

Studies of Particle Deformation and Microstructure Evolution Using High Strain Rate Particle Compression Test

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The deformation behavior of particles plays a significant role in achieving adhesion during cold spray. The deformation behavior of the particles is associated with the fracture of the oxide layer and recrystallization, which are the key elements of the quality of cold spray. Studies of particle compression have been made to understand the deformation behavior of a particle. However, the deformation behavior of particle under controlled load and precise and high strain rate is yet to be studied. Here, we show the oxide layer fracture pattern and recrystallization regime under controlled load with a precise and high strain rate. We found that the cracks in the oxide layer initially appeared on the equator of the particle and propagated towards the edge of the top surface. Meanwhile, on the top surface, the circumferential crack was developed. On the other hand, the nanoindentation result showed that the compressed particle under a high strain rate has an unusual load-displacement behavior. Our results demonstrate that the oxide layer fracture behavior corresponds to the adhesion mechanism suggested by previous studies. Our study also revealed that recrystallization takes place within the particle under a high strain rate. We anticipate this finding to give a general insight into the deformation behavior of particles during cold spray. For instance, since the recrystallization behavior at a given strain rate can be predicted through this study, the resultant grain size and shape, which is associated with mechanical properties, can also be predicted. Furthermore, the amount of strain and strain rate to form optimal adhesion can be evaluated.

1 Introduction

Cold spray technique allows coating and thick deposition in a solid state by spraying solid particles on a substrate at a high velocity [1-5]. This distinctive aspect of cold spray offers a multitude of advantages. As the adhesion is achieved in a solid state, the cold spray technique is inexpensive and makes it easier to make a thicker coating than other thermal spray techniques. Due to these advantages, cold spray is used throughout the engineering industry. For instance, cold spray can be used on machines' mechanical parts for repair and as additive manufacturing to create mechanical parts [2-5]. Thus, it is important to understand the coating mechanism of cold spray to improve its quality.

The adhesion mechanism of metallic cold spray is associated with the deformation behavior of the particle [6-10]. Hassani et al. used the LIPIT (Laser-Induced Particle Impact Test) to directly observe the impact-induced bonding behavior of single particles [11-12]. In the cold spray process, particles are largely deformed in a very short period. Ultrafast large plastic deformation of particles causes unique internal microstructure evolution such as dynamic recrystallization [13]. This microstructure formation is important for understanding the quality of cold-sprayed coatings and laminates. To this end, it is necessary to understand the relationship between deformation and microstructure formation in a high-rate strain environment. Still, it is difficult to precisely measure strain during deformation using LIPIT.

Nanoindentation is an indentation technique that applies micro-loading on a specimen and measures the indentation depth to evaluate mechanical properties. It is suitable for micro to nano scale evaluation. In conventional nanoindentation, a cone-shaped indenter, such as a Berkovich indenter, is pressed into the substrate. A particle compression test can be performed by changing the indenter to a flat indenter [14-16]. Furthermore, the recent development enabled indentation with high strain rate which prove useful in analyzing deformation behavior under high strain rate. When impact nanoindentation is employed for particle compression test, it is possible to determine the load applied to the particles and the deformation, and by observing the particles after the test, it is possible to determine the deformation behavior and the process of microstructure formation.

Our study focuses on the microstructure evolution of particles after a high-speed compression test. For this reason, monocrystal particles are used so that the change in crystal structure appears clearly. Moreover, a thick oxide layer was coated on the particle as in previous studies [16].

In this study, we have employed the nanoindentation technique to carry out a high-speed particle compression test, which will improve the understanding of the deformation regime during the compression test. In addition, scanning electron microscopy (SEM) observation was also made to analyze crack behavior around the particle.

2 Experimental procedures

2.1 Material

The gas-atomized aluminum powder THP-A20S manufactured by Toyo Aluminium K.K. Hino Works was used. The particles were polycrystalline particles produced by gas atomization. The particles annealed in a vacuum furnace at 200 °C for 3 hours. Then, the powder was coated with a zinc oxide layer by barrel sputtering, which allows for uniform coating. The target thickness of the zinc oxide layer is about 30 nm. In this study, both heat-treated aluminum particle with and without zinc oxide coating were subjected to following experiments and observation. For the impact nanoindentation test, described below, particles in diameter of $10 \mu\text{m} \pm 2\mu\text{m}$ were selected for the test.

2.2 Impact compression test by nanoindentation technique

For the particle compression test, a nano-indenter NanoFlip was inserted into SEM Hitachi S-3400N with aluminum particles on platinum coated sapphire glass substrates to perform in-SEM nanoindentation. An illustration of impact nanoindentation test is shown in **Fig. 1**. As shown in the diagram, a flat diamond indenter compresses Al particle. The indenter was withdrawn away from the particles prior to compression and compressed particles with nominal strain rate ($\dot{\epsilon}_{nom}$) of approximately 10^3 s^{-1} for impact tests. The nominal strain (ϵ_{nom}) is defined in Equation 1, and the calculation of the nominal strain rate is given in Equation 2 below,

$$\epsilon_{nom} = -\ln \left(1 - \frac{\delta}{d_0} \right) \quad (\text{Eq. 1})$$

$$\dot{\epsilon}_{nom} = \frac{\epsilon_{nom}}{t} \quad (\text{Eq. 2})$$

where, d_0 is the initial diameter, δ is the displacement, and t is the time. A built-in software of the NanoFlip was used to obtain load-indentation depth, time-indentation depth, and time-load graph for analysis.

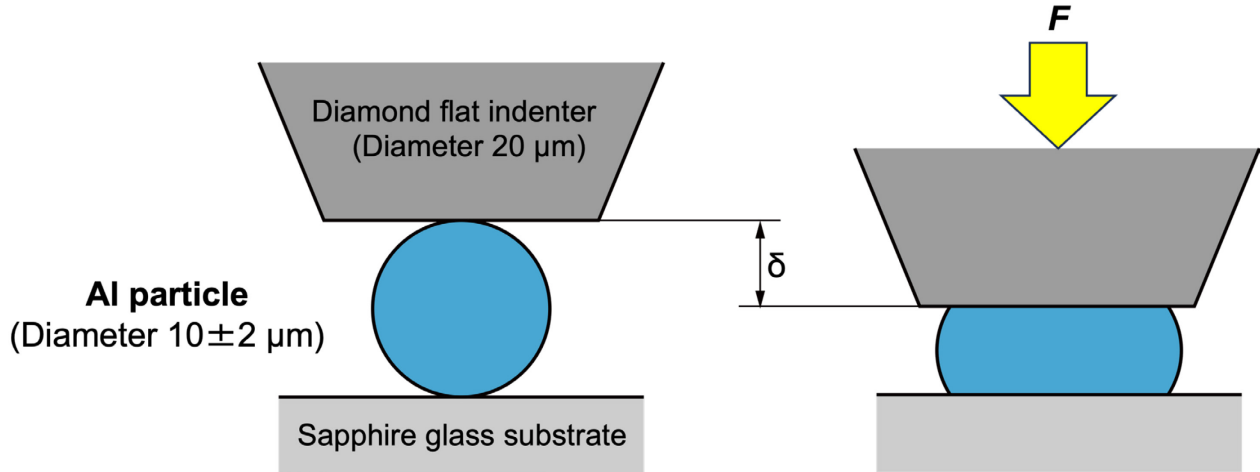


Figure 1. Schematic illustration of the particle compression test using impact nanoindentation,

2.3 SEM observation

The schematic image of a compressed particle is shown in **Fig. 2 (a)**. The compressed particle was coated by tungsten to make a protective layer and cut vertically by Hitachi FB-2200 FIB system to expose cross-section of the particle as shown in **Fig. 2 (b)**. The compressed particles such as shown in **Fig. 2 (a)** were then observed by SEM, Hitachi SU-70, at different angles for crack pattern identification. The deformed particles were then cross sectioned as shown in **Fig. 2 (b)** to study microstructure evolution.

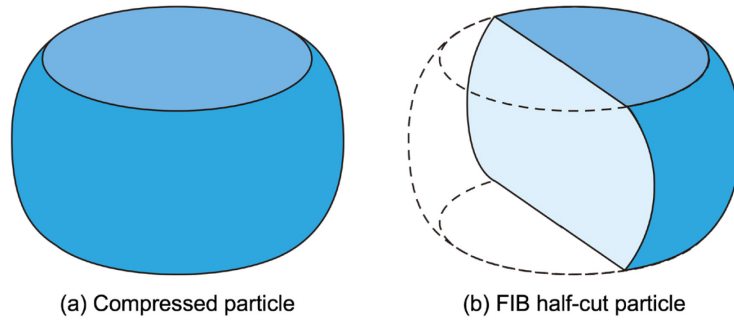


Figure 2. Schematic illustration of FIB fabrication of compressed particle, (a) as-compressed particle, and (b) vertically cut compressed aluminum particle.

3 Result and discussion

3.1 Prepared Particle

Fig. 3 shows the SEM image of heat-treated aluminum particles with and without zinc oxide coating. The cross-sectional observation results in **Fig. 3 (b)** and **(d)** were evaluated by backscattered electrons (BSE). In the BSE evaluation, the differences in composition and crystallographic orientation are indicated by differences in color. However, no grain boundaries appeared in the cross-section of the aluminum particles used in this study. TEM and EDX images of the monocrystalline aluminum particle with zinc oxide coating are shown in **Fig. 4**. In this figure, the outer shell of the particle is comprised of oxygen and zinc. This image shows that the zinc oxide is well coated on the aluminum particle.

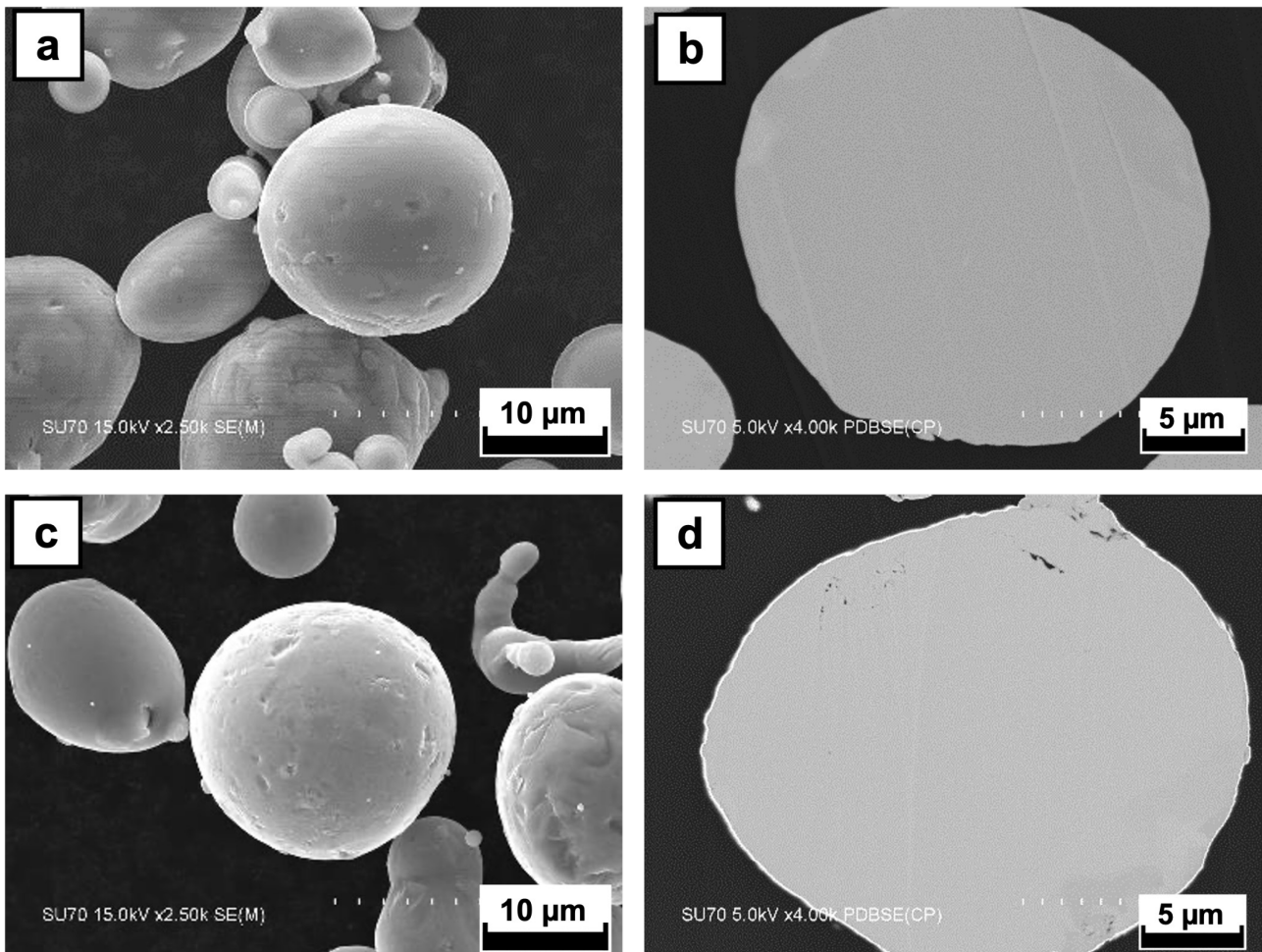


Figure 3. SEM image of tested Al particles, (a) SEM overview of monocrystalline Al particle without zinc oxide coating and (b) cross-section. (c) SEM overview image of monocrystalline Al particle with zinc oxide coating and its (d) cross-section.

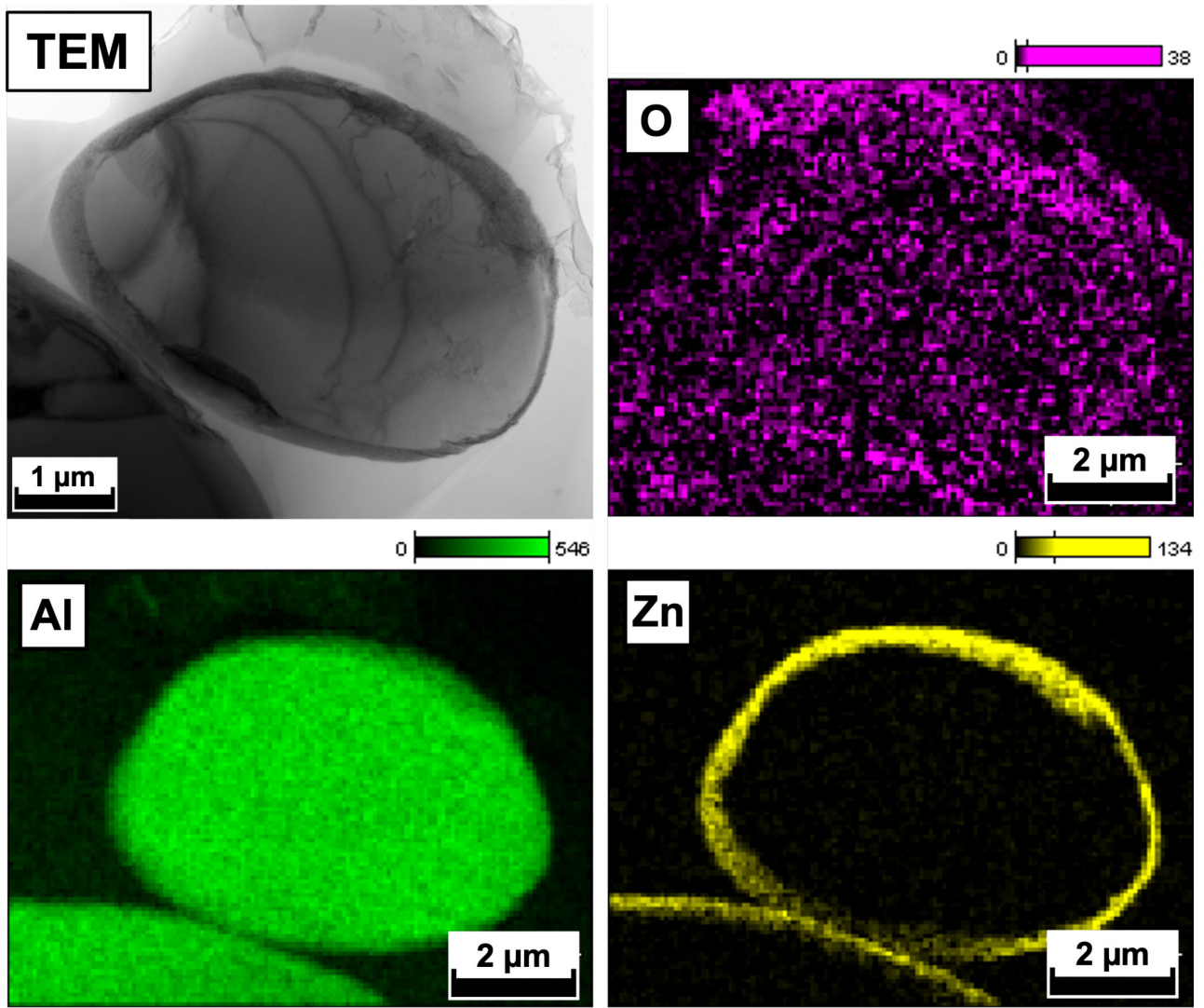


Figure 4. Cross-sectional TEM image and EDX mapping results of monocrystalline Al particle with zinc oxide coating.

3.2 Impact particle compression test

Monocrystal aluminum particles with and without zinc oxide coating with similar sizes were subjected to an impact nanoindentation test to yield the graphs shown in Fig. 5. In Fig. 5 (a) and (c), the compression depth of particles without oxide coating is higher than a particle with oxide coating under same experimental condition. This strongly agrees with our previous study of particle compression [16]. Nevertheless, the overall trend of the graphs of particles with and without oxide coating is similar, suggesting that the oxide coating does not significantly affect the deformation behavior of particles apart from the nominal displacement.

Fig. 5 summarizes the impact compression test results of both 10μm particles. **Fig. 5 (a)** shows that the compressive displacement increases almost linearly with time at the beginning of the test. And **Fig. 5 (b)** shows that the load returns to zero at approximately 400 μs for particles without coating and at 450 μs for particles with coating, indicating one impact test to this point. This point represents a single impact test. After this point, the particles no longer deform, and the indenter oscillates. Meanwhile, in **Fig. 5 (b)**, the load increases overall, although with several downturns before reaching its maximum level. Such a phenomenon is absent on the static test [14-16]. This indicates a force acting against the load on the indenter when the compression speed is high enough.

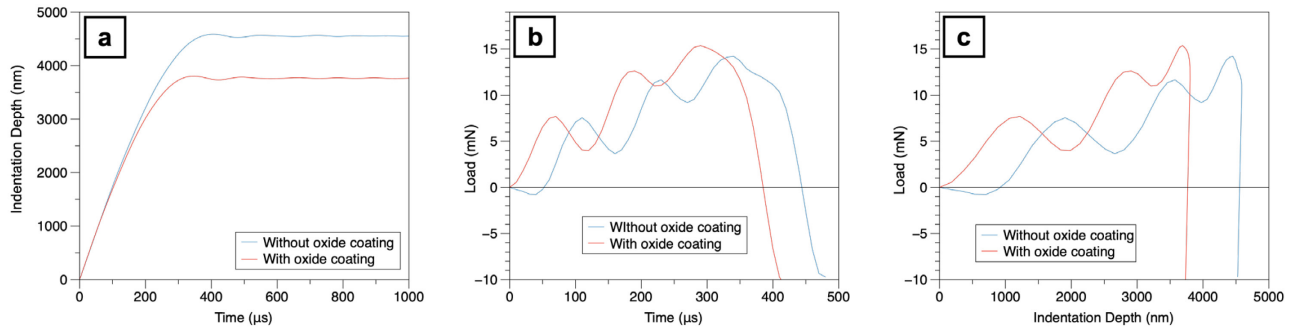


Figure 5. Impact particle compression results with and without zinc oxide coating monocrystal aluminum particles, (a) time-compression displacement, (b) time-load, and (c) compression displacement-load graphs.

3.3 SEM observation

A zinc oxide-coated monocrystal aluminum particle subjected to SEM observation is shown in Fig. 6. As shown in Fig. 6 (d), the particle develops a crack from its equator and reaches up to the edges of the flat upper surface. However, from the edge of the surface, the cracks no longer propagate and leave the middle of the flat area relatively intact as shown in Fig. 6 (b). These results agree with a study regarding oxide film fracture patterns at the interface of cold spray coating [10]. As this study suggests, the oxide film fracture is minimal at the so-called south pole of the particle, which corresponds to the middle of the flat surface. Also, the place where the best bonding was achieved corresponded to the place of crack initiation in this study. However, the tendency of crack initiation and propagation differs from the results of previous studies, which were obtained at low strain rates [16]. The reasons for this difference and the detailed morphology of the cracks need further investigation.

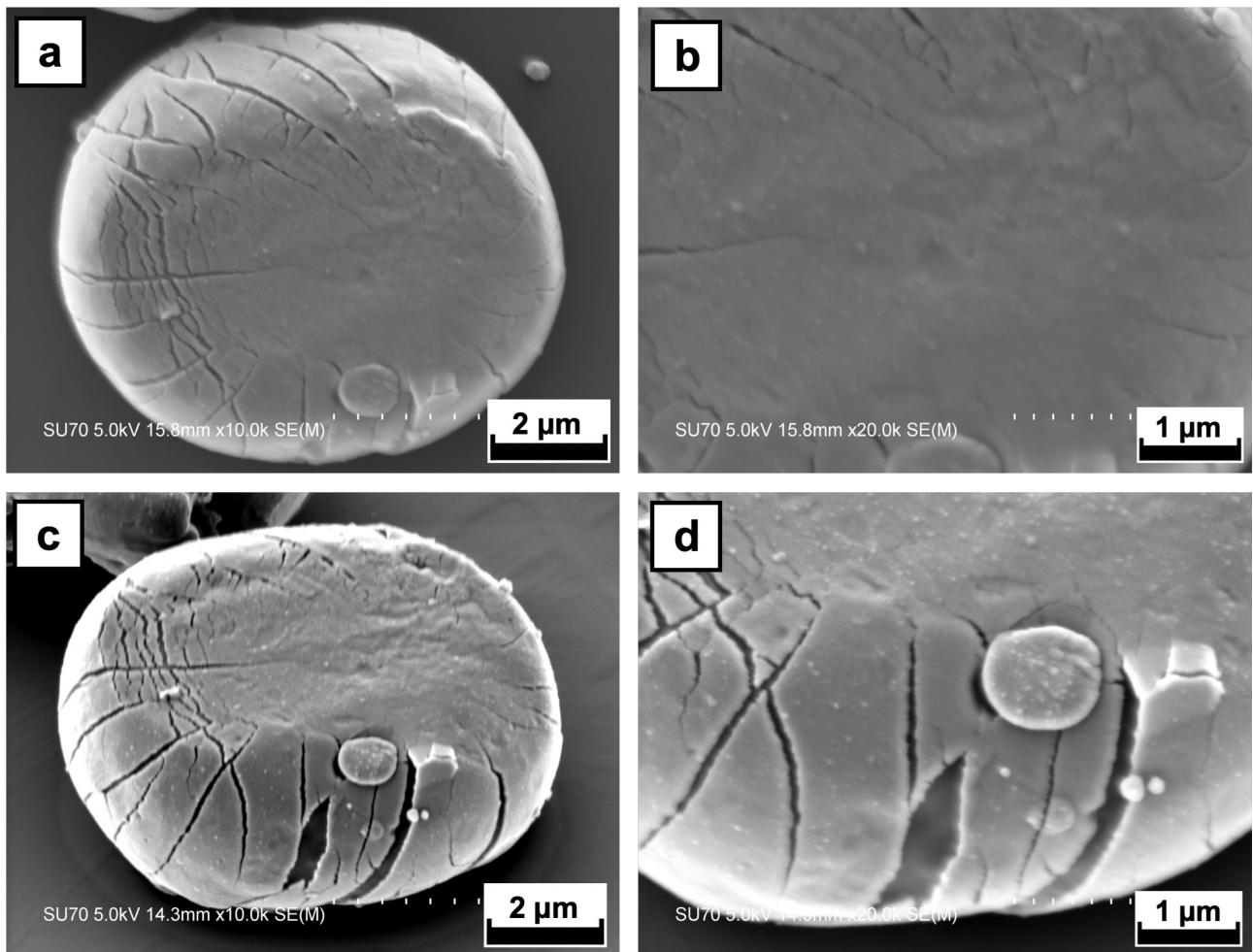


Figure 6. SEM observation of zinc oxide coating monocrystal aluminum particles after compression. (a) and (b) shows the image of the particle with 0° tilt at 10.0 k and 20.0 k magnification respectively. (c) and (d) shows the particle with 45° tilt at 10.0 k and 20.0 k magnification respectively.

The particle was sputtered by FIB and the cross-section was observed as shown in **Fig. 7**. In **Fig. 7 (b)**, the red region of **Fig. 7 (a)** was magnified with high contrast. **Fig. 7 (b)** shows a pattern which indicates the polycrystalline structure of the particle. This suggests that recrystallization has taken place during the compression test. Detailed analysis using EBSD, and other methods will be necessary in the future.

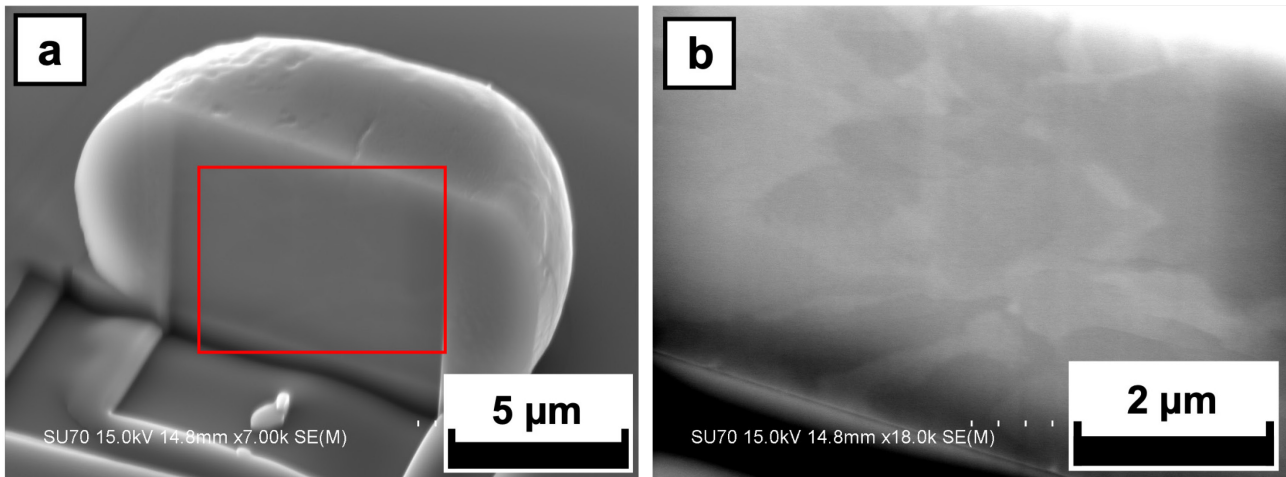


Figure 7. Cross-section observation of the particle after compression. (b) shows high magnification image of red region of (a).

4 Conclusion

In this study, we have carried out a set of experiments to analyze the deformation behavior of Aluminium particles with and without zinc oxide coating under high-speed compression. The oxide layer on the particle decreased the flattening of the particle but did not have a significant effect on the general deformation behavior. The result obtained by NanoFlip clearly showed that the deformation behavior of particles under the impact test differs from the static compression test. The impact test showed peculiar downturns in the load-indentation displacement plot, which we believe is due to the recrystallization process. The observation using SEM strongly supports that the declines in load are due to recrystallization behavior during the compression. Further study will be done employing EBSD to analyze the crystal structure of the compressed particle. We believe that this study will prove valuable in the field of cold spray technology.

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