

Numerical simulation of kinetics of the cobalt gradient change in WC–Co during liquid phase sintering

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ABSTRACT

During liquid phase sintering, WC–Co composite is a solid–liquid two-phase system in which the inter-particle spaces between solid WC grains are filled with liquid Co. When there exists heterogeneity with respect to cobalt content within the composite, liquid Co tends to migrate from regions with higher Co content to other regions with lower Co content, thus causing the redistribution of Co within the WC–Co composite. The kinetics of Co redistribution are of great practical importance for the liquid phase sintering of functionally graded WC–Co composite materials. This paper describes numerical simulations of the kinetic process of Co redistribution, demonstrating how the Co gradient in a WC–Co composite changes with time under various conditions.

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1. Introduction

Functionally graded WC–Co offers a solution to the trade-off between fracture toughness and wear resistance in WC–Co composites by establishing a cobalt gradient or WC grain size differences within the microstructure. Although various methods can be used to make functionally graded WC–Co composites, the liquid phase sintering (LPS) process remains the only economically viable manufacturing method. During liquid phase sintering, a WC–Co composite forms a solid–liquid two-phase mixture in which the inter-particle spaces between WC grains are filled with liquid Co. To manufacture functionally graded WC–Co, a gradient of cobalt content must be established, assuming the WC grain size is uniform throughout the material. The cobalt gradient can form during LPS via a number of means. For example, assuming a sample is composed of two layers of WC–Co with the same Co contents but different WC particle sizes, a Co gradient will form during LPS of this sample [1,2]. Assuming another sample is composed of two layers of WC–Co with the same WC particle sizes but different Co contents, the difference in cobalt content between the two layers, i.e., the initial cobalt gradient, will diminish during LPS. This phenomenon termed liquid phase migration – is similar to but different from the capillary-driven flow which involves the interaction among three-phase (solid, liquid and gas) interfaces. Liquid phase migration, noting that the changes of the Co gradients occur even when there are no pores present in the structure, is driven by the minimization of the interfacial energy in a two-phase mixture. The driving force for liquid phase migration can be expressed in

terms of the so-called liquid migration pressure within a composite material system [3–5]. When the liquid migration pressure is not uniform within a solid–liquid system, liquid will migrate from a location of lower liquid migration pressure to another location of higher liquid migration pressure, until the liquid migration pressure becomes uniform everywhere in the system. For a WC–Co material with no other additives, the liquid migration pressure has been found to depend on two primary factors WC particle size and liquid Co volume fraction, which has been quantitatively established in a recent study by the present authors [6], thus enabling the prediction of equilibrium Co gradient for any given WC particle size distributions.

The cobalt gradient in a fully sintered graded WC–Co composite depends, however, not only on the thermodynamic equilibrium, but also on the kinetic rate of the liquid migration process. For example, assuming a bi-layer WC–Co specimen is made by compacting WC–Co powders with uniform particle size but different cobalt content in each layer, this initial cobalt gradient in the green compact will be completely eliminated during sintering so long as the specimen is held for a sufficiently long time at the sintering temperature. On other hand, if the holding time at the sintering temperature is short, the initial cobalt gradient in the green compact could be maintained in the fully sintered part. In other words, a WC–Co composite with graded cobalt content can be manufactured by controlling the kinetic process of the liquid migration. The challenge is therefore to quantitatively understand the kinetics of liquid phase migration in order to be able to predict the length of time required for obtaining a desired Co gradient rather than the equilibrium Co gradient under various conditions. In another recent study by the present authors [7], the governing equation of liquid phase migration has been derived, and a numerical procedure

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for solving the governing equations was developed. In the present study, the governing equations and the numerical procedures were applied to quantitatively study and simulate the effects of material as well as process variables on the kinetic process and the time required to obtain any specified Co gradient in the material. These factors include sample thickness, WC particle size, and initial Co gradient. The results are expected to be valuable for design and manufacture of functionally graded WC–Co composite materials.

2. Governing equation and numerical method

2.1. Governing equation

As mentioned earlier, in order to quantitatively describe the liquid migration phenomenon, the governing equation that describes the migration process has been established [7], as shown in the following equation:

$$A_0(1-u_0)^{2/3} \frac{\partial(1-u)^{-2/3}}{\partial t} = -\frac{2}{3} \left[\frac{\partial}{\partial l} \left(\frac{k_p}{\mu} \left(\frac{1-u_0}{1-u} \right)^{2/3} A_0 \right) \frac{\partial P_m}{\partial l} + \frac{k_p}{\mu} \left(\frac{1-u_0}{1-u} \right)^{2/3} A_0 \frac{\partial^2 P_m}{\partial l^2} \right] \quad (1)$$

where A_0 is the cross-sectional area perpendicular to the liquid phase migration direction at initial time, m^2 ; t is time, s ; u is the liquid volume fraction ($=V_l/(V_l+V_s)$, where V_l and V_s are liquid volume and solid volume, respectively, m^3); u_0 is the liquid volume fraction at initial time; l is the distance in the liquid phase migration direction, m ; k_p is the permeability of the system, m^2 ; μ is the viscosity of liquid, $kg\ m^{-1}\ s^{-1}$ [8]; P_m is the liquid migration pressure, Pa. Since P_m and k_p are both dependent on the liquid volume fraction u , Eq. (1) is an equation with u as the dependent variable, and t and l as the independent variables. The left-hand term in the equation describes the changing rate of the liquid volume fraction at a given position and time due to liquid phase migration, while the right-hand term is the flux of the mass transfer due to the migration of the liquid phase. The flux of mass transfer by the migration of the liquid phase is proportional to the derivative of liquid migration pressure ($\partial P_m/\partial l$) and permeability (k_p) and inversely proportional to the viscosity of liquid phase (μ).

The challenge for establishing the governing equation of liquid phase migration lies in the fact that it is accompanied by the changes in shape and volume of the system. The volume and shape changes result in changes in the length in the direction of the liquid phase migration and the area across which the liquid phase migrates, which in turn leads to a considerably greater complexity for establishing and solving the governing equation. Mathematically, this is a partial differential equation with moving boundary.

For WC–Co system, it has been shown [6,7] that

$$k_p = 4.5 \times 10^{-4} d^2 u^{1.5} \quad (2)$$

and

$$P_m = 2048 \left[\frac{(1/u - 1)^{1/3} - 1.41u}{d^{0.4}} \right] \quad (3)$$

where d is WC particle diameter, m ; k_p is the permeability of the system, m^2 ; P_m is the liquid migration pressure, Pa; and u is the volume fraction of the liquid Co phase.

2.2. Numerical method

The governing equation of the liquid migration, i.e., Eq. (1), is a nonlinear partial differential equation that can only be solved numerically. The finite difference method, specifically the MacCormack method, was used in this study. The MacCormack method is

an explicit two-step (i.e., predictor–corrector) method that has been proved very efficient and thus popular for solving nonlinear partial differential equations [9]. It should be noted that l , the distance from one end of the two-phase system, varies with time in the equation. Its value must be obtained by summing up the values of Δl which are updated for each time step according to the relation:

$$\Delta l = \Delta l_0 \left(\frac{1-u_0}{1-u} \right)^{1/3} \quad (4)$$

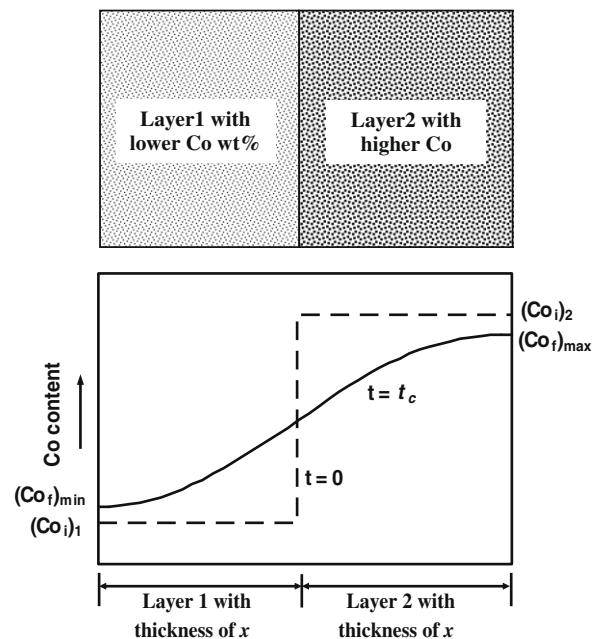
Since the space coordinates of all grid points are tracked (i.e., updated) for each time step, this method can be called as the grid-tracking technique. The finite difference equation of Eq. (1) and the solution algorithm are described in detail elsewhere [7].

3. Simulation results and discussions

In this study, all the numerical simulations were conducted for bi-layers of WC–Co composites with the two layers having the same WC particle size but different initial Co contents. The specimen configuration, the initial Co gradient, and the desired final Co gradient are illustrated in Fig. 1. The magnitude of the Co gradient is defined here as the difference between the maximum and the minimum Co contents within the bi-layer sample. Initially, the Co content was uniform within each layer; thus the initial amplitude of the Co gradient was $\Delta Co_i = (Co_i)_2 - (Co_i)_1$. At the end of the simulated sintering, as shown in Fig. 1, a varying Co content distribution profile developed across the two layers. The maximum and minimum Co contents were located at the two opposite ends of the bi-layer specimen. Therefore, the final amplitude of the Co gradient was $\Delta Co_f = (Co_f)_{max} - (Co_f)_{min}$. The ratio of the final to the initial amplitude of the Co gradient, $\Delta Co_f/\Delta Co_i$, decreased with time. The thickness of each layer was equal to the other. The sintering temperature was assumed to be 1400 °C.

3.1. The evolution of Co gradient with time

Fig. 2 shows the simulated profiles of (a) Co content, (b) permeability k_p and (c) liquid migration pressure P_m after different peri-



Initial amplitude of Co gradient $\Delta Co_i = (Co_i)_2 - (Co_i)_1$ at time $t = 0$;
Desired amplitude of Co gradient $\Delta Co_f = (Co_f)_{max} - (Co_f)_{min}$ at $t = t_c$.

Fig. 1. A schematic of bi-layer WC–Co sample.

ods of sintering time. It shows that the amplitude of the Co gradient decreased as the time increases and that the Co gradient was completely eliminated after six minutes. The specimen thickness in this simulation was 8 mm. This prediction was consistent with our experimental results, which showed that the cobalt gradient was eliminated in 5 min. More thorough validation of the simulation of the bi-layered WC–Co requires more data involving the transition of Co profile with time. These data, however, is not available to date. Therefore, a simulation was conducted by the present authors [7] for a unique case of Co gradient formation in WC–Co, of which the variation of Co profile with time was measured by Lisovsky [3]. In that case, a pre-sintered WC–Co bar was put into contact with liquid Co that had been saturated with WC at 1400 °C. The result showed that liquid Co migrated into the WC–Co bar from the liquid Co pool. Because the liquid Co phases inside the WC–Co and in the liquid Co pool were both saturated with WC at the same temperature, the compositions of liquid Co phases inside the bar and in the pool must be the same and thus no significant diffusion would occur. Therefore, the observed Co gradient in the WC–Co bar, after contact with liquid Co pool, can be attributed only to liquid phase migration. A schematic illustration of the experimental set-up and the Co profiles obtained by both experimental measurement and the simulation are shown in Fig. 3. The good agreement between the simulation and the experiment suggests that the governing equation and the solution algorithm which were also employed in this work are reliable. Therefore, one may expect that the formation of Co gradient in the bi-layered WC–Co specimens, a process that is governed by the same mechanism (liquid phase migration) in the same material system (WC–Co) can be treated using the same governing equation with different initial and boundary conditions.

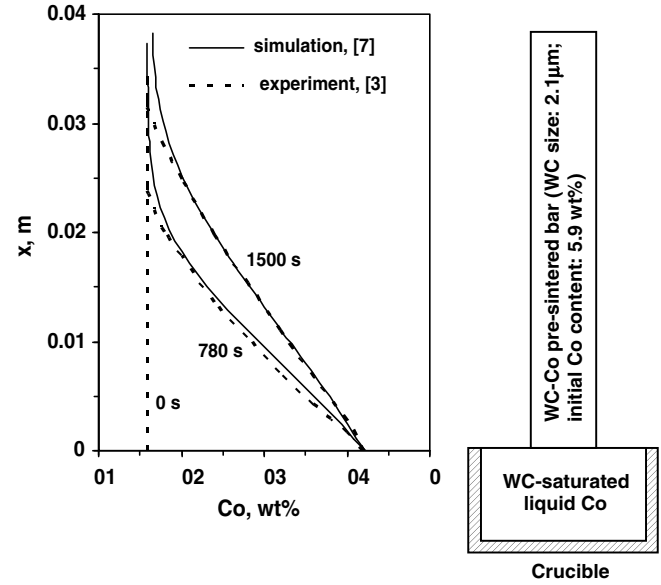


Fig. 3. Comparison between the simulation in this study and the reported experimental results [3] on the Co profile in a pre-sintered WC–Co bar after being put into contact with liquid Co at 1400 °C for different periods of time.

Needless to say, in order to obtain a desired final Co gradient in a fully sintered specimen, an optimal sintering time must be determined for a given initial cobalt gradient. The optimal sintering time at the temperature depends on the initial Co gradient and the desired Co gradient. It depends also on other factors such as WC particle size and sample thickness. In the following simulations, the effects of these factors on a characteristic sintering time t_c , defined as the time required for the amplitude of the cobalt gradient to decrease from an initial value to a specified value, are examined. The simulations were conducted by varying one variable at a time, so that the dependence of the characteristic sintering time on each of the factors is established. Overall, the methodology and the results of this study shed lights on how to control the kinetics of the liquid migration process to achieve the desired graded WC–Co composites.

3.2. Dependence of characteristic sintering time on bi-layer specimen thickness

The dependence of the characteristic sintering time on the thickness of a bi-layer WC–Co specimen was examined via three series of simulations – A, B and C. In each series, only the bi-layer thickness was changed, while other factors, such as WC particle size (d_{WC}), initial Co gradient (ΔCo_i), and the desired final Co gradient (ΔCo_f), were held constant.

The key parameters used in Series A simulations were as follows. The WC particle size was 1 μm ; the initial Co contents in two layers were 6 and 16 wt%, respectively, so that the initial amplitude of Co gradient was $\Delta Co_i = 16 - 6 \text{ wt}\% = 10 \text{ wt}\%$. The final Co contents were 8.5 and 13.5 wt%, respectively, such that the final amplitude of the Co gradient was $\Delta Co_f = 5 \text{ wt}\%$. The ratio of the final-to-initial amplitude of the Co gradient was therefore $\Delta Co_f / \Delta Co_i = 0.5$. These key parameters are also given in Fig. 4. The bi-layer specimen thickness for each simulated case in this series varied from 4 mm to 200 mm. Similarly, the key parameters used in the simulation Series B and C are also given in the same figure.

Fig. 4 depicts the dependence of the characteristic sintering time (t_c) on the specimen thickness (x) in logarithmic scale. The slopes of trend lines for all the three series are found to be 2, suggesting that

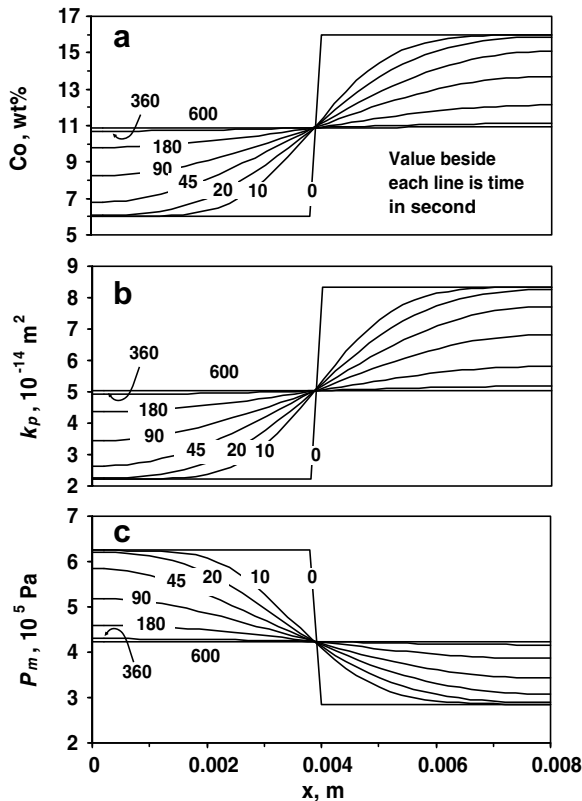


Fig. 2. Predicted distribution of: (a) Co content; (b) permeability k_p and (c) liquid migration pressure P_m in a 8 mm-thick WC–Co bilayer specimen sintered for different time periods. The initial Co contents are 6 wt% and 16 wt%, in the left and the right layers, respectively.

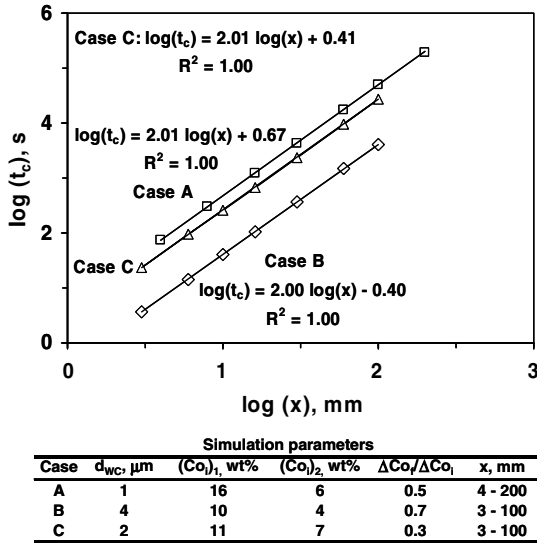


Fig. 4. Required time vs. bilayer thickness.

$$t_c = k_1 x^2 \quad (5)$$

where the value of the ratio (k_1) varies.

The above equation indicates that the required time length dramatically increases with the bi-layer specimen thickness. It is worth noting that the parabolic relationship between time and thickness is similar to the dependence of diffusion distance on time [10]. This similarity is attributed to the fact that the governing equation of liquid phase migration (i.e., Eq. (1)) is mathematically similar to the governing equations of diffusion (i.e., Fick's law).

3.3. The dependence of characteristic sintering time on WC particle size

According to the governing equation of liquid phase migration, Eq. (1), the mass transfer flux due to the liquid phase migration (i.e., the right hand term in the equation) is directly proportional to the product of the permeability k_p and the gradient of migration pressure P_m . Both k_p and P_m are dependent on WC grain size d , so liquid migration flux is dependent on d . Because $k_p \propto d^2$ and

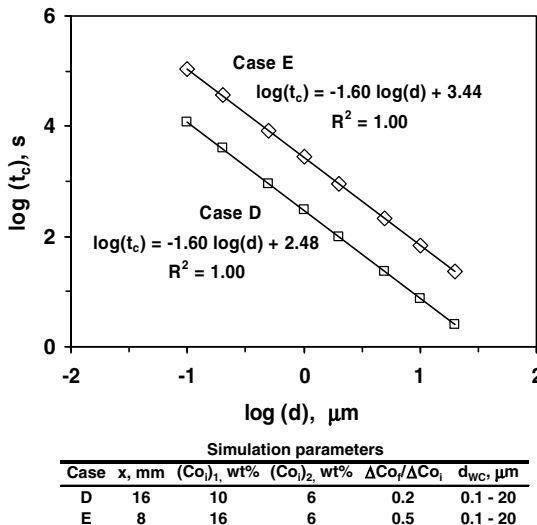


Fig. 5. Required time vs. WC particle size.

$P_m \propto d^{-0.4}$ (see Eqs. (2) and (3)), liquid migration flux is expected to be directly proportional to $d^{1.6} = d^{(2-0.4)}$, suggesting that the dependence of the required length of time for achieving a desired cobalt gradient on the WC particle size should be

$$t_c = k_2 d^{-1.6} \quad (6)$$

The simulation results of Series D and E, as shown in Fig. 5, where only the WC particle size was allowed to change while the other parameter was held constant, confirms the above relation.

3.4. The dependence of characteristic sintering time on initial amplitude of Co gradient

The dependence of the required time for achieving a desired cobalt gradient on the initial amplitude of the Co gradient was examined through two series of simulations (Series F and G). In each series, only the initial amplitude of the Co gradient was varied while the other parameters including the bi-layer thickness (x), WC particle size (d), the average value of the initial Co content ($aveCo_i$), and the ratio between the final to the initial amplitudes of the Co gradients ($\Delta Co_f / \Delta Co_i$), were held constant.

The values of the key parameters used in the Series F and G simulations are given in Fig. 6. The initial Co gradient in the simulations in Series F varied in the range between 2 and 12 wt%, while in Series G the range was between 2 and 16.

Fig. 6 depicts the dependence of the characteristic sintering time (t_c) on the initial amplitude of the Co gradient (ΔCo_i). The flat trend lines for both Series F and G lines indicate that the required time is independent of the initial Co gradient; that is,

$$t_c = k_3 \quad (7)$$

where the value of the ratio (k_3) depends on each simulation series having different key parameters, including WC grain size and sample thickness. Note that this relation was reached when the ratio of the final-to-initial amplitudes of the Co gradient ($\Delta Co_f / \Delta Co_i$) was fixed, suggesting that the time required for the Co gradient to decrease from $\Delta Co = 10$ wt% to $\Delta Co = 5$ wt% should be equal to that from $\Delta Co = 5$ wt% to $\Delta Co = 2.5$ wt%, and that from $\Delta Co = 2.5$ wt% to $\Delta Co = 1.25$ wt%, and so on. It is noted here that this relationship is similar to that of diffusion problems, suggesting that the kinetics of liquid phase migration is similar to that of diffusion processes. The result is useful for designing sintering cycles in practice.

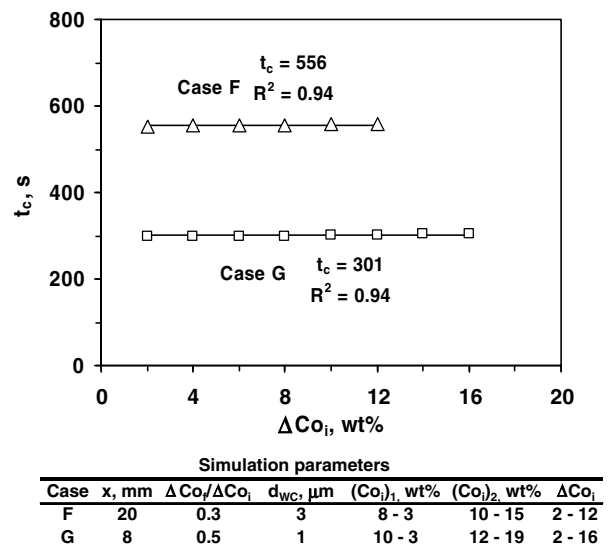


Fig. 6. Required time vs. initial Co gradient.

3.5. The dependence of characteristic sintering time on the ratio of the final-to-initial amplitudes of Co gradients

To further examine the dependence of the required time on the ratio of the final-to-initial amplitudes of Co gradients, $\Delta Co_f/\Delta Co_i$, three series of simulations J, H and L were conducted. In Series H, the bi-layer thickness was 30 mm; WC particle size was 2 μm ; and the initial Co contents for each layer were 7 wt% and 13 wt%, respectively. In Series J, the values used in the simulations for the bi-layer thickness was 8 mm; WC particle size was 1 μm ; and the initial Co contents for each layer were 6 wt% and 16 wt%, respectively. In Series L, the bi-layer thickness was 16 mm; WC particle size was 4 μm ; and the initial Co contents for each layer were 6 wt% and 10 wt%, respectively. During each simulation, the required time (t_c) for the ratio of the final-to-initial amplitudes of the Co gradient ($\Delta Co_f/\Delta Co_i$) to reach 0.9, 0.8, ..., 0.3, 0.2 and 0.1 were recorded.

Based on the plot of t_c vs. $\log_{10}(\Delta Co_f/\Delta Co_i)$ in Fig. 7, the required time length was found to increase with decreasing $\Delta Co_f/\Delta Co_i$, with the relationship as follows:

$$t_c = k_4[0.1 - \log_{10}(\Delta Co_f/\Delta Co_i)] \quad (8)$$

where the value of the ratio (k_4) varied with specific simulation cases.

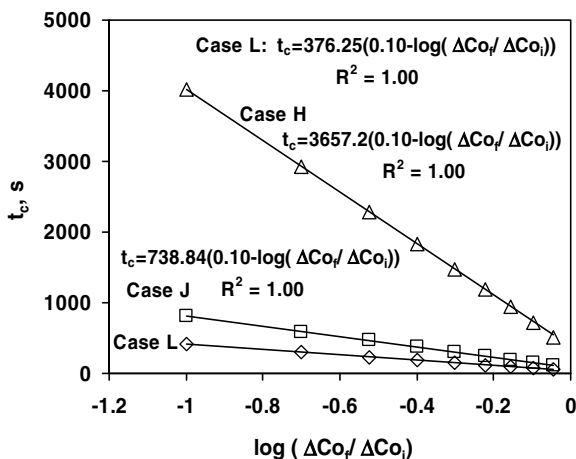
3.6. Dependence of characteristic sintering time on average initial Co content

The dependence of the characteristic sintering time on the average initial Co content ($\text{aveCo}_i = [(Co)_2 + (Co)_1]/2$) was examined through three series of simulations, K, M and N. In these simulations, only the average initial Co content (aveCo_i) was varied, while other factors were held constant.

Based on Fig. 8, which depicts the dependence of the characteristic sintering time (t_c) on the average initial Co content (aveCo_i), the following relationship was obtained:

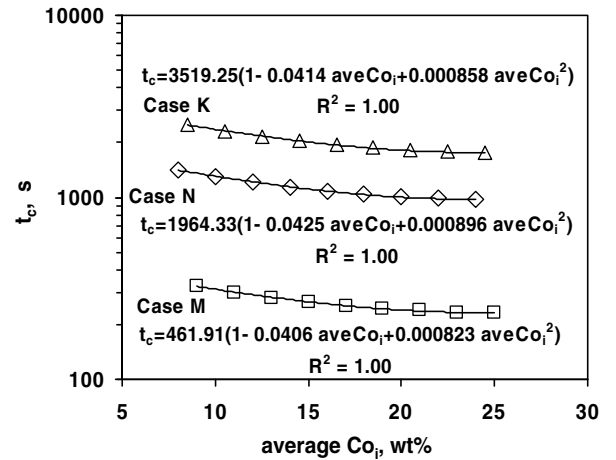
$$t_c = k_5(1 - 4.15 \times 10^{-2} \text{aveCo}_i + 8.59 \times 10^{-4} \text{aveCo}_i^2) \quad (9)$$

where the value of the ratio (k_5) varied with the simulation series. Note that the values of 4.15×10^{-2} and 8.59×10^{-4} are the average numbers of the three regression relations shown in the figure.



Simulation parameters						
Case	$d_{WC}, \mu\text{m}$	$(Co)_1, \text{wt}\%$	$(Co)_2, \text{wt}\%$	x, mm	$\Delta Co_f/\Delta Co_i$	
H	2	13	7	30	0.1 - 0.9	
J	1	16	6	8	0.1 - 0.9	
L	4	10	6	16	0.1 - 0.9	

Fig. 7. Required time vs. ratio of final and initial Co gradient.



Simulation parameters						
Case	$d_{WC}, \mu\text{m}$	x, mm	$\Delta Co_f/\Delta Co_i$	$(Co)_1, \text{wt}\%$	$(Co)_2, \text{wt}\%$	$\text{aveCo}_i, \text{wt}\%$
K	4	40	0.1	5 - 21	12 - 28	8.5 - 24.5
M	1	8	0.5	4 - 20	14 - 30	9 - 25
N	2	20	0.2	6 - 22	10 - 26	8 - 24

Fig. 8. Required time vs. average initial Co content.

According to Fig. 8, the required time decreases with the increase of the average initial Co content, and thus with the average Co content at any time during the entire simulated sintering process. This result suggests that the cobalt migration in a system with a higher Co content and thus a larger volume fraction of liquid would be slightly faster than that in a system with a lower cobalt content. From the standpoint of permeability as in Eq. (2), a higher liquid volume fraction will lead to higher permeability and thus to faster liquid migration; while, from the standpoint of the migration pressure as in Eq. (3), a higher liquid volume fraction will result in a lower liquid migration pressure and thus a slower liquid migration. The observed relation of a slightly faster liquid migration for systems with a higher liquid volume fraction is the combined effects of the above two factors.

3.7. The dependence of characteristic sintering time as a function of all key parameters

Based on all the previous series of simulations, the characteristic sintering time is dependent on the sample thickness (x), the WC particle size (d), the ratio between the desired final Co gradient and initial Co gradient ($\Delta Co_f/\Delta Co_i$) and the average initial Co content (aveCo_i), with the established relations as $t_c \propto x^2$, $t_c \propto d^{-1.6}$, $t_c \propto [0.1 - \log_{10}(\Delta Co_f/\Delta Co_i)]$ and $t_c \propto (1 - 4.15 \times 10^{-2} \text{aveCo}_i + 8.59 \times 10^{-4} \text{aveCo}_i^2)$, respectively. Therefore, it is reasoned that the dependence of the characteristic sintering time can be expressed as a function of all the above key factors as follows:

$$t_c = k_6 x^2 d^{-1.6} [0.1 - \log_{10}(\Delta Co_f/\Delta Co_i)] (1 - 4.15 \times 10^{-2} \text{aveCo}_i + 8.59 \times 10^{-4} \text{aveCo}_i^2) \quad (10)$$

where k_6 is a constant independent on all the above key factors.

Regression of the data from all the simulations in this study leads to a best-fit value for $k_6 = 18.77$. Therefore, Eq. (10) becomes

$$t_c = 18.77 x^2 d^{-1.6} [0.1 - \log_{10}(\Delta Co_f/\Delta Co_i)] (1 - 4.15 \times 10^{-2} \text{aveCo}_i + 8.59 \times 10^{-4} \text{aveCo}_i^2) \quad (11)$$

where t_c is the required time, s; x is sample thickness, mm; d is WC particle size, μm ; ΔCo_f is the final amplitude of the Co gradient, wt%; ΔCo_i is the initial amplitude of the Co gradient, wt%; and aveCo_i is the average initial Co content, wt%.

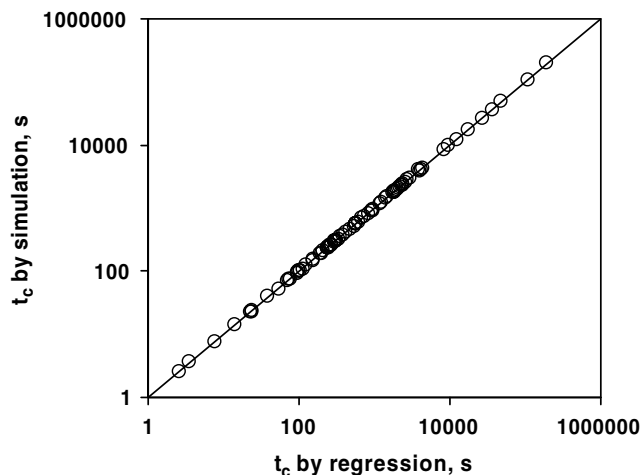


Fig. 9. Required time obtained through direct numerical simulation vs. required time calculated from Eq. (11).

Fig. 9 shows the correlation of the simulated characteristic time from the simulations of the effects of individual parameters with the calculated characteristic sintering time according to Eq. (11) (denoted as t_c by regression). The agreement between the two is satisfactory. It suggests that Eq. (11) can be used to predict the required sintering time for obtaining a desired Co gradient from a pre-designed initial Co gradient under the conditions covered in this study. This is again expected to be valuable for design and manufacture of functionally graded WC–Co composites.

This study focused on the kinetic process of Co redistribution during liquid phase sintering of WC–Co. The methodology and the results of this study, however, are expected to be useful also for understanding the kinetic processes of liquid phase migration in other materials systems. The governing equation of liquid phase migration is not limited by the type of material system. Further studies of the liquid phase migration in other materials systems

are necessary to improve our understanding of this interesting and useful phenomenon.

4. Summary

A numerical simulation has been conducted with the aim of quantitatively understanding the kinetic processes in the sintering of WC–Co composite with a Co gradient via liquid-phase-sintering of bi-layered WC–Co composites. For this purpose, the governing equation of the liquid phase migration has been solved numerically to simulate the change of the Co gradient vs. time under various conditions. The effects of key factors, including sample thickness (x), WC particle size (d), initial Co gradient (ΔCo_i), the desired final Co gradient (ΔCo_f), and the average initial Co content ($aveCo_i$), on the time (t_c) required to obtain the desired Co gradient have been quantitatively established as follows:

$$t_c = 18.77x^2d^{-1.6}[0.1 - \log_{10}(\Delta Co_f/\Delta Co_i)](1 - 4.15 \times 10^{-2}aveCo_i + 8.59 \times 10^{-4}aveCo_i^2)$$

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