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Production of $LaPO_4$ coatings using a novel ultrasonically-assisted plating technique

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ABSTRACT

Dense $LaPO_4$ and $LaPO_4$–Al_2O_3 coating films were produced on the surface of stainless steel substrates at room temperature and ambient pressure with the aid of a novel ultrasonic-based mechanical coating method, which we call UMCA. The main emphasis was on examining the conditions necessary for the successful coating operation and characterizing the as-deposited coatings for the thickness, uniformity and surface morphology. The experimental results suggested that hardness and thermal conductivity of balls and substrate are key parameters influencing the coating efficiency. The coated samples showed an improved hot corrosion resistant in Na_2SO_4–NaCl molten salts at 950 °C.

1. Introduction

Recently, there has been increasing interest in developing the coating of metal components with ceramic materials. Although considerable progress has been made in this area, there still remain many practical limitations in combination of materials for the substrates and coatings. A typical example of the material combinations that presents a considerable challenge for coating technology is the case when an easily oxidized or/and easily melted metals needs to be coated with a refractory and/or chemically inert ceramics. In many cases, such coating processes can be realized only inside vacuum or high-temperature chambers that impose limitations on the area of surface to be coated. This makes the coating process expensive and reduces its efficiency. Furthermore, such a process encounters the problem that oxidation of the metal substrate surfaces cannot be avoided even at ultra low pressures.

In an attempt to overcome the above problems, the authors have proposed a novel method for treatment of metallic surfaces, which include coating and/or armouring of the surfaces with powdered materials. Because the method is based on mechanical activation through high-energy ball impacts and uses the energy of ultrasonic vibrations to accelerate the balls, we termed it as ultrasonic mechanical coating and armouring (UMCA). In the previous paper [1], we introduced the concept of the UMCA method and reported some results of the method examination by using Al and Ti powder particles as a substrate and coating material, respectively. The UMCA method has a number of attractive features and competitive advantages that makes it a process worthy of further investigation. First, it occurs at ambient pressures and temperatures by using relatively cheap ultrasonic equipment. Second, the process and related equipment are conceptually designed in such a way that there is no limitation regarding the area of the surfaces that can be coated. Third, extremely high frequencies of balls-to-surface collisions (about one million per second and square meter) and high energy of the balls provide a means of peeling the oxide film from the substrate during the treatment and of hammering the particles into the substrate surface layers for a relatively short time. Both these phenomena can provide a better contact between particles and matrix.

In the present study, we made an attempt to apply the UMCA method in order to produce coatings of monazite-type lanthanum phosphate, $LaPO_4$. In recent years, the lanthanum phosphate has attracted the interest of both research and industry, mainly due to its unique high-temperature stability and chemical inertness. Potential applications of the lanthanum phosphate include, but not limited to, such an area as thermal barrier coatings for materials used in extreme environments [2]. However, the above-mentioned thermal and chemical stability of lanthanum phosphate makes it difficult or even

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impossible to produce the lanthanum phosphate coatings by conventional techniques. The goal of the present experiments was to deposit the $LaPO_4$ coatings on stainless steel samples. Such coatings are of considerable industrial interest for application to components working under very high temperatures and chemically aggressive environment, for example in the waste treatment industry. Our main emphasis was on examining the conditions necessary for the successful coating operation and characterizing the obtained coatings for the thickness, uniformity and surface morphology. The characterization was carried out using scanning electron microscopy (SEM), electron probe microanalysis (EPMA) and laser microscopy. Besides, in order to estimate the coating performance characteristics, the coated samples were subjected by hot corrosion tests in a Na_2SO_4–NaCl melt at 950 °C.

2. Experimental

The experiments were carried out in a laboratory unit described in details in our previous work [1]. Fig. 1 schematically presents one of the main parts of the unit, the bowl-shaped resonance chamber. A certain number of stainless steel (hereinafter STS) or ZrO_2 balls (diam. 3 mm) were put into the chamber fixed beneath the surface to be coated. The chamber was attached to an ultrasonic transducer powered with a high-frequency generator at a resonant frequency in the range of 21–22 kHz at an electrical power of about 1 kW. The ball number was varied in the range of 100 –200.

In the above-cited earlier study [1], we investigated the version of the UMCA method in which Ti powder particles for coating were put into the resonant chamber together with STS balls. Oscillations of the chamber initiated an acoustic gas streaming that forced the particles to fly inside the chamber. This resulted in adhering a part of the particles on the sample surface (Al plate), and hammering the particles into the surface layers of the substrate to produce the Ti coating.

Attempts to apply the same technique to the coating of $LaPO_4$ were unsuccessful because of a very low degree of coverage. The reason, presumably is that the $LaPO_4$ particles were much smaller than the Ti particles. As a result, they were carried away from the substrate by the acoustic streaming. This prevented the particles from adhering to the substrate. Therefore, in the present work, we examined another modification of the UMCA method which was that the samples were first precoated with a $LaPO_4$ particle-containing ethanol suspension

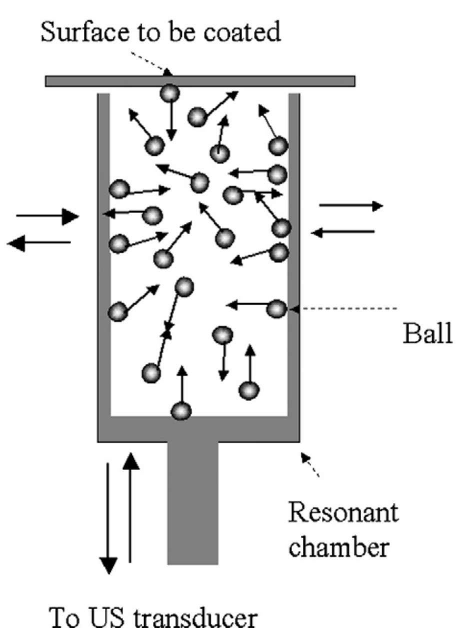

Fig. 1. A representation of the process concept.

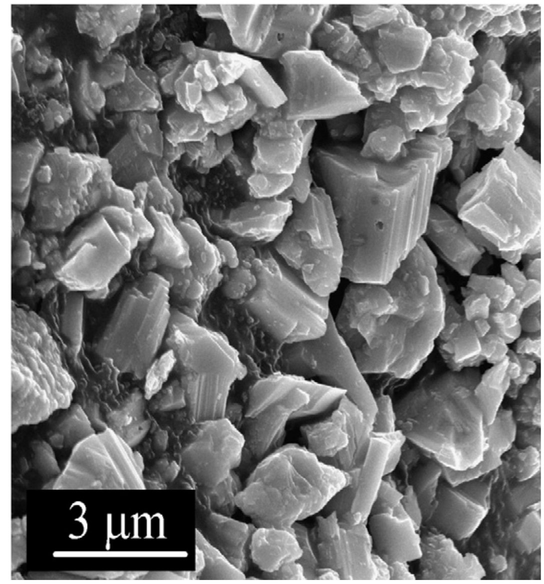

Fig. 2. A SEM view of $LaPO_4$ particles after calcination.

and then the precoated surface was treated under Ar atmosphere by balls in the resonant chamber as shown in Fig. 1.

The precoating procedure was performed in the following way. The surface of as-obtained substrates, stainless steel (SUS304) square plates of size 50×50×2 mm, was rinsed with ethanol and dried. Lanthanum phosphate powder in amount of about 1 g was added to ethanol and stirred into suspension. The volume ratio of ethanol to powder was about 3:1. A few drops of the suspension were placed on the surface and then the substrate was inclined at small angles to the horizontal for letting the drops spread out slowly to cover the entire substrate After evaporating ethanol, the substrate was fixed at the top of resonance chamber for coating treatment. The treatment time was 3, 5 or 10 min per one coating operation. The number of coating operations, N_c was ranged from 1 to 3. In the case $N_c>1$, the precoating of the sample surface with the particle-containing ethanol suspension was done after completing each coating operation.

All the coatings were produced at room temperatures. To prepare the $LaPO_4$ powder, rhabdophane-type $LaPO_4{\cdot}0.5H_2O$ was calcined at 1273 K for 6 h [3,4]. After that, the particle size distribution was investigated by SEM. Fig. 2 shows a typical SEM view of the $LaPO_4$ particles after calcining. Two types of particles can be seen in the picture. The first one is agglomerates of smaller particles with size down to 0.5 µm. The second one is non-agglomerated single particles with the size up to 3 µm. In order to examine the possibility for producing composite coatings, in some experiments, the lanthanum phosphate particles were premixed with Al_2O_3 powder at a weight ratio of 1:1, and then an ethanol suspension of the mixture was spread over the sample surface to be treated as explained above. After the treatment, all samples were rinsed with ethanol and cleaned ultrasonically in a water–ethanol mixture bath.

Besides, some samples were precoated with the $LaPO_4$ containing ethanol suspension after preliminary treatment of the as-obtained substrate surface with balls. The purpose of these experiments was to elucidate whether there is an effect of preliminary mechanical activation of the substrate surface on the coating efficiency.

3. Results and discussion

3.1. Degree of coverage and coating uniformity

Among the important characteristics of a coating are the degrees of coverage, α and the coating uniformity. The latter is defined as the