IN-SITU INVESTIGATION OF THE COOPERATIVE MATERIAL TRANSPORT DURING THE EARLY STAGE OF SINTERING BY SYNCHROTRON X-RAY COMPUTED TOMOGRAPHY

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ABSTRACT

A better fundamental understanding of the sintering process requires experimental data describing in 3D particle rearrangement processes, especially particle rotations. Therefore, synchrotron computed tomography (SCT) is used to investigate the particle rearrangements during sintering. The specimens consisting of spherical monocrystalline copper powder were measured in a special in-situ furnace. The data is analyzed by a custom image analyzing software based on photogrammetric methods to determine the coordination, the centre approach and rotation of each particle in a 3D specimen. The sintering of monocrystalline copper spheres analyzed using SCT is compared to literature data of different test arrangements and dimensions.

INTRODUCTION

Sintering is the most essential processing step of powder metallurgical production. Heat treatment of an assembly of loose or compacted particles results in the growth of interparticle bonds and shrinkage of the component. The driving force for the growth of sintering necks is the need to minimize the surface area and surface energy. The material transport takes place by diffusion of vacancies from the neck surface to the contact grain boundary or the particle surface. These mechanisms have been investigated since the 1940's and are now well understood. Extensive reviews have been written by J.E. Geguzin[1], H.E. Exner[2, 3], W. Schatt[4] and R.M. German[5].

Beside neck growth, cooperative material transport, e.g. by rotation, also contributes to the densification of sintered components. In the literature, asymmetric sinter necks[4], tensions due to non-uniform neck growth[6] or enhanced grain boundary energies due to the different crystallographic orientations of the contacting particles[7] are discussed and thought to be the major driving forces for rotations. However, a fundamental understanding of these processes has not been achieved because of the inaccessibility of experimental data by conventional analysis. To investigate these rotations in three-dimensional samples, high-resolution synchrotron X-ray computed tomography (SCT) is the only adequate method. First investigations of the sintering process using SCT have been made by A. Vagnon et al.[8] and O. Lame et al.[9]. Measurements of the cooperative material transport during sintering have been made by M. Nöthe et al.[10].

By combining synchrotron tomography with methods of photogrammetric image analysis it becomes possible to perform in-situ measurements and to obtain quantitative data of the particle rearrangement during sintering inside of three-dimensional specimens.

EXPERIMENTAL

Three-dimensional specimens for tomographic measurements have been produced from monocrystalline spherical copper particles. These particles were produced based on the Sauerwald process[11] from polycrystalline copper powder provided by the company ECKA Granulate GmbH & Co. KG. Selection of the spherical particles was accomplished by rolling the particles down a tilted glass panel or mirror. Asymmetric particles rolled down in a curve caused by their geometry and could be rejected.

In preparation for the tomography measurements the loose packing of particles with radii of approximately 30µm were filled into a 1.3mm diameter silica capillary and fixed in a pre-sintering step at 600°C. After this step, the capillary was removed to achieve free sintering without any external influences during the following measurements. Preliminary measurements of samples in silica capillaries showed an approximately linear increase of rotation, density and centre approach as a result of the different thermal expansion coefficients of silica and copper. Thus measurements of free sintering samples are necessary.

The synchrotron tomography measurements were conducted at the European Synchrotron Radiation Facility (ESRF) in Grenoble (France) at the beamline ID 15 A. The use of a white beam (polychromatic radiation) allows the measurement of 851 radiograms within a time of just 56 seconds which is sufficient to obtain one entire tomogram of high quality. A complete turn including the radiograms, 51 flatfield measurements, the turn back of the rotation stage and the readout of the CCD-camera required 3 minutes and 32 seconds. Due to the short measuring times it was possible to heat the specimens up continuously in a special in-situ furnace during the investigations. Two specimens were sintered with a heating rate of 10K/min. One of the samples was sintered with additional dwell times of one hour at 650°C, 750°C, 850°C, 950°C and 1050°C.

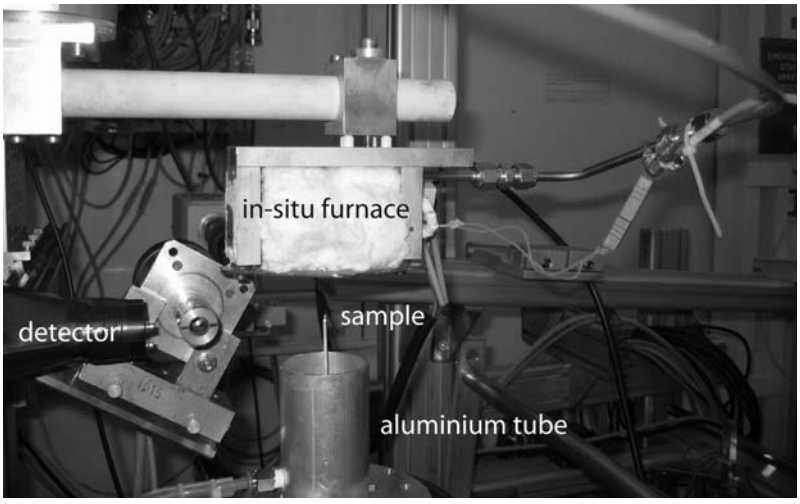

Figure 1: Tomography setup including the in-situ furnace. Here, the furnace has been lifted to change the sample.

The sintering process took place in a reducing atmosphere consisting of 96% He and 4% H_2. Since a rotation of the specimen was necessary a hole at the bottom of the furnace was required. Therefore an aluminium tube was installed from the manipulation stage close to the bottom of the furnace (Figure 1). By passing inert gas into the bottom of the aluminium tube it was possible to achieve a small overpressure in the tube and in the furnace chamber. This results in a gas flow out of the 3mm gap between tube and furnace and no oxygen could enter into the furnace chamber which enables a constant reducing atmosphere.

The analysis of the measured data was performed by custom made image analysis software based on photogrammetric methods. This software allows detecting the centre positions of all copper spheres in a tomogram by searching for homogeneous spherical areas exceeding a given grey value[12]. The radii of these areas have to be slightly smaller than the radii of the measured spheres. The results are the approximate particle centres. In a next step, the precise positions of surface points are determined by interpreting virtual grey value lines from the detected centres to beyond the surface of the particles. Using these determined points the exact positions of the centres can be calculated.

Furthermore the software is able to detect coordination partners and can track every single sphere across several sintering steps.

Using the given data it is possible to investigate the rotations of the particles with respect to their coordination partners, the relative density, the centre approach and the shrinkage of the specimen. The contact partners are detected by analysing the grey value lines between a particle and its nearest neighborhood. The rotations are calculated by fitting each sphere and its coordination partners to their positions in another sintering step and evaluating the average change of the angle between the coordination partners. The error of the analysis is about 0.075 voxels. This corresponds to approximately 0.1°. The centre approach is calculated by the distance to the next coordination partner. Thus, new and broken contact partners during sintering are excluded from the analysis.

RESULTS AND DISCUSSION

Figure 2 shows the cumulative rotation in two specimens exposed to a heating rate of 10K/min. The figure displays almost no rotation up to 950°C. At this temperature the sample with dwell time shows first small rotations. Only above the dwell time at 1050°C a significant increase of cumulative rotation occurs. However, the value of the rotation of less than 0.7° after one hour dwell time is still very small compared to measurements on a plate-sphere-model[13] or the one-dimensional measurements on a row of monocrystalline copper spheres by K.-P. Wieters. In both cases rotation values of up to 25° have been detected. Furthermore, these big rotations were measured between 600°C and 900°C[14]. In this range of temperature the sintering contact areas are still very small and the diffusion rates increase. Thus the rotations driven by misorientations of the contact partners increase as well. Caused by the high degree of freedom the driving force was sufficient to initiate some rearrangements. In this study, the particles possess between 6 and 7 contact partners on average (see Figure 4). Because of these high coordination values of the particles the driving forces counteract one another and, as a result, hamper particle rearrangements. Furthermore, the particle rearrangement shown in Figure 2 is correlated with the particle centre approach (see Figure 3). Thus, the contribution of the rotation shown in Figure 2 due to the misorientations in the contact areas is very small, but occurs by unequal centre approach during sintering at high temperatures. The rearrangement caused by misorientations can be negligible during solid state sintering in three-dimensional specimens.

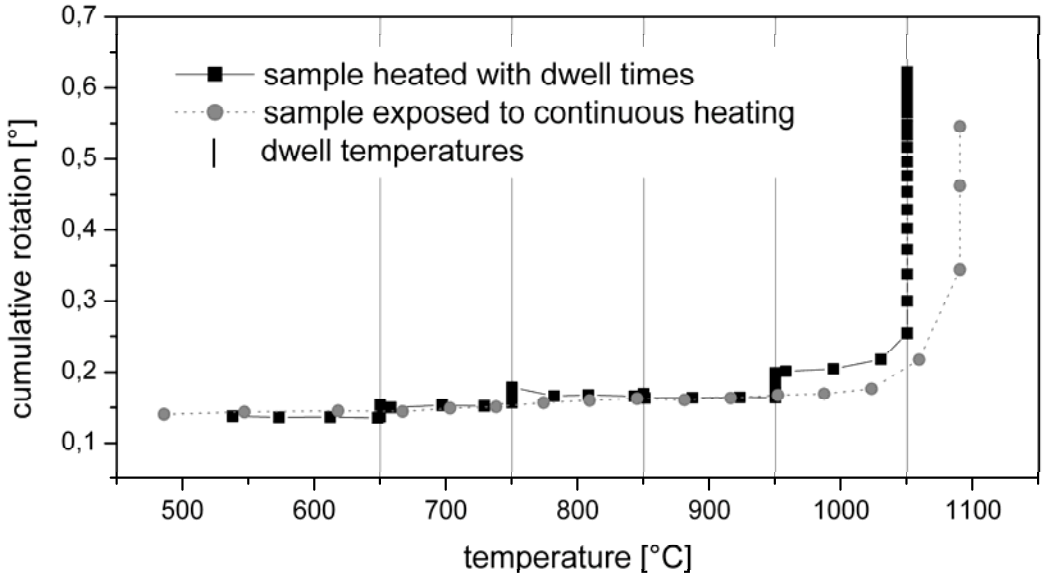

Figure 2: Differential and cumulative rotation vs. temperature. Monocrystalline spherical copper particles with radii of 30µm. Heating rate: 10K/min. Dwell time 60min for each temperature.

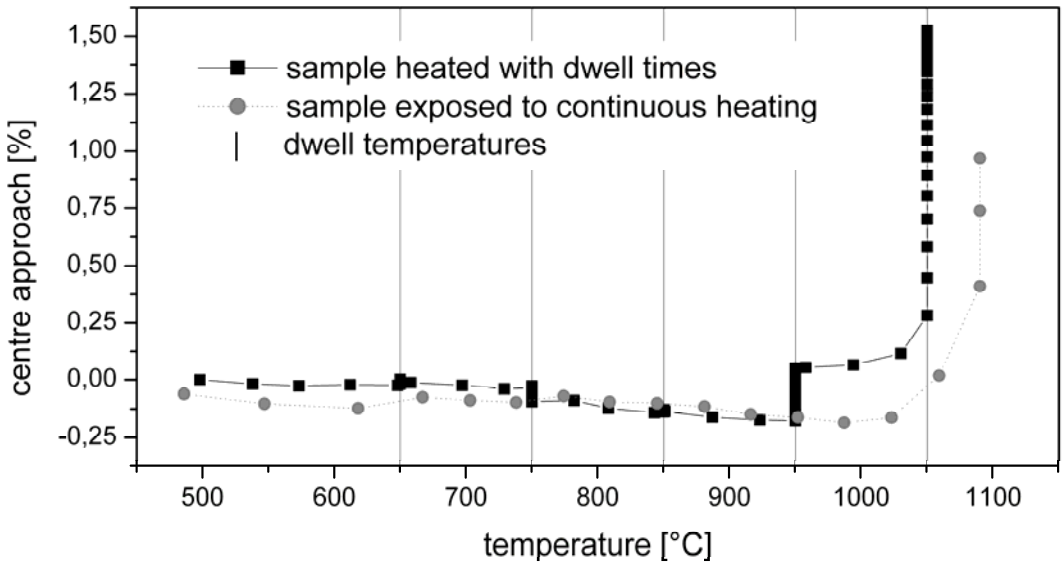

Figure 3: Centre approach vs. temperature. Monocrystalline spherical copper particles with radii of 30µm. Heating rate: 10K/min. Dwell time 60min for each temperature.

Thermal expansion has already been subtracted from the curves in Figure 3. The small decrease up to 950°C can be explained by the small rotation values in the same temperature range (Figure 2). Above 950°C the centre approach starts and is stronger than the potential expansion induced by particle rearrangements. A steep increase follows at 1050°C.

During heating up to 950°C no significant difference between the two specimens could be detected. The continuously heated sample shows a centre approach and rotation above 1000°C similar to the specimen heated with dwell times above 950°C. The reason for this behavior is the dwell time of 60min. In this additional time centre approach at lower temperatures and a simultaneous rotation takes place.

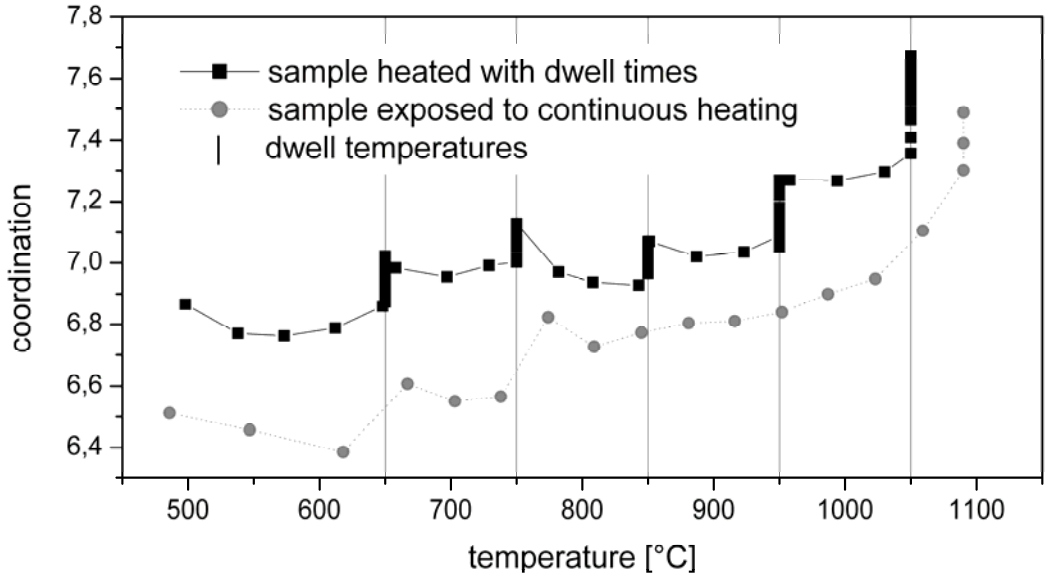

Figure 4: Number of contact partners vs. temperature. Monocrystalline spherical copper particles with radii of 30µm. Heating rate: 10K/min. Dwell time 60min each.

The sample sintered with dwell times shows a decrease of coordination at the beginning of each heating period up to 950°C. This behavior is correlated with the negative centre approach in the mentioned temperature range. This demonstrates that even very small rotation values below 950°C influence the sintering process. The influence of the cooperative material transport during the heating periods exceeds the centre approach. In contrast, the centre approach during dwell periods is more intense than the dilatation of the specimen caused by cooperative material transport.

CONCLUSION

High resolution synchrotron tomography was found to be an adequate method to investigate the cooperative material transport in three-dimensions.

Results obtained on one-dimensional samples cannot be transferred to 3D. The reason is the number of contact partners. One-dimensional specimens have fewer restrictions regarding their degrees of freedom, while a particle in a three-dimensional sample cannot rotate without moving its contact partners too. This is why the measured rotations are comparatively small.

Further experiments showed a kind of intrinsic rotation of the particles during sintering around their own centre. This kind of rotation cannot be measured with the presented method. Therefore, in new specimens each sphere has been marked using a Focused Ion Beam. By tracking these markers the measurement of self rotations becomes possible. Results of these further measurements will be published soon.

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