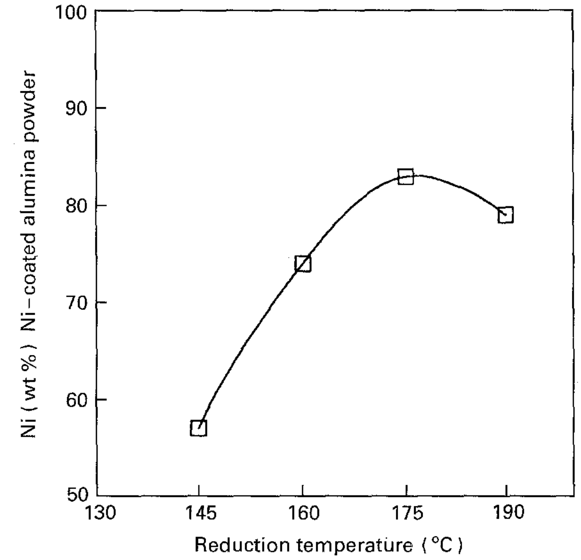

Figure 6 Effect of temperature on nickel deposition on alumina particles (600 r.p.m.; other conditions the same as for Fig. 2).

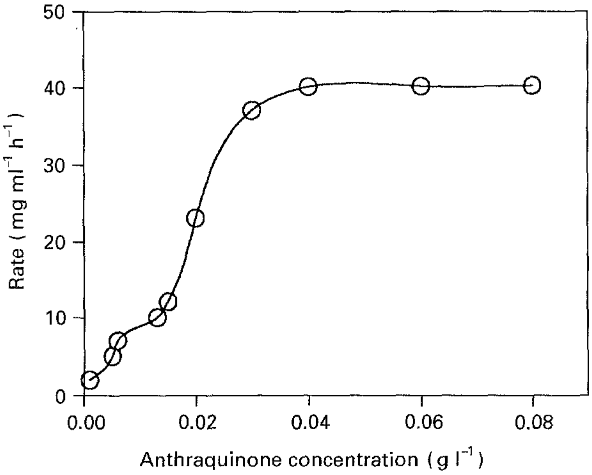

Figure 7 Effect of anthraquinone addition on nickel precipitation rate (600 r.p.m.; other conditions the same as for Fig. 2).

in the solution is shown in Figs 3 and 4. The coated amount increases with hydrogen pressure up to 2760 kPa and then decreases. As can be seen in Equation 1, the rate of nickel precipitation should increase with hydrogen pressure, but hydrogen-ion concentration also increases with hydrogen pressure, which favours the reverse reaction. The electron microscope pictures of the cross-section of the nickel-coated alumina powder prepared under various hydrogen pressures are shown in Fig. 5.

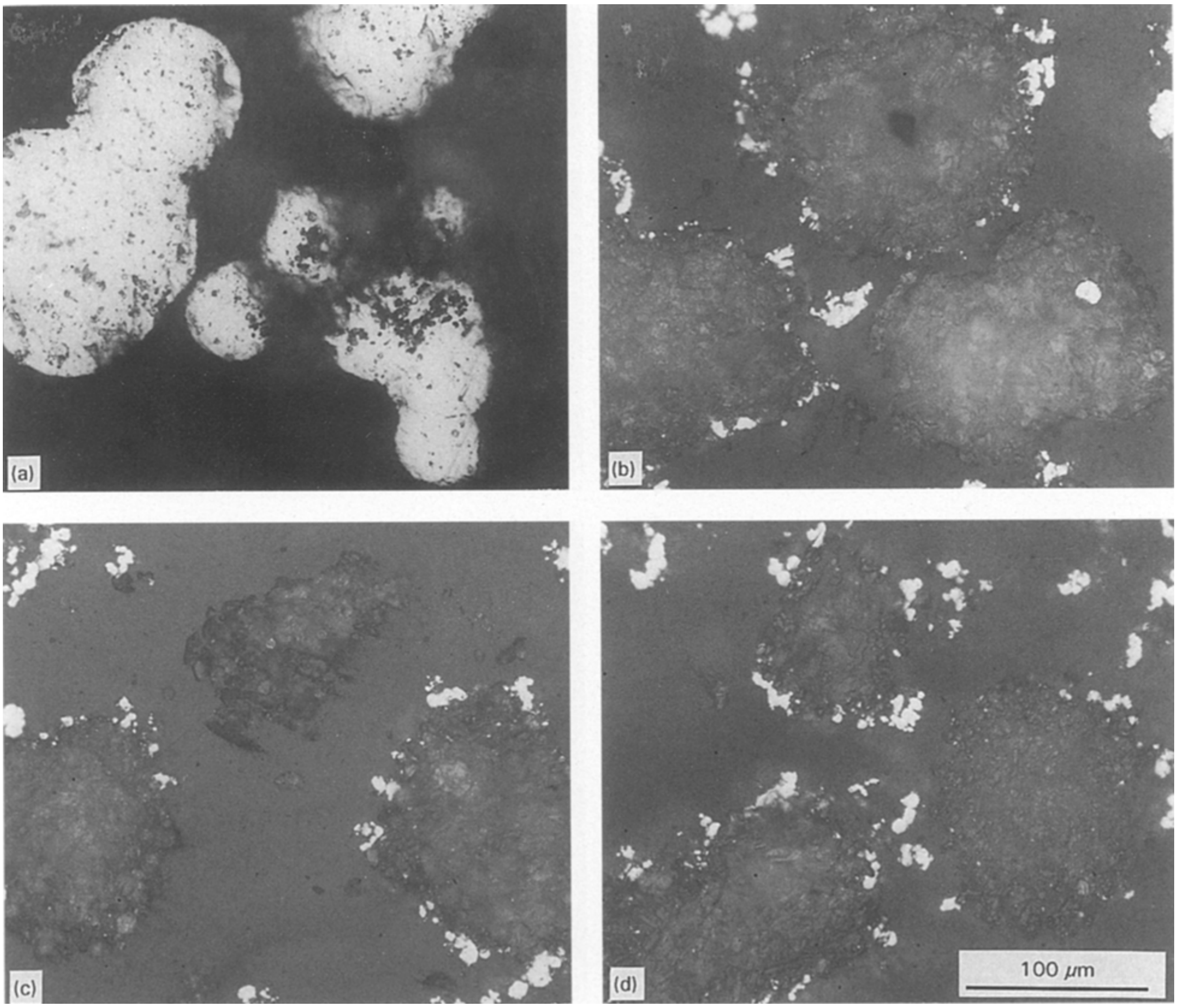

Figure 8 Cross-section of nickel-coated alumina particles produced at different concentrations of Fe^{2+}: (a) 0 g, (b) 0.5 g, (c) 1.0 g, (d) 1.5 g.

The effect of temperature is shown in Fig. 6, indicating that the rate of coating is highest at 175 °C. The X-ray analysis revealed only alumina and nickel, without any iron.

The rate of nickel precipitation increased with the amount of surfactant anthraquinone up to $0.04\ g\,l^{-1}$ and then levelled off, as can be seen in Fig. 7.

The Fe^{2+} ion was added to the solution to prevent the agglomeration of nickel and to obtain uniform coating. As illustrated in Fig. 8, nickel did not deposit on the alumina surface when no Fe^{2+} ions were present, and the deposited amount of nickel increased with Fe^{2+} concentration.

4. Conclusions

From the results of this experimental investigation on the preparation of nickel-coated alumina composite powder by hydrogen reduction in an autoclave, the following conclusions are drawn.

1. The rate of nickel precipitation increases with $[NH_3]/[Ni^{2+}]$ molar ratio up to 1.6, then decreases with a further increase in this ratio.

2. The addition of the surfactant anthraquinone increases the nickel precipitation rate up to 0.04 g anthraquinone/l but has no further effect above this concentration.

3. The addition of Fe^{2+} ion is necessary to obtain a uniform coating of nickel on alumina.

4. The rate of nickel precipitation is affected by stirring rate, hydrogen pressure, and temperature. The optimum values of these variables found in this work were 600 r.p.m., 400 p.s.i. (1 kPa = 0.145 p.s.i.), and 175 °C, respectively.

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Received 28 April
and accepted 16 November 1994