In-situ analysis of the cooperative material transport during sintering of copper powder

Introduction

Sintering of metals is usually explained by a two particle model which describes neck growth and centre approach due to the attempt to minimize the surface energy. Unfortunately the model is incomplete if applied to 3D samples. In addition to a particle centre approach, cooperative material transport occurs in 3D - the movements of entire particles relative to surrounding particles, for example particle rotations [1]. These rotations are usually attributed to tensions due to asymmetric interparticle contacts [2], inhomogeneous centre approaches [2] and the necessity to form low energy grain boundaries [3].
The existence of particle rotations was shown by Herrmann and Gleiter [4] using a sphere plate experiment, by Wieters using rows of spheres [1] and by Exner using 2D arrangements of monocrystalline spherical copper particles. However, inside 3D specimens the cooperative material transport remains nearly unexplored.
Computer tomography, especially high resolution synchrotron computer tomography (SCT), is a suitable method that permits an investigation of particle movements in 3D samples. First results were obtained by ex-situ analyses [5, 6]. These experiments proved the existence of particle rearrangements in the 3D case. Furthermore, it became clear that the rotation of particles in 3D specimens can be analysed quantitatively using high resolution SCT in combination with photogrammetric image analysis. It becomes possible to measure rotation processes inside of 3D specimens and an extended database of experimental data will enable to improve existing computational simulations of sintering processes.

Results

Two specimens consisting of monocrystalline spherical copper particles with sizes of either 56-60 µm (sample 1) or 72-80 µm (sample 2) were analyzed using synchrotron tomography at the ESRF (ID15A) in July 2007. About 3000 (sample 2) or 8000 (sample 1) spheres were pre-sintered at 600°C in silica glass capillaries with a diameter of 1.3 mm. The glass capillaries were removed prior the measurements to avoid constrained sintering. The measurements took place in a special in-situ furnace in a reducing atmosphere. The heating rate was 10 K/min from room temperature up to 1050°C. Subsequently to a dwell time of 10min the specimens were cooled down. The acquisition of an entire tomogram required 50 seconds.
A third specimen consisting of monocrystalline copper with a particle sizes of 160-200 µm was measured in a glass capillary from room temperature till 1050°C. Each sphere was marked with a 5x5x5 µm hole manufactured by a focus ion beam (FIB). These markers enable to measure the orientation of each sphere.
In order to analyze the behaviour of each copper particle it is necessary to determine the accurate positions of all particles and to track them through the entire sintering process. Therefore, grey value lines are calculated from the centres beyond the surfaces of the spheres to detect the surface points. The resulting centres and radii of the particles are determined with a minimum precision of 0.3 voxels by regression analysis. Each particle can be traced during the entire sintering process and the coordination partners as well as the local density can be determined.
The particle rotation to each other is the most important aspect of this study of sintering processes in 3D. The rotation of a particle is calculated by the variation of the angles between the respective particle and its persistent contact partners. In this case the cumulative rotation of the particles is used, i.e. the rotation is always referenced to the first sintering step.
Fig. 1 (sample 1) and Fig. 2 (sample 2) show the cumulative rotation angle and the centre approach of the particles plotted versus the temperature. The centre approach as a scale of the contribution of the two particle model to the

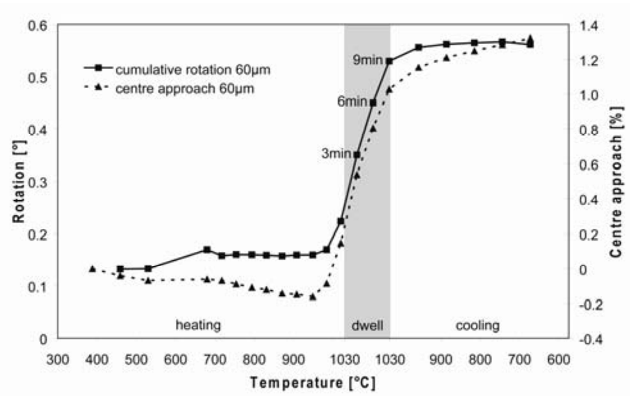

Fig.1: Rotation and centre approach vs. temperature; 56-60µm

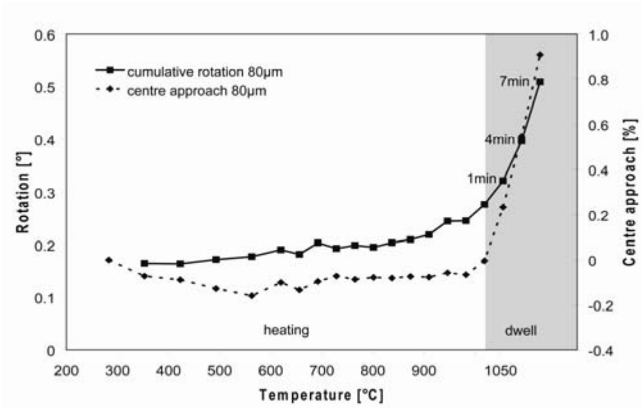

Fig.2: Rotation and centre approach vs. temperature; 72-80µm

densification was measured by the average distance to the next coordination partner of all particles. The inherent bias of the rotation measurement caused by the error of the image analysis is about 0.13° in both specimens. The calculated average of the rotations shows a statistical error in the range of $1*10^{-3}$ to $6*10^{-3}$ degrees.
Specimen 1 shows no cumulative rotation below 1000°C. At higher temperatures and during the dwell time the alteration rate of the cumulative rotation shows a steep increase. During the cooling no significant alteration of the rotation was observed. The centre approach shows a similar curve progression.
Specimen 2 shows an insignificant increase of cumulative rotation up to about 950°C. With further increase of temperature including dwell time the alteration rate of the rotation shows an extensive growth. The densification due to particle centre approach, as well as the macroscopic shrinkage of the specimen start almost immediately after the increase of rotation.

Fig.3 (sample 3) shows a FIB hole in each sphere. The analysis of the specimen is not yet completed, but it will be possible to detect the exact position of the holes.
At high temperatures the material transport mechanisms contributing to the observed sintering processes are intensified. Consequently both, a measurable particle centre approach and rotations can occur.
Thus, by recent improvements of spatiotemporal resolution of SCT and photogrammetry it has become possible to investigate quantitatively the structural developments in sintering samples.

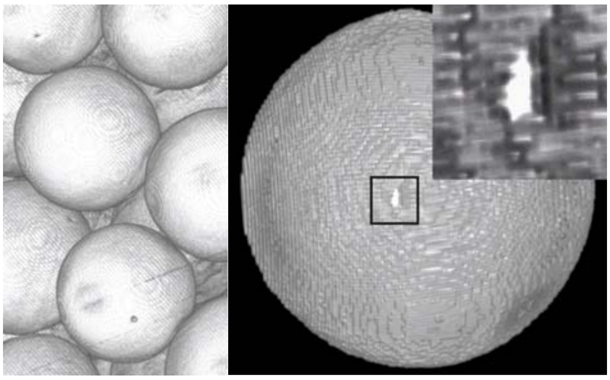

Fig.3: Tomogram of copper spheres with FIB holes

Conclusion

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