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# Sintering driving force of $\text{Al}_2\text{O}_3$ powders at the initial stage of pulse electric current sintering under thermoelastic diffusion

Zhang Long\* and Zheng Heng-wei

## Abstract

**Background:** The theoretical investigation of the rapid sintering mechanism under pulse electric current sintering has no unified understanding. Especially for non-conductive powder, since there is no current flowing directly through the powder materials, the driving force in the neck growth mechanism becomes a key problem and needs to make progress.

**Methods:** The sintering driving force of nonconductive  $\text{Al}_2\text{O}_3$  powders at the initial stage of pulse electric current sintering is investigated under the thermoelastic diffusion coupling transmission with the consideration of non-Fourier and non-Fick effect.

**Results:** The concentration diffusion flux, which is caused by the local concentration gradient, and the thermal diffusion flux act as additional driving forces for the surface curvature driving.

**Conclusions:** Equal-sized particles model reveals that these fluxes exert the dominant influence on sintering driving force for volume and simultaneous surface and volume diffusion mechanisms. In particular, the sintering driving force is remarkably increased at the postperiod of the initial stage of sintering.

**Keywords:** Pulse electric current sintering, Sintering driving force, Generalized thermoelastic diffusion, Thermal diffusion flux

## Background

Pulse electric current sintering (PECS) achieves rapid sintering and densification by enabling a high-intensity pulse current to flow directly through indenters and graphite die. It is achieved by using the Joule heat generated by the large current to rapidly heat up the powder under the coupling of multiphysical fields (thermoelastic diffusion) (Antou et al. 2015; Semenov et al. 2017). But the research of sintering driving force is relatively lacking because of the complex effects of thermal, electrical, and mechanical processes on mass transport (Antou et al. 2015).

Considering the coupling effect of the temperature field, mass diffusion, and deformation field, Nowacki established the thermoelastic diffusion theory, which implied that the propagation speed of temperature and mass diffusion is infinite (Nowacki 1974a; Nowacki

1974b; Nowacki 1974c). Based on the mass diffusion non-Fick's law and the non-Fourier heat conduction law, Sherief (Sherief et al. 2004) posited the generalized thermoelastic diffusion theory, which described the temperature and mass diffusion propagation in limited speed, as well as the coupling effect of temperature, deformation, and mass diffusion in elastic solids. And at present, few literatures have used generalized thermoelastic diffusion theory to analyze the rapid sintering mechanism of PECS.

According to the classical mass diffusion of Fick's law, the concentration gradient is the direct driving force of mass diffusion. Based on the generalized thermoelastic diffusion theory, at the early stage of PECS, the distributions of the temperature and stress fields and of the mass concentration exert an interaction coupling effect at the neck area of particles under transient thermal shock. The influence of temperature gradient and strain are considered in the main concentration diffusion equation. Therefore, the driving forces of mass migration

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during neck sintering consists of three parts: (1) the neck vacancy concentration gradient affected by intrinsic Laplace stress, additional pressure, and thermal stress (Zhang et al. 2016); (2) the temperature gradient caused by the non-uniform temperature distribution at the neck (Olevsky and Froyen 2009); and (3) the concentration gradient resulting from the influence of the temperature and stress fields.

A 3D double equal-sized spherical model based on thermoelastic diffusion interaction is established for nonconductive alumina powder to explore the influence of temperature and concentration field distribution on the sintering driving force at the initial stage of PECS.

## Methods

### Non-Fick's law and generalized thermoelastic diffusion theory

Classical Fick law describes the mass diffusion phenomenon under an assumption that mass disturbances propagate at infinite speeds; such an assumption is reasonable for static or quasistatic mass transfer processes. By conducting theoretical analysis and experimental research, Huai (Huai et al. 2000) and Jiang (Jiang et al. 1996) proved that transient mass transfer follows a wave propagation mechanism that is similar to a heat wave. They defined the process as a diffusion wave, and the corresponding diffusion wave velocity was defined as the mass propagation velocity. Analogous to the transient heat transfer non-Fourier's law, the classical Fick's first law can be revised as the non-Fick's law as follows:

$$J + \tau \frac{\partial J}{\partial t} = -D \nabla C \tag{1}$$

where  $J, D, \nabla C$ , and  $\tau$  denote the diffusion flux, the diffusion coefficient, the concentration gradient, and the diffusion relaxation time, respectively. When the diffusion relaxation time tends to zero, the mass propagation velocity approaches infinity; Eq. (1) is degraded into the classical Fick's first law.

Considering the interaction among elastic effect, heat effect, and mass transfer in the classic thermoelastic theory, Nowacki (Nowacki 1974a; Nowacki 1974b; Nowacki 1974c) proposed the classic thermoelastic diffusion theory. For the extreme heat and mass transfer process, Sherief (Sherief et al. 2004) deduced the basic equations of generalized thermoelastic diffusion (GTD) theory, which considers the non-Fourier effect of heat conduction and the non-Fick effect of mass diffusion.

Equation of motion:

$$\mu u_{i,jj} + (\lambda + \mu) u_{j,ij} - \beta_1 T_{,i} - \beta_2 C_{,i} = \rho' \ddot{u}_i \tag{2}$$

Equation of heat conduction:

$$\begin{aligned} \rho' C_e (\dot{T} + \tau_0 \ddot{T}) + \beta_1 T_0 (\dot{e} + \tau_0 \ddot{e}) \\ + a' T_0 (\dot{C} + \tau_0 \ddot{C}) \\ = k' T_{,ii} \end{aligned} \tag{3}$$

Equation of mass diffusion:

$$D \beta_2 e_{,ii} + D a' T_{,ii} + \dot{C} + \tau \ddot{C} - D b' C_{,ii} = 0 \tag{4}$$

where  $D, C, u_i$ , and  $T$  denote the diffusion coefficient, the concentration, the displacement vector, and the temperature increment related to the reference temperature  $T_0$ , respectively.  $k, \rho'$ , and  $C_e$  represents the thermal conductivity, the density, and the specific heat at constant strain.  $e$  is the components of the strain tensor. The constants  $a'$  and  $b'$  are measures of the thermo-diffusion and diffusive effects, respectively.  $\tau$  is the diffusion relaxation time, and  $\tau_0$  is the thermal relaxation time.  $\alpha_c$  is the linear diffusion expansion coefficient,  $\alpha_t$  is the linear thermal expansion coefficient,  $\lambda$  and  $\mu$  are the Lamé elastic constants and  $\beta_1 = (3\lambda + 2\mu)\alpha_t$ ,  $\beta_2 = (3\lambda + 2\mu)\alpha_c$ . Here,  $\lambda$  and  $\mu$  can be expressed in terms of the elastic modulus  $E$  and Poisson ratio  $\nu$ , written as:

$$\lambda = \frac{Ev}{(1 + \nu)(1 - 2\nu)}, \mu = \frac{E}{2(1 + \nu)} \tag{5}$$

The above field variables may be regarded as the functions of the position vector  $\mathbf{x}$  and time variable  $t$ , and the partial derivative forms can be expressed as:

$$u_{i,jj} = \frac{\partial^2 u_i}{\partial x_j^2}, u_{j,ij} = \frac{\partial^2 u_j}{\partial x_i \partial x_j}, \ddot{u}_i = \frac{\partial^2 u_i}{\partial t^2}$$

where the components of the position vector  $\mathbf{x}$  are  $x_1, x_2, x_3$ .

Equations (2), (3), and (4) are the governing equations of generalized thermoelastic diffusion theory. The equations considered not only the influence of concentration, temperature, and stress-strain relation but also the limitations of temperature and mass propagation velocity. The equations are the coupled equations of temperature, stress, strain, and their concentration combined with the corresponding displacement, temperature, and concentration boundary conditions and initial conditions. These equations can obtain the specific solution of the transient heat and mass transfer problem. The corresponding thermal stress in the PECS process is calculated by:

$$\sigma_T = 2\mu e + \delta_{ij}(\lambda e - \beta_1 T - \beta_2 C) \tag{6}$$

which considers the influence of temperature, strain, and concentration. Here,  $\delta_{ij}$  is Kronecker symbol.

When ignoring the influence of the concentration, only considering the coupling effect of strain and temperature, the Eqs. (2)–(4) degraded into the

generalized thermoelastic theory (GT) as follows (Zhang et al. 2016):

$$\rho \ddot{u}_i = (\lambda + \mu) u_{j,ij} + \mu u_{i,jj} - \beta_1 T_{,i} \tag{7}$$

$$k' T_{,ii} = \rho' C_\epsilon (\dot{T} + \tau_0 \ddot{T}) + \beta_1 T_0 (\dot{\epsilon} + \tau_0 \ddot{\epsilon}) \tag{8}$$

The corresponding thermal stress is:

$$\sigma_T = 2\mu\epsilon + \delta_{ij}(\lambda\epsilon - \beta_1 T) \tag{9}$$

**Calculation model and parameters**

Three kinds of diffusion mechanisms (surface, volume, and simultaneous surface and volume diffusions) are considered. At the initial stage of PECS, surface diffusion causes neck growth but not shrinkage. Thus, the center-to-center distance does not change, as shown in Fig. 1a. Under volume diffusion, the neck grows and the center-to-center distance decreases, resulting in shrinkage and densification. This finding is observed in Fig. 1b, where  $a$  is the neck radius and  $c$  is half of the length of the cord;  $c = 0.74a$  (Wang 1990). In Fig. 1c,  $c = 0.74a_v$ ,

and  $a_v$  is the neck radius produced by the volume diffusion only. The neck curvature radii are approximately  $\rho = a^2/2r$  (surface and simultaneous diffusions) and  $\rho = a^2/4r$  (volume diffusion), where  $r$  is the radii of the particle. The neck growth rate is defined as  $X = a/r$ . This paper considers only the initial stage of PECS, that is,  $X \leq 0.3$ .

The 3D finite element model is established to solve the distributions of the temperature and concentration fields. Step temperature and additional pressure are applied on both ends of the particles,  $x = -r, 3r$ :  $T = \bar{T}^* 1(t)$ ,  $\bar{T} = 1273$  K and  $P = 35$  MPa. The rest of the area of the particles is adiabatic and stress-free. The thermoelastic diffusion parameters (Wang et al. 2010; Young and McPherson 1989; Kumar et al. 2014; Rao and Cutler 1972) are shown in Table 1. Relevant partial differential equations (PDEs) are solved using Comsol Multiphysics 3.5.

**Results and discussion**

**Temperature and concentration field distributions**

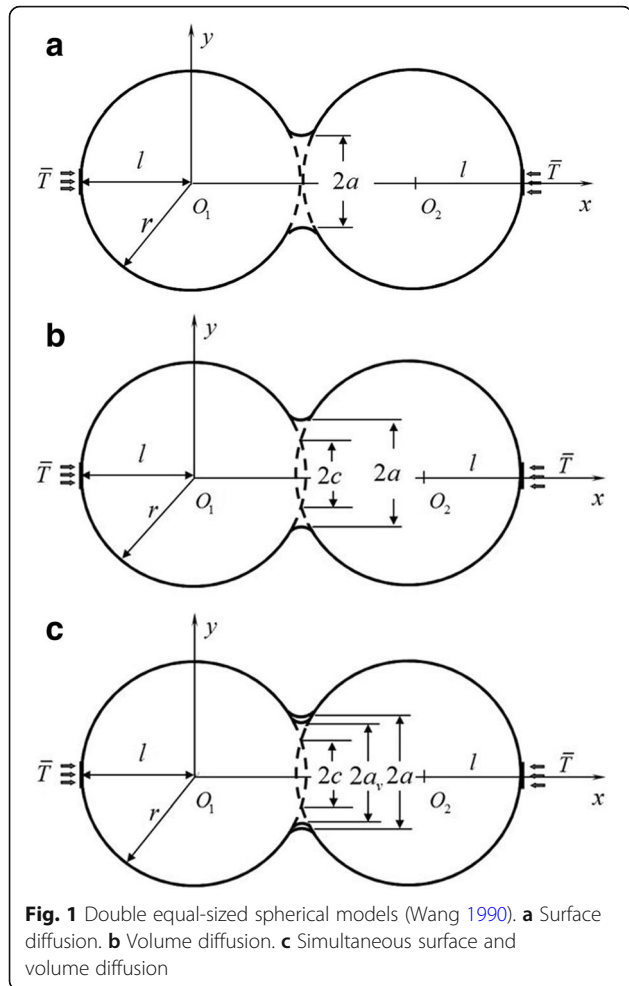
For the volume diffusion mechanism in the generalized thermoelastic diffusion theory that considers the heat conduction non-Fourier effect, the temperature is in the form of wave propagation in the particle system. Firstly, superposition is generated at the edge of the sintering neck, resulting in local high temperature at the edge of the neck as shown in Fig. 2a, and then the heat wave continues to propagate to the center of the sintering neck forming a local high temperature in the center of the neck, and the maximum temperature can be reached to 1573 K, as shown in Fig. 2b.

At the same time, the changes in temperature and stress cause a change in concentration in the neck area. The thermal wave at the neck forms the local high temperature and produces a greater larger thermal stress, thereby causing significant changes to the mass concentration of neck sintering, driving neck material migration, and ultimately providing an additional driving force for neck growth. Affected by temperature and stress, the concentration of the edge of the neck initially increases at the neck edge (Fig. 3a) and then forms the high local concentration and the concentration gradient at the center of the neck (Fig. 3b). The high concentration gradient drives the neck material migration to the edge of the neck.

**Diffusion flux under volume diffusion**

For the condition of a high heat rate under pressure, the vacancy concentration difference  $\Delta C$  of PECS in consideration of the intrinsic Laplace stress, the thermal stress, and the additional applied pressure is proposed as follows (Zhang et al. 2016):

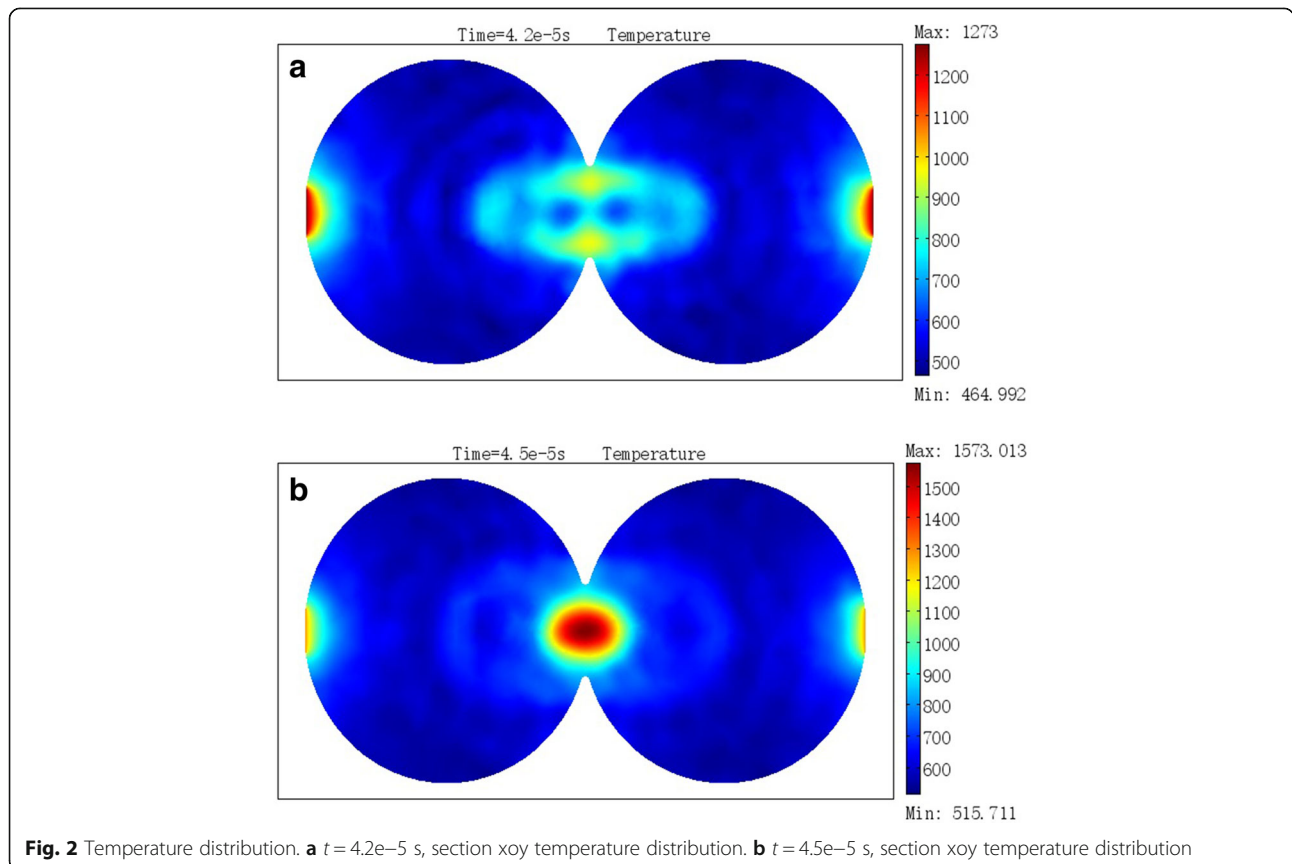
Volume diffusion:



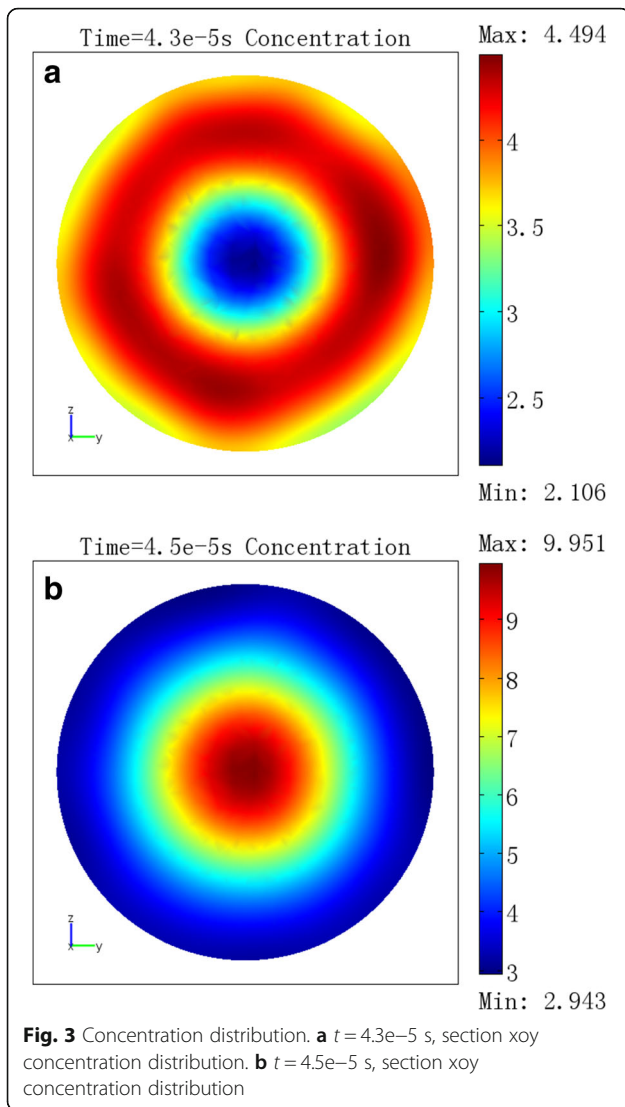
**Fig. 1** Double equal-sized spherical models (Wang 1990). **a** Surface diffusion. **b** Volume diffusion. **c** Simultaneous surface and volume diffusion

**Table 1** The thermoelastic diffusion parameters of  $Al_2O_3$

Name	Symbol	Value	Unit
Reference temperature	$T_0$	293	K
Density	$\rho'$	3900	$kg \cdot m^{-3}$
Specific heat	$C_\epsilon$	900	$J \cdot kg^{-1} \cdot K^{-1}$
Thermal conductivity	$k'$	27	$W \cdot m^{-1} \cdot K^{-1}$
Thermal expansion coefficient	$\alpha_t$	$8.0 \times 10^{-6}$	$K^{-1}$
Elastic modulus	$E$	380	GPa
Poisson ratio	$\nu$	0.22	
Thermal relaxation time	$\tau_0$	0.2	s
Diffusion expansion coefficient	$\alpha_c$	$2.34 \times 10^{-4}$	$m^3 \cdot kg$
Diffusion coefficient	$D$	$0.63 \times 10^{-8}$	$kg \cdot s \cdot m^{-3}$
Measures of the thermodiffusion effects	$a'$	$2.32 \times 10^4$	$m^2 \cdot s^{-2} \cdot K^{-1}$
Measures of the diffusive effects	$b'$	$3.061 \times 10^6$	$m^5 \cdot kg^{-1} \cdot s^{-2}$
Diffusion relaxation time	$\tau$	0.02	s
Volume of vacancy	$\Omega$	$2.7 \times 10^{-29}$	$m^3$
Boltzman constant	$k$	$1.3806505 \times 10^{-23}$	
Enthalpy of vacancy motion	$H_m$	3.8	eV



**Fig. 2** Temperature distribution. **a**  $t = 4.2e-5$  s, section xoy temperature distribution. **b**  $t = 4.5e-5$  s, section xoy temperature distribution



$$\Delta C = (C_0 \Omega / a^2 k T) (4 \gamma r + 4 P r^2 / \pi + a^2 \Delta \sigma_T) \quad (10)$$

Simultaneous diffusion:

$$\Delta C = (C_0 \Omega / a^2 k T) (2 \gamma r + 4 P r^2 / \pi + a^2 \Delta \sigma_T) \quad (11)$$

where  $\gamma$  and  $P$  denote the surface energy and the additional applied pressure, respectively;  $\Omega$  is the volume of vacancy,  $C_0$  presents equilibrium concentration of the vacancy of stress-free zone, and  $k$  is Boltzman constant.  $\sigma_T$  is the thermal stress caused described in Eq. (6). The corresponding vacancy diffusion flux is

$$J_{\text{curv-t}} = -D \frac{\Delta C}{\rho} \quad (12)$$

In hot-pressure (HP) sintering, that is,  $\sigma_T = 0$ , the corresponding vacancy diffusion flux is described as  $J_{\text{curv}}$ .

The thermal diffusion flux driven by the temperature gradient is expressed as (Olevsky and Froyen 2009):

$$J_{\text{ther}} = -\frac{D C H_m}{k T} \frac{\nabla T}{T} \quad (13)$$

Here  $H_m$  is the enthalpy of vacancy motion, and  $\nabla T$  is the temperature gradient.

Affected by temperature and stress, the concentration diffusion flux expression is

$$J_{\text{conc}} = -D \nabla C \quad (14)$$

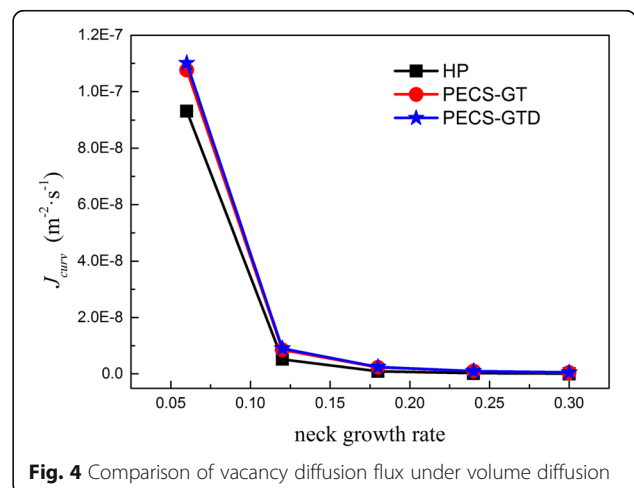
Figure 4 compares the vacancy diffusion flux of the HP, the generalized thermoelastic (GT), and the GTD theory under volume diffusion mechanism. Considering the effect of thermal stress, the vacancy diffusion flux calculated by GT and GTD theories greatly increased, and the values of GTD is little higher because GT theory only considers the influences of strain and temperature.

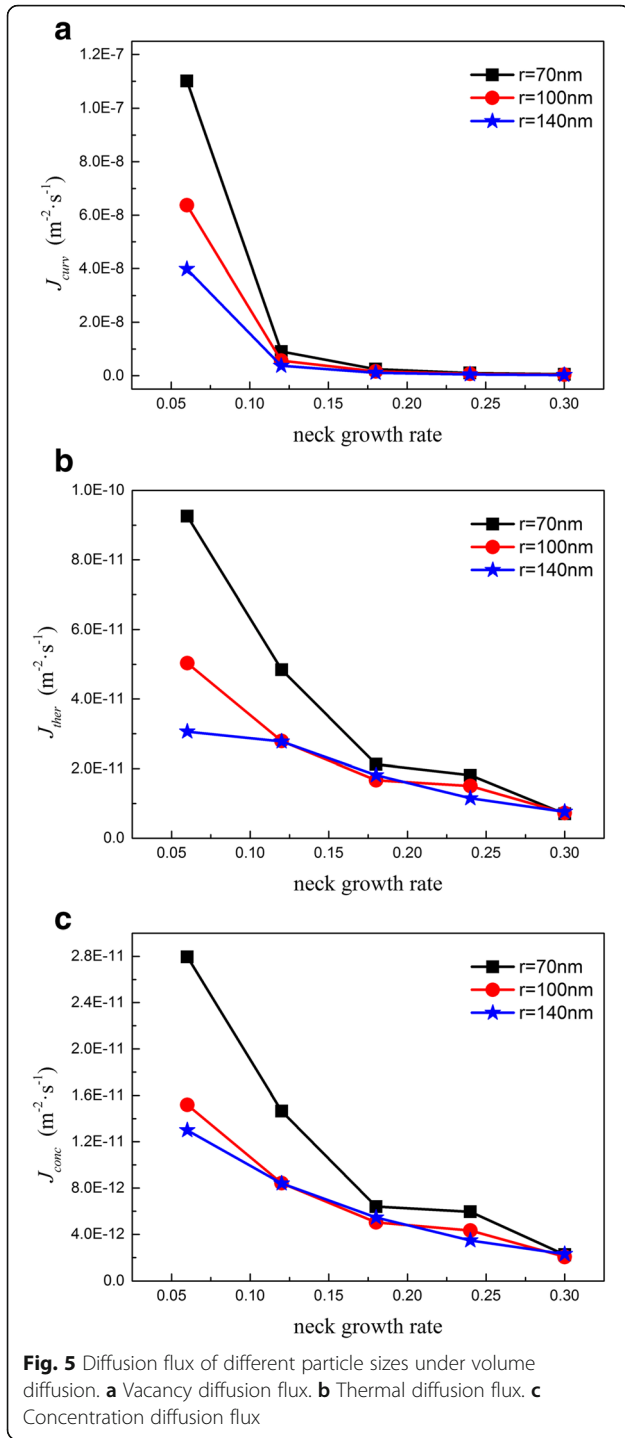
Particle size has a significant effect on the diffusion flux. As the particle size increases, the vacancy of the diffusion flux, the thermal diffusion, and the concentration of the diffusion fluxes were significantly reduced, especially at the initial stage, as shown in Fig. 5. Numerical results show that large particles have small sintering driving forces; thus, the fine powder accelerates the sintering process in a short time. This finding is consistent with actual sintering practices (Hungria et al. 2009; Lu et al. 2012).

By introducing the particle radius,  $r$ , the ratio of the thermal diffusion flux to the conventional HP curvature-driven flux is given by (Johnson 1990).

$$\frac{J_{\text{ther}}}{r^2 J_{\text{curv}}} = \frac{\nabla T H_m \rho}{T 2 \Omega r^2} / \left[ \left( \frac{\gamma}{\rho} - \frac{\gamma}{a} \right) + \frac{4 P r^2}{\pi a^2} \right] \quad (15)$$

The ratio of the concentration diffusion flux to the conventional HP curvature-driven flux can be written as





$$\frac{J_{conc}}{r^2 J_{curv}} = \frac{\nabla C \rho}{r^2 \Delta C} \tag{16}$$

According to Eqs. (10) and (11), the ratio of PECS vacancy diffusion flux to the conventional HP curvature-driven flux is.

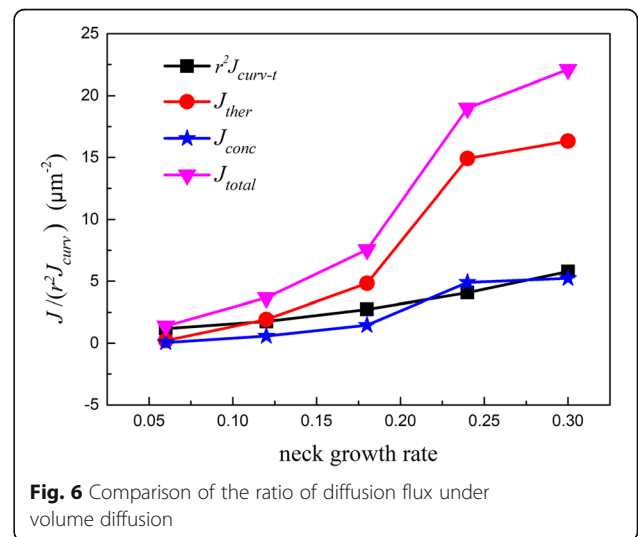
Volume diffusion:

$$\frac{J_{curv-t}}{J_{curv}} = \frac{4r\gamma + 4pr^2/\pi + a^2\Delta\sigma_T}{4r\gamma + 4pr^2/\pi} \tag{17}$$

Simultaneous diffusion:

$$\frac{J_{curv-t}}{J_{curv}} = \frac{2r\gamma + 4pr^2/\pi + a^2\Delta\sigma_T}{2r\gamma + 4pr^2/\pi} \tag{18}$$

The ratios of the PECS vacancy diffusion flux, the thermal diffusion flux, the concentration diffusion flux, and the total of three diffusion fluxes to the conventional HP curvature-driven flux are shown in Fig. 6. At the initial stage of PECS, along with the sintering process, the ratio of thermal diffusion flux to the conventional curvature-driven flux has an obvious increasing trend because the contact area between particles is smaller, the intrinsic Laplace stress is infinite, and the free-surface curvature-driven diffusion is dominant. Then, the contact area increases with the neck growth and the intrinsic Laplace stress decreases, causing the ratio of the diffusion flux to increase. The neck material migration is mainly driven by thermal diffusion. The concentration diffusion flux caused by the change of temperature and stress, retain a relatively higher value, that is, about three to five times that of the HP processes. Relative to the PECS vacancy and the concentration diffusion fluxes, the influence of thermal diffusion flux formed from the neck non-uniform temperature distribution on the sintering driving force is dominated especially in the late initial stage of PECS that remarkably increases the sintering driving force. Therefore, for the double equal-sized spherical model under volume diffusion mechanism, the neck growth-driven force is mainly provided by the temperature gradient.



**Diffusion flux under simultaneous diffusion**

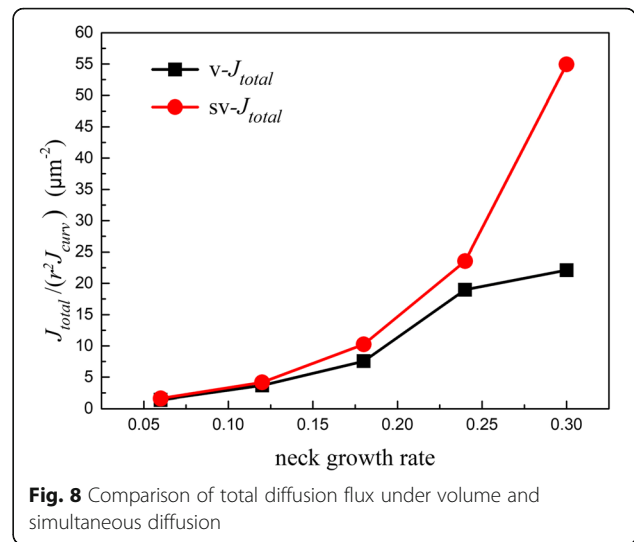
Similar to the volume diffusion mechanism, thermal wave produces superposition at the neck under a simultaneous diffusion mechanism forms the local high temperature. The neck mass concentration change, caused by temperature changes, forms high concentration gradient, promoting the mass migration of the neck center to the neck surface.

Three kinds of the ratio calculated by Eqs. (15), (16), and (18) under simultaneous diffusion are shown in Fig. 7. The thermal diffusion flux accounted for nearly half of the total diffusion flux, meaning that thermal diffusion is the main driving force of the neck mass migration and that the concentration diffusion flux cannot be ignored. Thermal stress, thermal diffusion, and concentration gradient greatly improved the driving force of the neck mass migration, which explains the reason why PECS possesses higher efficiency than the traditional hot-press sintering.

The ratio of total diffusion flux under volume ( $v - J_{total}$ ) and simultaneous diffusions ( $sv - J_{total}$ ) is shown in Fig. 8. With the increase of neck growth, the ratio of total flux increases significantly; in the late initial stage of PECS, the values of simultaneous diffusion is higher than volume diffusion. In fact, the diffusion mechanism of the PECS process is a series of transient process under the action of varieties of diffusion mechanism. The change tendency of simultaneous diffusion explains that the PECS is finished in a short time.

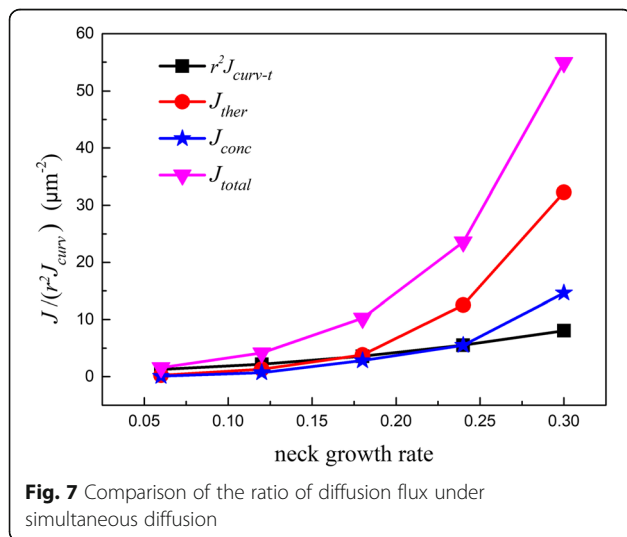
**Conclusions**

The sintering driving force at the initial stage of PECS is investigated by introducing the generalized thermoelastic diffusion theory and considering the coupling effect of strain, temperature, and concentration. The following conclusions are drawn:



**Fig. 8** Comparison of total diffusion flux under volume and simultaneous diffusion

1. The thermal wave in the neck produces a superposition, thus forming the local high temperature. Temperature change causes the neck mass concentration to change, thereby forming the local concentration gradient and providing additional driving force for neck growth. The thermal stress, which includes the influence of strain, temperature, and concentration, improves the vacancy diffusion flux in the neck.
2. For double equal-sized spherical model under volume diffusion and simultaneous diffusion, the thermal diffusion flux driven by the high-temperature gradient formed from the neck non-uniform temperature distribution is the dominant sintering driving force, especially at the late initial stage of PECS. This flux remarkably increases the driving force, promoting neck mass migration. At the same time, the total ratio of the diffusion flux of simultaneous diffusion is greater than the volume.



**Fig. 7** Comparison of the ratio of diffusion flux under simultaneous diffusion

**Abbreviations**

GT: Thermoelastic theory; GTD: Generalized thermoelastic diffusion; HP: Hot pressure; PDEs: Partial differential equations; PECS: Pulse electric current sintering

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### Authors' contributions

ZL and ZH conceived and designed the study. ZL wrote the paper. ZH reviewed and edited the manuscript. Both authors read and approved the final manuscript.

### Competing interests

Both authors declare that they have no competing interests.

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