Monte Carlo Potts Simulation of Grain Growth of Solid Grains Dispersed in a Liquid Matrix

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Abstract— Liquid phase sintering is a process in which solid grains coexist with a liquid matrix. This process has important applications in processing of several engineering materials. Examples of these applications are high-speed metal cutting tools, alumina substrates for packaging silicon chips and barium titanate electrical capacitor. Grain growth in liquid phase sintered materials occurs by Ostwald ripening. The purpose of this paper is developing Monte Carlo Potts model to simulate Ostwald ripening in liquid-phase sintered materials. Ostwald ripening is simulated by treating two phases, solid grains dispersed in a liquid matrix as a two-dimensional square array of sites. Each site of the solid-phase grains (phase A) is given a random positive number between 1 and Q where Q=100 for all the simulation. The sites of the liquid phase (phase B) are assigned only one negative number, q_B = -1. It is found that the grain growth is controlled by volume diffusion for volume fraction of the solid grains ranging from 40% to 90%. The grain growth exponent has the value, n=3, in agreement with the theoretical value of Ostwald ripening.

Index Terms— Monte Carlo, Potts model, grain growth, grain size, sintering, Ostwald ripening.

I. INTRODUCTION

Practically all engineering ceramics are sintered by a liquid phase, which is a viscous glass. In this state, the solid grains solubilize in the liquid causing wetting of the solid. Consequently, an emergent capillary force attracts the grains together. Thus, liquid phase sintering has essential applications in materials processing and enhancing their properties for commercial applications. The microstructure of liquid phase sintered materials evolves with time by Ostwald ripening. Ostwald ripening is a process in which small grains in a liquid matrix decrease in size until they vanish while large grains grow and the mean size will increase with time while the number of grains will decrease. In such a process, the interfacial area reduction drives the diffusional mass removal from areas of high interfacial energy to areas of low interfacial energy.

Lifshitz, Slyozov and Wagner [1,2] have theoretically investigated Ostwald ripening process named LSW theory for volume fractions close to zero. They assumed that the second phase grains are spherical and the mean distance between the grains is large compared with their dimensions. This implies that the interaction between the grains may be ignored and the volume fraction of the dispersed phase is very small. The LSW theory predicted that in the long time limit the average grain radius asymptotically changes with time as t1/3. The LSW theory has provide a way to find the interfacial energy between the matrix and the dispersed phase. The LSW approach has a difficulty to be tested by experiment or computer simulation because it is limited to zero volume fraction of the growing phase while experiments investigate finite volume fractions. Several authors [3-6] have attempted efforts to modify the LSW to study the effect of volume fractions on the growth behavior. They found that the variation of grain size with time does not depend on the volume fraction.

The Ostwald ripening driven mass transfer may significantly change the microstructure of two-phase materials. This change in the microstructure happens by means of small grains shrinking and moving their mass to large grains. Consequently, the average grain size increases with time and the number of the second phase grains drops with time.

The materials microstructure affects their mechanical properties; therefore, it is very important to understand the change of materials microstructure to improve their performance in industrial applications. Because Ostwald ripening controls materials microstructure, we have to study Ostwald ripening process to understand the microstructural evolution of materials. The theoretical methods used to model grain growth [7] and Ostwald ripening [1,2] made many simplified assumptions to make the problem easy to understand. To diminish these simplified assumptions, several analytical models have been developed [8-20].

The main purpose of this paper is to develop a numerical approach for simulating Ostwald ripening in system, which has conserved volume fractions. It is based on our Monte Carlo Potts computer simulation

model for grain growth in two-phase systems [10-11]. Not only this Monte Carlo Potts model permit one to display the microstructural evolution during Ostwald ripening but also to get information about average grain size and grain size distribution. The emphasis will be on the microstructural evolutions and on their comparison to experimental observations.

Furthermore, the grain size variation with time during Ostwald ripening will be discussed. For Ostwald ripening of second phase grains embedded in a matrix, it is shown that the mean grain size varies with time as t1/n, where n is the grain growth exponent. The growth exponent for Ostwald ripening is n=3 in case of volume diffusion controlled growth [1,2]. Base on the computer simulation results, the growth exponent will be determined.

Next section will cover the simulation method. Section III is devoted to cover Otwald Ripening simulation, followed by conclusion.

II. SIMULATION METHOD

Monte Carlo Potts model implemented by Solomatov et al. [11] to study grain growth in two-phase systems was developed to study the grain growth in liquid phase. The structure of the solid phase in the liquid phase is initialized using a 400×400 square array of sites. A site represents a domain with a specific orientation of crystalline lattice. The orientation of the individual grains is described with the help of spin.

The initial microstructure used in this study was generated by randomly occupying the lattice with the desired volume fraction of phase A and phase B. Phase A represents the solid polycrystalline material and phase B is the liquid matrix. Each site of phase A is given a random number between 1 and Q while the sites of phase B are given only one state represented by a negative number -1. The two phases were produced by defining boundary energies between sites of phase A and the sites of phase B so that the components can separate into two phases. Eab is the solid-liquid (A-B) interfacial energy, Eaa is the grain boundary energy between two solid grains (A-A) of different orientation. The liquid phase cannot have liquid-liquid interfaces in the simulations, Ebb=0. The total interfacial energy of the system is given by,

$$E_{total} = \frac{1}{2}\sum_{i=1}^{N}\sum_{j=1}^{N} E(i,j)\left[1-\delta\left(q_i - q_j\right)\right] \qquad (1)$$

where E(i, j) is the boundary energy between site i and j, q_i = -1 is the spin of the i^{th} site. N is the total number of lattice sites. δ is the Kronecker delta function. The interfacial energy Eab is higher than Eaa or Ebb. The total number of the sites of phase A and phase B kept constant.

The microstructure evolves as a result of Ostwald ripening process which transfer mass from one grain of phase A to another grain of the same phase through the liquid matrix (phase B). This is numerically done as follows. A site is randomly selected and its neighbor is randomly selected from its eight nearest neighbors. If both sites belong to different phases, the site of phase A is flipped to a new orientation. The orientation randomly selected from 1 to Q. Then, the A- and B- sites are allowed to exchange their spins. In order to check if the new orientation is accepted or rejected, it is necessary to use the exchange probability function determined by Boltzmann statistics

$$P(\Delta E) = \begin{cases} 1 & \Delta E \le 0 \\ \exp(-\dfrac{\Delta E}{kT}) & \Delta E \ge 0, \end{cases} \qquad (2)$$

where k is the Boltzman constant and T is the temperature. A random number between 0 and 1 is selected. If the number is less than P(ΔE), the move will be accepted; otherwise, it will be rejected. After each attempt, the time is incremented by 1/N, where N is the total number of lattice sites. The time is given in units of Monte Carlo steps (MCS) which represents N attempted orientaions.

III. SIMULATION OF OSTWALD RIPENING

The microstrucral evolution of Ostwald ripening is controlled by the ratio of the solid-solid and the solid-liquid boundary energies, Eaa and Eab. Therefore, Ostwald ripening was simulated by using the following values, Eab=1.0, Ebb=0.0 and Eaa=2.5, Q=100 and grain volume fractions ranging from 40% to 90%.

Figure 1 displays the simulated microstructures at several times for different volume fractions of solid grains (phase A) at the value of T equal to 1.3. The solid grains are white and the liquid phase is gray. These microstructures are quite similar to experimenal results [21]. Evidence of Ostwald ripening is the decrease of the number of grains and the increase of the mean grain size with time. This evolution in microstrucure occurs due to the fact that small particles eventually shrik and disappear and their atoms transfere to large grains by volume diffusion. Figure 2 exhibits the dependence of mean grain size on time. The mean grain size and the standard deviation are calcluated after running each case ten times.

The standard deviation is not indicted because it is found to be very small. As can be seen from Figure 2, the system goes through a transitional regime and eventually approach the self-similar regime after time varies between time t ≈ 103 - 104 depending on the case of solid fractions. We can find the value of the growth exponent by taking the slope of the curves

shown in Figure 2.

The growth exponent is very sensitive to change in time; therefore, the slope is calculted in a time window with width equal to 5. The slope is plotted as a fuction of time in Figure 3. It can be seen from Figure 3 that the slope in the self-consistent regime is close to the value 1/3 which is the predicted value for Ostwald ripening of grain solids in a liquid matrix where grain growth is controlled by long-range diffusion and the typical diffusion is the seperation distance between the solid grains.

IV. CONCLUSION

The Monte Carlo Potts model has been developed to simulate Ostwald ripening by choosing the suitable parameters. A two-phase system composed of solid grains in a liquid matrix was simulated by determining the value of the solid-solid and liquid-solid boundary energies. It is found that the microstructural evolution of the two phases reaches a self-similar regime where power-law relationship was obtained. The value of the grain growth exponent close to the value of n=3 in agreement with the theoretical value of Ostwald ripening.

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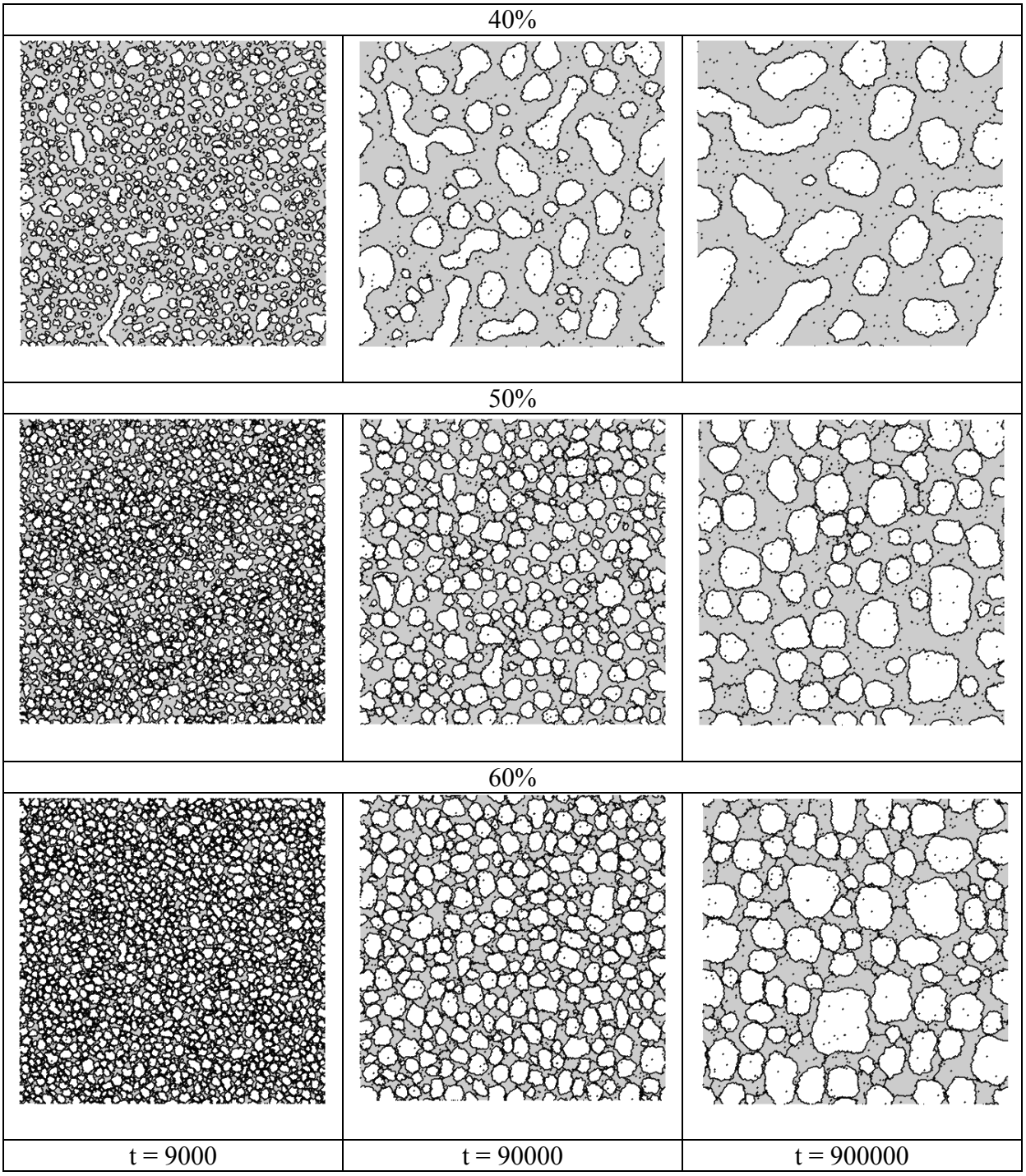

Figure 1. Simulated microstructure of solid-liquid systems with different solid fractions. Solid grains (phase A) are white and liquid (phase B) is gray

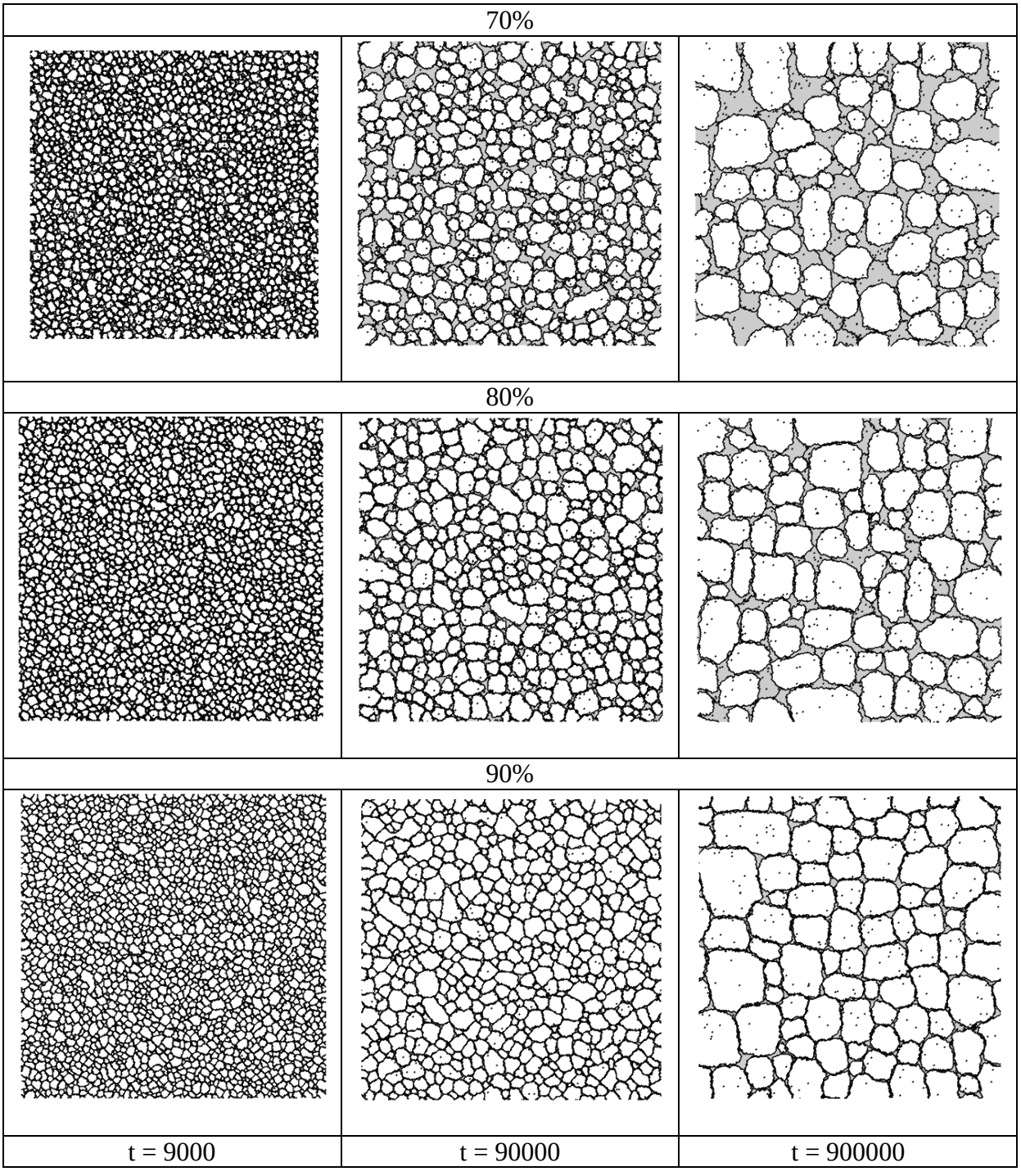

Figure 1. Continued

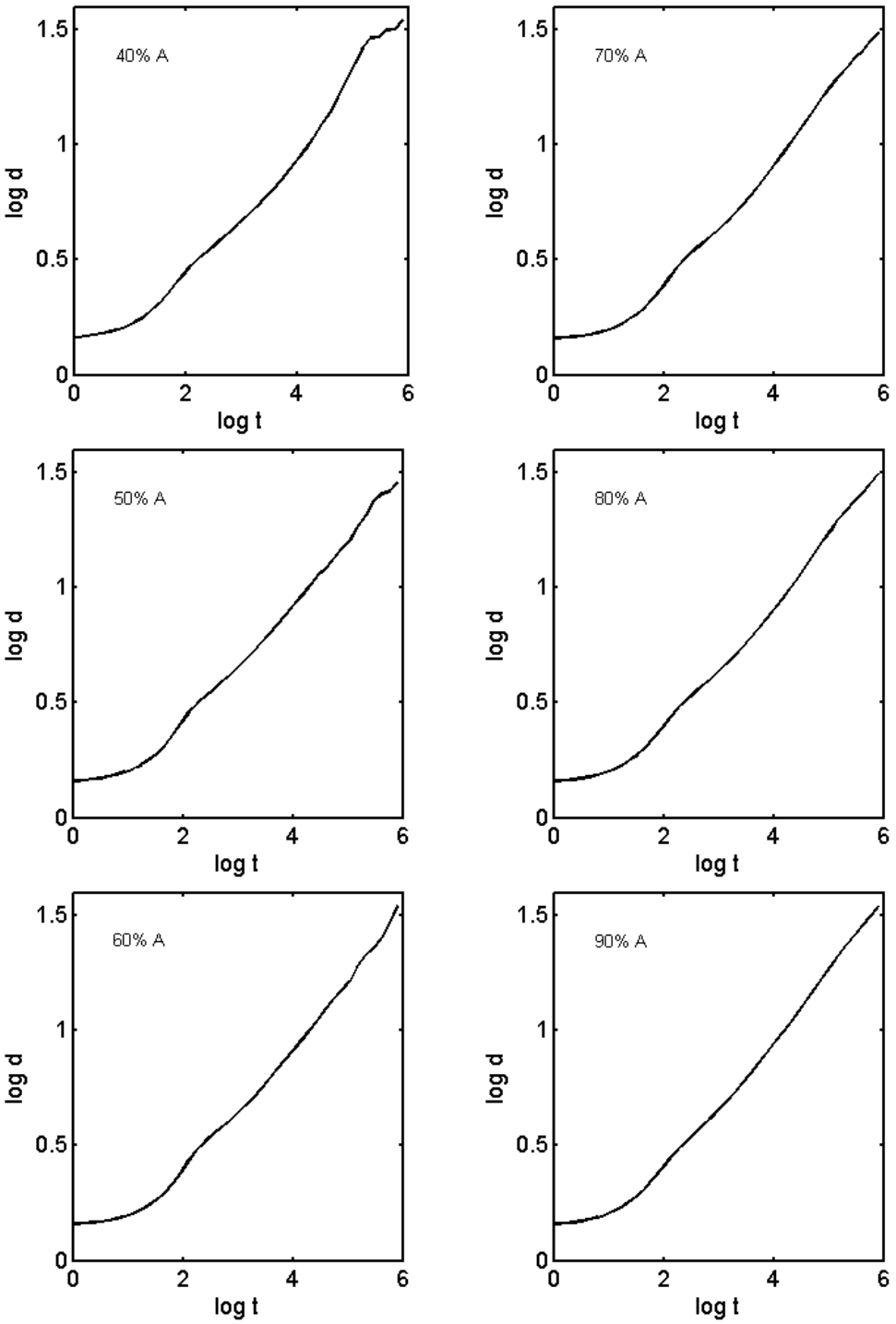

Figure 2. The mean grain size change with time. The volume fractions of solid grains varies from 40% to 90% as indicated.

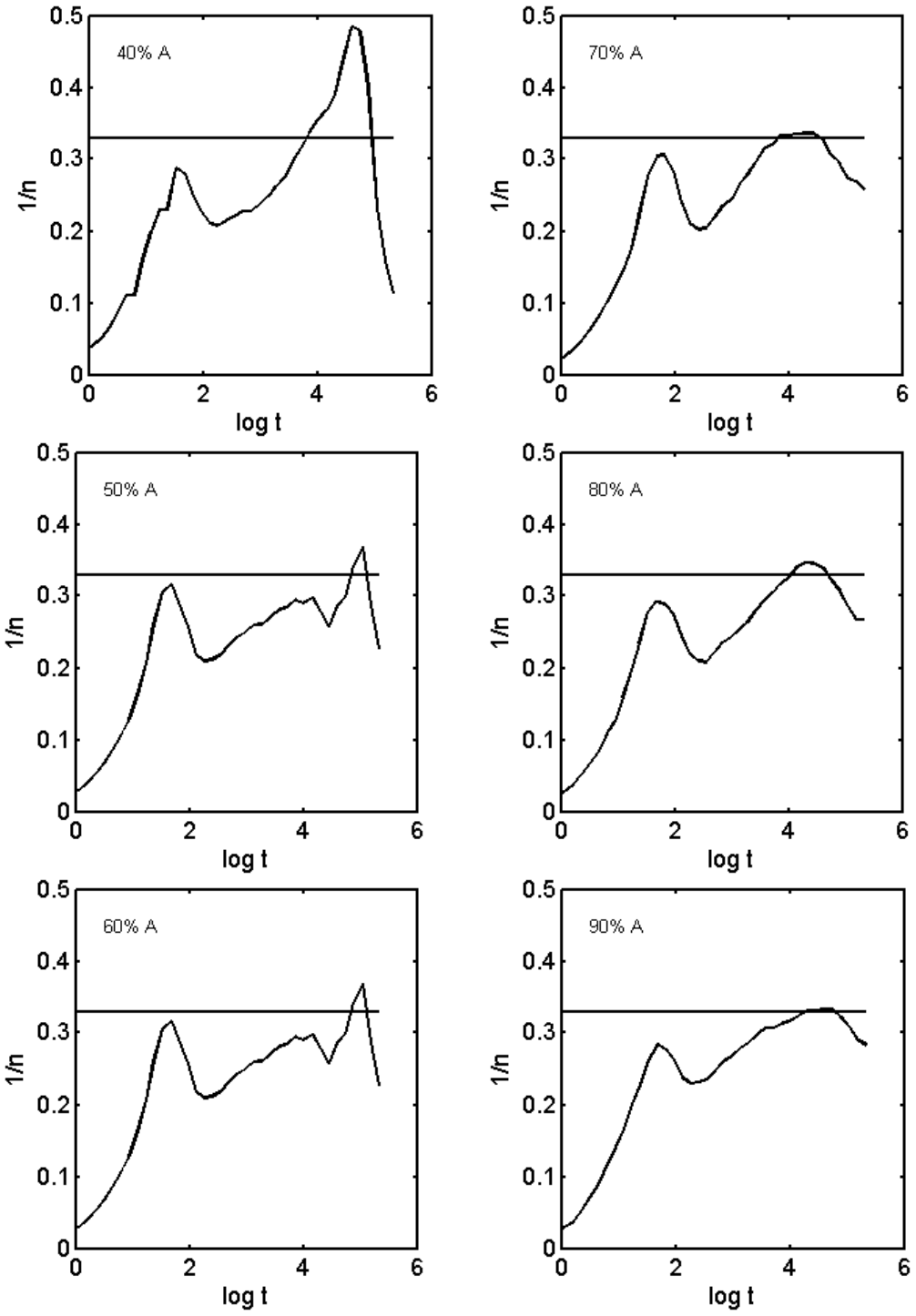

Figure 3. The grain growth exponent as a function of time for various fractions of the solid grains.

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