

True Sedimentation and Particle Packing Rearrangement during Liquid Phase Sintering

Jong K. Lee¹, Lei Xu² and Shu Zu Lu³

¹Dept. of Materials Science and Engrg., Michigan Technological University, Houghton, MI, USA

²Dept. of Materials and Metallurgical Engrg., New Mexico Institute of Mining and Technology, Socorro, NM, USA

³School of Materials Science and Engrg., University of Science and Technology Beijing, Beijing, CHINA
^ajk1103@mtu.edu, ^blxu@nmt.edu, ^cszlu@mater.ustb.edu.cn

Abstract

When an alloy such as Ni-W is liquid phase sintered, heavy solid W particles sedimentate to the bottom of the container, provided that their volume fraction is less than a critical value. The sintering process evolves typically in two stages, diffusion-driven macrosegregation sedimentation followed by true sedimentation. During macrosegregation sedimentation, the overall solid volume fraction decreases concurrently with elimination of liquid concentration gradient. However, in the second stage of true sedimentation, the average solid volume fraction in the mushy zone increases with time. It is proposed that the true sedimentation results from particle rearrangement for higher packing efficiency.

Keywords: liquid phase sintering, macrosegregation, composition gradient, sedimentation

Diffusion-driven Sedimentation and True Sedimentation

In liquid phase sintering (LPS), dissimilar metal or ceramic powders are mixed and pressed to form a green compact of the desired geometry. In the presence of gravity, LPS with a relatively low volume fraction for solid phase produces nonuniform structures. When the solid phase is denser than the liquid phase, the solid phase sinks, whereas it floats to the top of the sample if it is lighter. Therefore, a liquid zone emerges either above or below the two-phase zone (mushy zone). The extent of the mush zone diminishes with the sintering time; i.e., the mushy zone sediments. This is termed sedimentation. Sedimentation is ubiquitous in LPS, provided that the solid phase volume fraction is less

than a critical value. This work proposes a mechanism responsible for the later stage of sedimentation, *true sedimentation*. Courtney et al. [1-3] proposed a model to explain observed sedimentation phenomena: the sedimentation was ascribed to macrosegregation induced by the initial settling of particles and then ensuing elimination of the composition gradient in the liquid zone by diffusion. However, after the composition gradient is eliminated in the liquid zone, sedimentation can still be observed with an increased sintering time. An illustrative schematic is shown in Fig. 1. Imagine, for example, sintering a Ni-65 wt.% W alloy. For an initial sintering, the sample would show an interface between the liquid zone and the mushy zone, as shown in Fig. 1a. Since the initial melting involves a minute dissolution of W, the liquid composition is practically

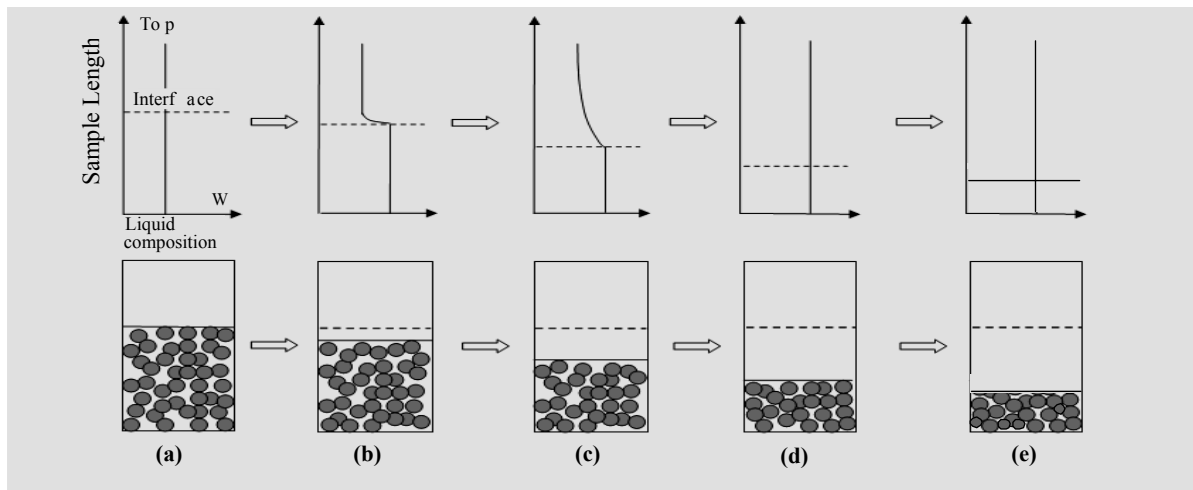


Fig. 1. Schematic for structural development in liquid phase sintering when gravity-induced macrosegregation occurs.

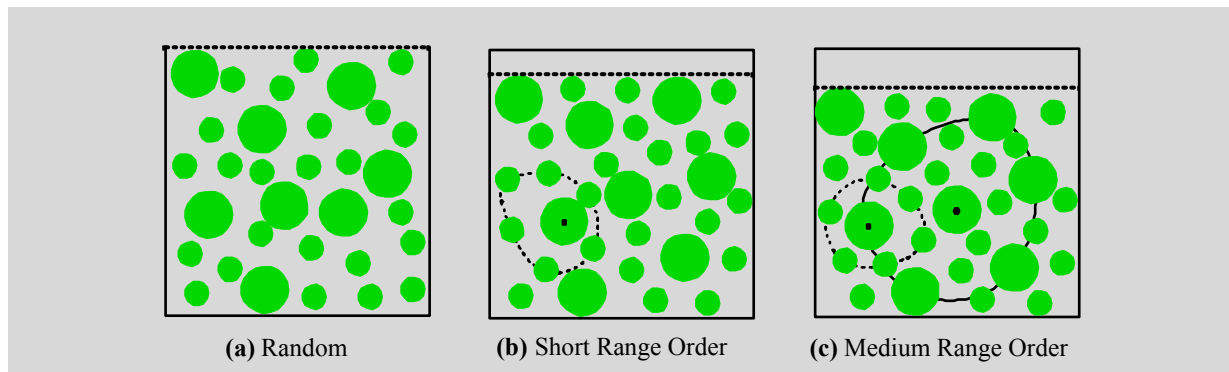


Fig. 2. Schematic for particle distribution from random to short range order and then to medium range order during true sedimentation.

uniform throughout the sample length: this is depicted as a vertical line in the upper figure of Fig. 1a.

After the settling of W particles and formation of a skeleton structure, some solid W dissolves into the immersed liquid until the equilibrium liquidus composition is reached. The liquid in the liquid zone has a nearly pure Ni composition, whereas the liquid in the mushy zone has the liquidus composition. Consequently, a sharp composition gradient arises across the interface between the liquid and mushy zones (Fig. 1b). The stage that takes most of the sintering time happens hereafter. The composition gradient is gradually reduced and eventually disappears mainly through the long-range liquid diffusion of W (Figs. 1c and 1d). Even after the composition gradient is eliminated in the liquid phase, sedimentation can still be observed with an increased sintering time. This is shown as a further-reduced level of the mushy zone in Fig. 1e compared to that in Fig. 1d. This retreat of the mushy zone in the absence of a composition gradient is termed *true sedimentation*. Several possible mechanisms were suggested for the true sedimentation: they are directional particle coarsening, mushy zone deformation [4], and solid state sintering, but none of them is satisfactory.

There has been a great progress on the phase transition behavior of colloidal suspensions. In particular, Sirota et al. [5] found that as the volume fraction of polystyrene spheres (of ~ 100 nm diameter) increases, the system undergoes structural changes from liquid to a colloidal crystal of bcc at first and then to that of fcc. Certainly, not all of the results on colloidal particles are applicable to LPS, as the colloidal particles have Brownian motions. Nevertheless, the increase in the packing density from the disorderly liquid to bcc, and

to fcc appears to have a strong correlation to the sedimentation behavior in LPS. Further, the work of Krichevsky and Stavans [6] on the correlation effects during Ostwald ripening demonstrates a good evidence that there takes place some ordering activities, i.e., packing rearrangement among the settling solid particles in the mushy zone. Figure 2 presents a schematic in which, due to the Gibbs-Thomson effect, particle rearrangement may occur from random to short range order then to medium range order distribution during true sedimentation. For simplicity, a bi-disperse particle system is depicted, where each of the large particles is surrounded by small particles in a short range order distribution (marked with a dotted curve) and then large particles develop a loose network in a medium range order distribution (indicated with a solid curve).

References

1. T. H. Courtney, Y. Du, and S. Z. Lu, *Metall. and Mater. Trans.*, 31A (2000), 2907.
2. Y. Du, T. H. Courtney, and S. Z. Lu, *Acta Mater.*, 51 (2003), 445.
3. L. Xu, S. Z. Lu, and T. H. Courtney, *Scripta Mater.*, 46 (2002), 887.
4. Y. Liu, D. F. Heaney, and R.M. German, *Acta Mater.*, 43 (1995), 1587.
5. A. K. Sood, *Solid State Phys.*, 45 (1991), 1.
6. O. Krichevsky and J. Stavans, *Phy. Rev. E.*, 52 (1995), 1818.