

A DRY ULTRASONIC-BASED METHOD FOR MECHANICAL COATING

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Abstract. This work presents a new ultrasonic-based dry mechanochemical method for armouring or coating metallic surfaces with other metal or ceramic materials. Two modifications of the method have been examined. In the first one, hard balls and metal or ceramic powder are put into a bowl-shaped resonant chamber which is fixed beneath the surface to be coated. In the second one, only balls are put into the chamber while the surface is previously precoated with a suspension of a liquid and powder. The chamber is set into high frequency vibration by an ultrasonic transducer attached to the chamber bottom. This initiates a chaotic motion and collision of the balls and powder particles, and hammering them into the metallic surface. Experimental examination was performed using substrates of Al and stainless steel, powders of Ti, SiC, and Al₂O₃ and a number of liquids for precoating. The results revealed that the method allows the production of various coatings and armoured layers on the metallic surfaces at room temperatures regardless of difference in the properties of materials used.

1. INTRODUCTION

Despite significant advances in the coating technology, there remain many practical limitations in combination of substrate and coating materials to be used. Most of these limitations are caused by the fact that many advanced materials, which could provide new or significantly improved coating performance, differ widely in their properties. For example, it is very difficult to apply a coating of an easily oxidized or/and easily melted metal on the surface of a more refractory metal or ceramic material.

Recently, there have been attempts to produce coatings using the principle of mechanical alloying process. In the process, samples were put into ball mill jars of vibrating type together with the balls and coating powder, and then treated for a certain time. It is found that dense thick coatings with good adhesion can be produced by mechanical alloying of

Ti samples with Al powder with consequent annealing for intermetallic phase formation [1,2]. However, one of the big disadvantages is that size of sample to be coated is limited by dimensions of ball mill jars.

In this work, we introduce a new dry method designed for coating and armouring of metal surfaces with powdered metallic, ceramic or composite materials. Inasmuch as the method uses ultrasonic energy, and is based on the principles of mechanochemical activation and mechanical alloying, we called it UMCA (Ultrasonic Mechanochemical Coating and Armouring). The proposed method has no limitation on the area of surfaces that can be processed that makes it usable for many practical applications. Other attractive features of the method are as follows. 1) The coating or armouring process can be performed under room temperature and atmospheric pressure. 2) The method can

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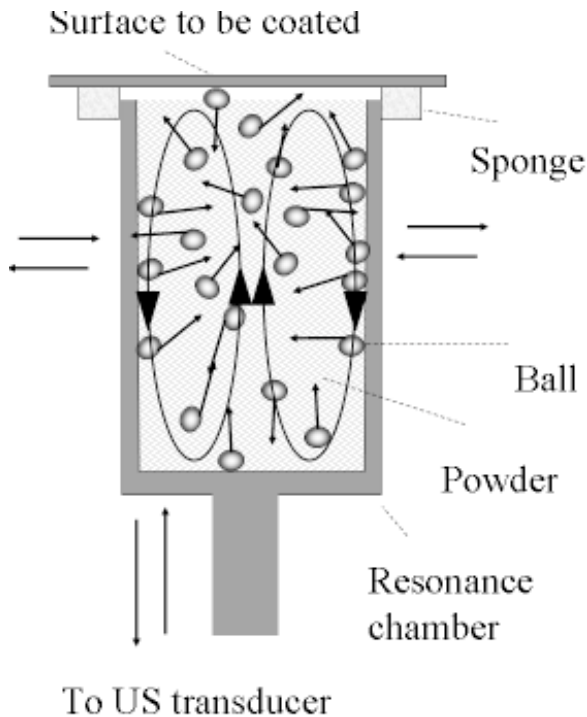


Fig. 1. A representation of the process concept.

provide a rapid cold-hardening and nanostructuring of powder particles before coating, as may be necessary. 3) The method can be applied itself or can be combined with other techniques of surface treatment.

The following sections introduce the method concept and present some experimental results of the method examination.

2. THE METHOD CONCEPT AND ITS EXPERIMENTAL REALIZATION

The proposed method can be performed in one or few stages depending on the goals and material system used. Fig. 1 explains the process concept. Balls made of a hard material (stainless steel, ZrO_2 or other) are put into a bowl-shaped resonant chamber which is fixed beneath the surface intended to be coated or armoured with powdered materials. In the first modification of the process, the powdered material is also put into the chamber. In the second one, the powder layer is deposited on the surface before fixing it above the resonant chamber. The chamber is designed in such a way that all its parts are vibrated at a fixed resonance frequency which is set by an ultrasonic transducer

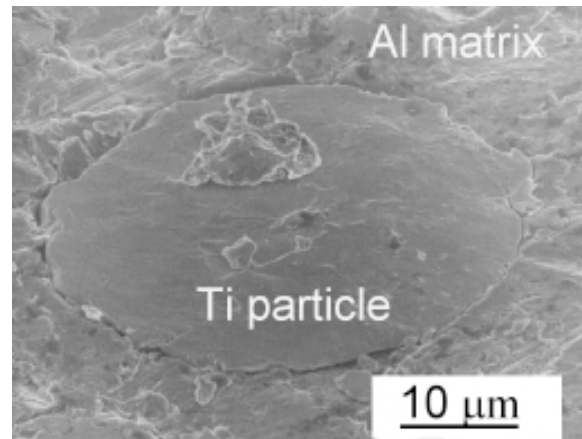


Fig. 2. A SEM view of a Ti particles hammered into Al matrix.

attached to the chamber bottom. The paired arrows shown outside of the chamber in Fig. 1 indicate its vibration in vertical and radial directions. The chamber vibration results in a vigorous acceleration and chaotic motion of the balls inside the resonance chamber that leads to their collisions with each other and with the upper surface to be treated.

Besides, the chamber vibrations cause small and large-scale acoustic streaming inside the chamber. The large-scale streaming entrains the particles into an eddy motion inside the chambers that causes transferring a part of the particles to the sample surface and adhering on it. When a ball collides with such an adhered particle, this causes hammering the particle into the sample surface layer. Two closed loops in Fig. 1 show one of the possible pattern of the large-scale acoustic streaming while the arrows indicate the stream directions.

In general, the coating method and related equipments are conceived such that either the resonant chamber can move over a surface or, the surface can shift relative to the chamber since it may be necessary to treat a wider surface area. However, at the present stage of our study, we investigate a simplified version of the method in which both the chamber and surface are fixed relative to each other.

The method was examined with three substrate/powder material systems: Al/Ti, Al/SiC, stainless steel/ Al_2O_3 (substrate/powder), although most of the results presented are related to the first sys-

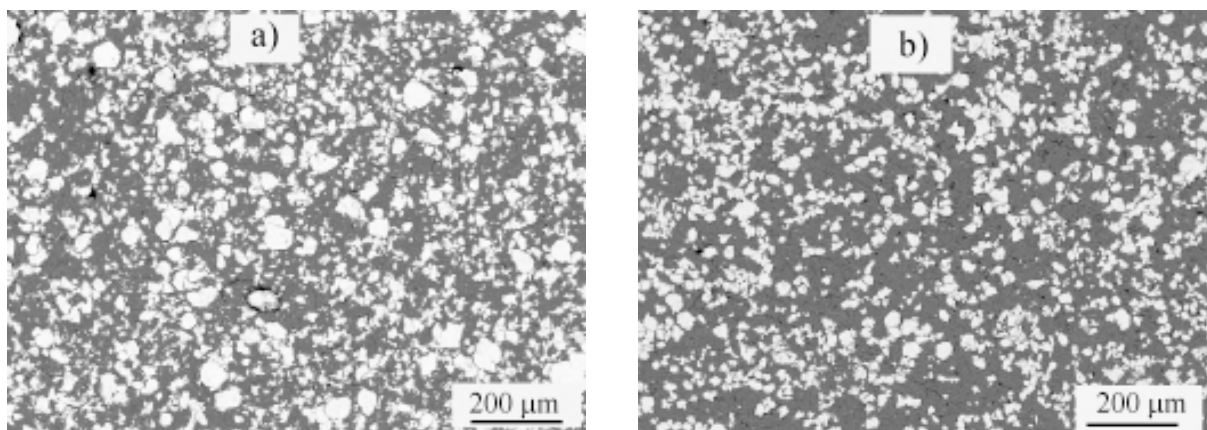


Fig. 3. Back scattered electron images of Al surface (black) coated with Ti particles (white). Plate surface conditions: a) dry, b) previously wetted with 25% glycerol aqueous solution.

tem. The experiments were performed by using a laboratory-scale equipment. A sample plate of 50x50 mm in size and 2~3 mm in thickness was fixed at the top of the resonance chamber with inner diameter 30 and height of 60 mm. The chamber was set into vibration at a frequency of 21~22 kHz and amplitude of 10~12 μm by using a magnetostrictive transducer powered with a 1 kW ultrasonic generator. The effect of ball number on the ball-to-plate collision efficiency was investigated preliminarily. Thereafter, their number was kept constant at 92 with diameter of 3 mm. The maximum size of powder particles was 45 (Ti), 3 (Al_2O_3) and 0.27 (SiC) μm , respectively, although majority of the Al_2O_3 and SiC particles existed as agglomerates.

Three variants of the process were examined. They differed in the conditions of preparation of the sample plate surface: 1 – as-obtained, 2 – preliminarily wetted with a liquid, 3 – precoated with a mixture of powder and liquid. In the first and second cases, the powder charge of 0.5 g was put into the chamber together with balls. In the third case, only balls were put into the chamber. The liquids used were ethanol, aqueous solutions of glycerol and acetone solutions of stearic acid. The main differences between them, essential for the present experiments, are the evaporation rate and viscosity. Easily evaporated ethanol is less viscous. The glycerol aqueous solutions, on the contrary, are hardly evaporated and very viscous. As for the acetone solutions of stearic acid, despite the fact

that the acetone evaporates rapidly, the stearic acid remains on the surface as a thin grease layer. All experiments were performed inside a sealed box under an Ar gas atmosphere. The processing time was ranged from 10 to 60 min. After processing, the surface of the samples was cleaned by wiping and rinsing with alcohol to remove the reminder of particles slightly adhered to the surface, and then analyzed by SEM, EPMA and SIMS (secondary ion mass spectrometry).

3. RESULTS AND DISCUSSION

Collision of a ball with particles adhered on the plate surface results in hammering the particles into the plate surface layer. The experimental data suggested that the hammering effect is more pronounced when hardness of powder particles is higher than that of sample plate, as for example in the Al/Ti and Al/SiC systems. Fig. 2 shows typical SEM views of Ti particles hammered into Al matrix. As can be seen, the upper surface of the particles became flat although the original particles had convex polygonal shape. Further observations of cross-sections of the sample plates after ultrasonic treatment revealed that Ti particles are flattened out and their bottom parts are completely incorporated into the Al matrix.

When Ti powder was put into the resonance chamber, about 57% of the dry Al surface was coated with the Ti particles after 10 min of the surface treatment. This result was obtained by taking

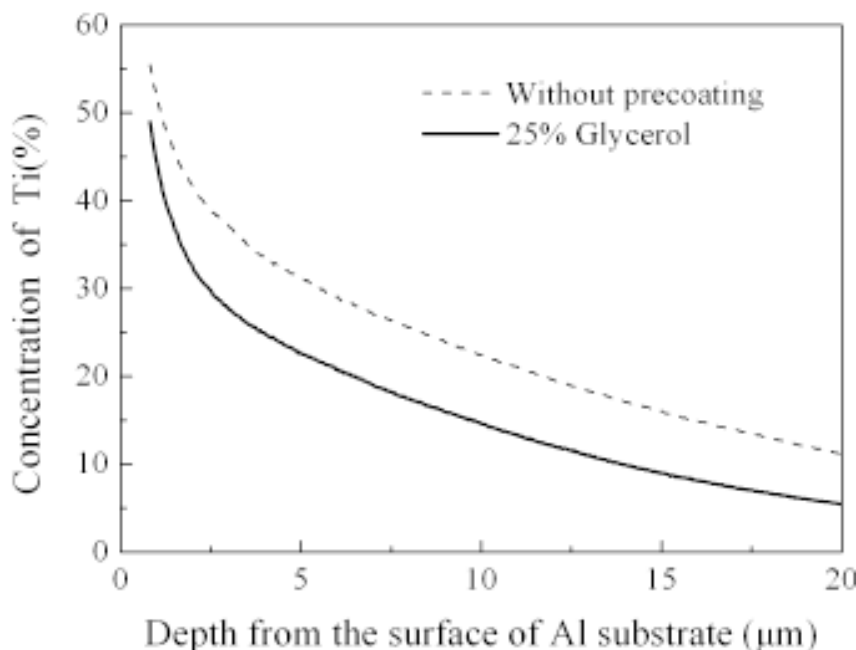


Fig. 4. Variation of Ti concentration with distance from sample surface.

a back scattered electron image of a part of the surface shown in Fig. 3a and by the further processing of the image by Scion Image software. It is interesting that after the surface treatment a high proportion of particles after the treatment had significantly smaller size as compared to original one. This finding appears to suggest that the particles are subjected to grinding during the impact treatment with balls although the grinding mechanism is not clear yet. Such surfaces, which are only partly covered with particles embedded in the surface layer, can be termed armoured surfaces. The similar samples were obtained for Al/SiC and SUS304/Al₂O₃ material systems, although the proportion of area coated with particles, referred as to the degree of coverage, for the SUS304 plate was slightly less than that for Al plate. It is expected that a combination of the particle embedding and cold-hardening of the surfaces should impart higher strength properties and wear resistance to the surfaces. Characterization of armoured surfaces produced by the UMCA method will be the subject of our further studies.

It is important to note here that the degree of coverage was almost unchanged with the further increase in the impact treatment time up to as long as 60 min. In an attempt to clear up the reason, we

conducted a set of experiments to measure the frequency of ball collisions per unit area, f_b and portion of the surface area struck by the balls during ball-to-plate collisions per unit time, S_b . The measurements were based upon findings that the balls leave marks on the sample surface in the form of small cavities after collision. In the present experiments with the ball number of 92, f_b and S_b were estimated to be $1.06 \cdot 10^6 \text{ s}^{-1} \times \text{m}^{-2}$ and $5.62 \cdot 10^{-2} \text{ s}^{-1}$, respectively. The reciprocal of S_b gives the minimum time needed for surface treatment that imply the time during which every part of surface has been struck with a ball at least once. Under the given experimental conditions, the time is about 18 seconds. Consequently, during the 10-min treatment, the balls strike the same place about 33 times that would be quite enough to produce a thick coating layer on the surface provided that a sufficient amount of particles have been adhered to the surface. Thus, one of the possible ways to increase the degree of coverage is to improve the capturing efficiency of particles by the surface.

Assuming that particles can bounce off the dry surface on collision, a series of experiments was carried out using previously wetted surfaces. None of the liquids tested, namely ethanol, aqueous solutions of glycerol and acetone solutions of stearic

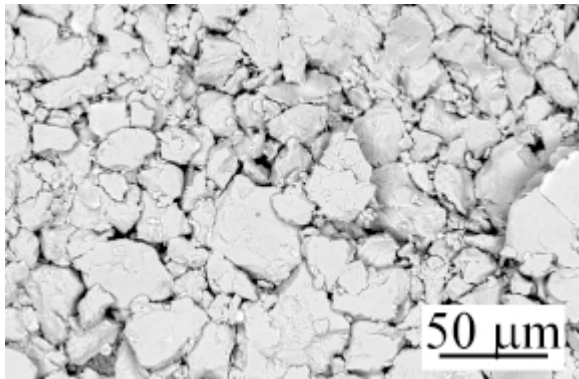


Fig. 5. A SEM image of Al surface fully coated with Ti particles.

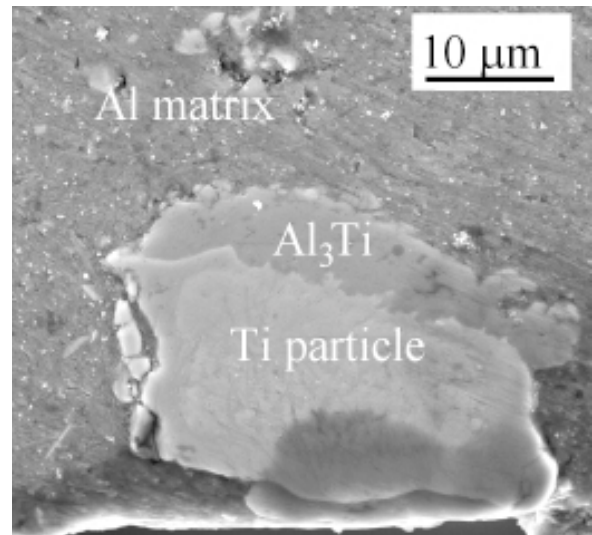


Fig. 6. A cross-sectional view of a Ti particles after annealing.

acid gave better results. Furthermore, sometimes the degree of coverage was even less than that for the dry plate, as presented in Fig. 3b for the case of the surface previously wetted with 25% glycerol aqueous solution.

The thickness of armoured layers was measured with SIMS. The measurement results are shown in Fig. 4 as depth profiles of Ti concentration for two sample plates: 1 – dry, 2 – wetted with 25% glycerol aqueous solution. It is clear that in the first case the layer was deeper. For example, at the depth of 20 μm , the Ti concentration was about 10% for the dry plate against 5% for the previously wetted. Most likely the layer of high-viscosity glycerol on the plate surface dissipates the energy of balls that results in a significant weakening of the hammering effect.

In the next set of experiments, the surface was previously coated with a suspension of Ti particles in the above mentioned liquids, and then the precoated surface was treated ultrasonically with balls. Here, the results were very different depending on what liquid was used in suspension. In the cases of glycerol or stearic acid solutions, the degree of coverage remained unchanged or decreased as compared with the dry surface under the same experimental conditions. However, when ethanol was used in suspension, the degree of coverage was significantly improved. The surface of Al plate was fully covered with Ti particles after the 10-min treatment. The best results were obtained when the ball treatment process was carried out in 2 or 3 stages, each of which was followed by precoating procedure. A SEM view of such a fully coated surface is presented in Fig. 5. The

coating was performed in 3 stages. The coating thickness was estimated to be within 30 μm . Taking into account the particle flattening, such a coating should consist of 2–3 layers of the Ti particles. Thus, short time of evaporation of liquid from the precoated suspension appears to be an important feature of the coating process. Although requiring further examination, the time of evaporation should be shorter than the treatment time so that to ensure a good contact between the plate and particles after evaporating.

Finally, one more important feature of the UMCA method should be mentioned here. Because the interaction time between balls and particles at the sample surface is very short, their deformation or/and embedment into the surface layer occur with very high speeds. This can cause cracking and peeling of thin oxide films, inevitably presented on the surfaces of such easily oxidized metals as Al and Ti, and provide, thus, a better contact between particles and matrix that is of basic importance for the mechanical alloying and coating technology, especially from the standpoint of diffusion bonding or further heat treatment. However, we have not yet direct evidence of the cracking and peeling phenomena. Additional experiments revealed the formation of a Al_3Ti layer on the Ti-coated Al plate after annealing at 640 $^\circ\text{C}$ during 120 min under Ar atmosphere. Fig. 6 is a cross-sectional view of a Ti particle embedded into the Al matrix. The picture

was obtained after the sample annealing. EMPA analysis of the sample showed the presence of a Al_3Ti layer at the particle surface. The layer thickness was estimated to be 6–7 μm .

4. CONCLUSIONS

The present study introduces a new ultrasonic-based dry method for armouring or coating metallic surfaces with other metal or ceramic materials, and presents the experimental results on the method examination. The proposed method offers several attractive features that may make the method very useful for many practical applications. One of them is the possibility to cover a wide surface area under room temperature conditions by using a broader combination of coating materials and substrates. Particularly, the examination results

showed that Al surface can be coated with Ti or SiC particles for a relatively short time. It is the authors belief that the method can be effectively used in itself, for example for producing composite metal-ceramics coatings, and in combination with other techniques of surface processing. Besides, the proposed method seems to be attractive for obtaining multi-layer coatings comprising different metal and ceramic particles.

REFERENCES

- [1] S. Romankov, W. Sha, S.D. Kaloshkin and K. Kaevitser // *Surface and Coatings Technology* **201** (2006) 3235.
- [2] S.E. Romankov, S.D. Kaloshkin and L.Yu. Pustov // *Fizika Metallov I Metallovedenie* **101** (2006) 1, In Russian.