Modelling of powder consolidation using electro heating assisted by mechanical loading

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Abstract. The model of the process of reactive sintering assisted by mechanical loading is suggested. The conjugate heat exchange of powder mixture is taken into account. The powder mixture motion is described as viscous liquid with effective viscosity. Mechanical sub problem is one dimensional because friction near the wall is assumed negligible small. Conjugate thermal conductivity problem includes thermal conduction equations for various materials (reactive mixture and walls of the camber. Heat release is possible due to external electrical heating, viscous dissipation and chemical reactions. Kinetical equations correspond to detailed reaction scheme. The problem is solved numerically with special algorithm. As a result the composition of the mixture is obtained for different time moments. The final composition is not uniform.

1. Introduction
There are a lot of technologies of new materials synthesis assisted by mechanical loading together with the heating by different methods. Process of powder consolidation using electricity as the main energy component is widely known and is used in various technologies. Depending on material types, experimental conditions, geometry, preliminary preparation, the way of loading, ones distinguish hot press sintering (HP); hot isostatic pressing (HIP); spark plasma sintering (SPS), and etc [1]. As a result, we come to composite materials with different structure, composition and properties. The most accepted mechanism of the pulsed electric current sintering includes the heating in the gap between neighbouring due to the micro-spark discharge, the heating by joule heat due to electrical current flowing from particle to particle and mass exchange between particles initiated by temperature gradient, electro migration and inelastic strains. The possibility of liquid phase formation in the contact between particles was experimentally confirmed for example in [2]. Numerous physical and mathematical models exist in this field allowing describing the various details of the sintering. Mathematical modelling allows to understand physical phenomena accompanying the new materials production and to give the preliminary prognosis for the choosing of technological parameters [3].

The first major publication on computer simulation of sintering appears in 1965 [4], where finite-difference method has been applied for the sintering of spheres. Nowadays, various approaches are used for the modelling of sintering assisted by pressure and electric field: Monte Carlo, finite difference, discrete element, finite element, neural network etc. Even so, when commercial software’s are available, there are a lot of problems in the modelling [3].
Based on physical lows and continuum mechanics, many authors describe the temperature change in complex fields, residual stresses, density, porosity viscosity and mechanical properties evolution, the junction formation between individual particles etc. [5-14].

Theory and technologies of sintering are described in [9, 12, 15-17 ] Special attention was paid to the generation and evolution of new ideas, elements of the physical theory of sintering during different time periods. The key role in these theories belongs to theoretical physicists, Frenkel, Herring, and Lifshits.

One of the most popular sintering theory [9, 12, 14] includes the relations between stress tensor’s components $\sigma_{ij}$ and strain rate tensor’s component $\varepsilon_{ij}$

$$\sigma_{ij} = 2\eta_0 \left[ \varphi \varepsilon_{ij} + \left( \psi - \frac{1}{3} \varphi \right) \delta_{ij} \right] + p_L \delta_{ij} \tag{1}$$

where $\eta_0$ is the shear viscosity of a fully dense material, $\varphi$ and $\psi$ are the normalized shear and bulk viscosities, $\delta_{ij}$ is the Kronecker symbol ($\delta_{ij} = 1$ if $i = j$ and $\delta_{ij} = 0$ if $i \neq j$), $\varepsilon = \varepsilon_{kk} = \varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33}$. Effective sintering stress $p_L$ is the product of the local sintering stress $p_{LO} = 3\alpha/G$ ($\alpha$ is the surface tension, $G$ is the product of the local sintering stress) and of the normalized effective sintering stress $p_L = (1-\theta)^2/2$.

The normalized shear and bulk viscosities $\varphi$ and $\psi$ are defined as:

$$\varphi = (1-\theta)^2; \quad \psi = \frac{2}{3} \frac{(1-\theta)^3}{\theta}.$$  

Porosity changes in accordance of kinetic equations depending on problem under study.

The finite element simulation [3 and 18] predicts several phenomena, including grain growth, densification, and distortion based on constitutive equations applicable to a linear viscous compressive material. Temperature is given as uniform. In these papers, the relations between strains and stresses are taken in form

$$\varepsilon_{ij} = \frac{1}{2\eta} \sigma_{ij} + \frac{1}{3K} \left( \frac{\sigma_{kk}}{3} - \sigma_s \right) \delta_{ij} \tag{2}$$

where $\sigma_{ij}$ - deviatoric part of stress tensor, $\sigma_s$ - sintering stress, $\eta$ and $K$ are effective shear and bulk viscosity. The values $\sigma_s, \eta, K$ depend on relative density, surface energy corresponding to some laws depending on sintering stage.

Note, that equations (1) and (2) are practically equivalent. Immediately from (1), we can obtain

$$\varepsilon_{ij} = \frac{1}{2\eta_0 \varphi} \left[ \sigma_{ij} - \frac{\sigma_{kk}}{3} \right] + \frac{1}{3\psi} \frac{1}{2\eta_0} \left( \frac{\sigma_{kk}}{3} - p_L \right) \delta_{ij}$$

Hence, $\eta = \eta_0 \varphi; \ K = 2\eta_0 \psi$ and $\sigma_s = p_L$.

One can suggest the generalization for (1) and (2), based on known analogy between viscous and elastic stresses [ ] and taking into account

A majority of the existing papers connecting with the sintering is devoted to the compaction stage. Only few attempts have been made to develop the approach combining the compaction and sintering stages. For example, in the papers [19-21], sintering process is modeled within an idealized Representative Volume Element. The intrinsic deformation of both the solid phase and the melt phase are taken into consideration, whereby elastic deformation is ignored. The macroscopic properties of sintered material are found via computational homogenization of the representative element.

However, not uniform temperature field connecting with the conjugate heat exchange in real technological conditions, and chemical conversions which lead to composition and properties change
together with porosity evolution (for example, for intermetallic systems [22,23]) are not taken into account in macroscopic continual models.

In this paper we study the stress-assisted sintering process for the case of intermetallic composite formation in the conditions of conjugate heat exchange.

2. Problem formulation

Problem formulation corresponds to Fig.1.

![Figure 1. Illustration to the problem formulation](image)

and is based on following assumptions: the wall friction absents; the flow is one-dimensional; the powder mixture flow is similar to viscous fluid; the thermal problem is two dimensional due to conjugate heat exchange; the coordinate system is cylindrical; the heating is carried out by Joule heat; the porosity changes in accordance with kinetical equation; the chemical heat release is possible and corresponds to the simplified reaction scheme:

\[ Ti + Ni \rightarrow T_{i}Ni \]
\[ 2Ti + Ni \rightarrow T_{i}2Ni \]
\[ Ti + 3Ni \rightarrow T_{i}Ni_{3} \]
\[ T_{i}Ni + 2Ni \rightarrow T_{i}Ni_{3} \]

This gives the features of the model in comparison with [24, 25].

The sub problem

\[ \sigma = \left( \zeta + \frac{4}{3} \mu_{V} \right) \frac{dV}{dz} \quad (3) \]
\[ \rho \rho_{s} \left[ \frac{\partial V}{\partial t} + V \frac{\partial V}{\partial z} \right] = \frac{\partial \sigma}{\partial z} \quad (4) \]
\[ z = \xi(t) : -p(t) = \sigma = \left( \zeta + \frac{4}{3} \mu_{V} \right) \frac{dV}{dz} \quad (5) \]
\[ z = \frac{h_{2}}{2} : V = 0; \; \sigma = 0, V = 0, t = 0 \quad (6) \]

describes the flow in the region with moving boundaries. Here \( p(t) \) is the given law of external load change; \( \rho \) - is relative density, connecting with the porosity, \( \rho = 1 - \theta \); \( \sigma = \sigma_{zz} \) is stress tensor component in the loading direction; \( V \) is the component of velocity vector in this direction.
Porosity changes in accordance with the equation [17]

$$\frac{d\theta}{dr} = k(1-\theta) \frac{\partial V}{\partial z}. \tag{7}$$

In general case, coefficient $k$ can depend on pressure, temperature and other parameters

$$k = k(P,T,Y_k),$$

where $P = \frac{1}{3} \sigma_{ik}$, and $Y_k$ are the species concentrations ($k = 1,2,3,4,5$ for Ti, Ni, TiNi, Ti$_2$Ni, TiNi$_3$).

In the powder medium, the processes of heat conductivity, structure formation and chemical transformations take place. Due to heat losses to the walls from reacting mixture, thermal processes are two-dimensional. Thermal conductivity equation for the reacting mixture contains the various volume heat sources:

$$c_2 \rho_2 \partial_t T_2 + V \frac{\partial T_2}{\partial z} = \frac{\partial}{\partial z} \left( \lambda_2 \frac{\partial T_2}{\partial z} \right) + \frac{1}{r} \frac{\partial}{\partial r} \left( \lambda_2 r \frac{\partial T_2}{\partial r} \right) + W_{ch} + W_H + \sigma \frac{\partial V}{\partial z}. \tag{8}$$

Here term $W_H$ describe the electrical heating

$$W_H = R_e I^2(t),$$

where $I(t)$ is the current, $R_e$ is effective electrical resistance depending on temperature and medium structure. This claims a special investigation. In this paper, we believe that dependence of resistance on time is known from experiment [26]. The last term in (8) corresponds to viscous dissipation. Stress and velocity fields follow from mechanical sub problem. Chemical heat release includes the heat of all reactions.

Corresponding to the reaction scheme, we have five kinetical equations:

$$\frac{dY_k}{dt} = \sum_{i=1}^{4} \omega_i v_{ki}, \quad k = 1,2,3,4,5 \tag{9}$$

where

$$\omega_i = z_i \phi_i (Y_j) \exp \left( - \frac{E_k}{RT} \right) \equiv \omega_i (z,r,t).$$

It is enough for numerical solution only four equations, because $Y_1 + Y_2 + Y_3 + Y_4 + Y_5 = 1$.

For four reactions we have

$$\omega_1 = Y_1 Y_2, \quad \omega_2 = Y_1^2 Y_2, \quad \omega_3 = Y_1 Y_3^2, \quad \omega_4 = Y_3 Y_2^2.$$

The values $z_i$ can take into account the reaction retardation by summary product, similarly to [27, 28].

Hence, for chemical heat release we shall find

$$W_{ch} = \sum_{i=1}^{4} Q_i \omega_i, \quad Q_i = - \sum_{k=1}^{5} h_k v_{ki}.$$

Parameters for kinetical equations, heats for the reactions were found based on thermodynamical data similarly to [29].

Usual thermal conductivity equations take a place in the areas 1 and 3 (Fig.1):

$$c_k \rho_k \frac{\partial T_k}{\partial t} = \frac{\partial}{\partial z} \left( \lambda_k \frac{\partial T_k}{\partial z} \right) + \frac{1}{r} \frac{\partial}{\partial r} \left( \lambda_k r \frac{\partial T_k}{\partial r} \right), \quad k = 1,3 \tag{10}.$$
Boundary conditions between materials also correspond to Fig.1. We believe that ideal thermal contact exists in interfaces «1-2»; «1-3» and «2-3». The symmetry conditions are assumed in the axis \( r = 0 \) and in the plane \( z = h_2 / 2 \) (cross-section A-A).

Heat exchange with environment is carried out by radiation, for example

\[
    z = -h_1 + \xi(t); \quad \lambda_1 \frac{\partial T_1}{\partial \xi} = -\sigma_0 c_1 \left( T_1^4 - T_0^4 \right); \\
    r = R_3; \quad \lambda_3 \frac{\partial T_3}{\partial \xi} = -\sigma_0 c_3 \left( T_3^4 - T_0^4 \right).
\]

At the initial time moment we have:

\[
    T = T_0, \quad \sigma = 0, \quad V = 0, \quad \eta = \eta_0(z), \quad \xi = 0; \\
    Y_1 = Y_{10}, \quad Y_2 = Y_{20}, \quad Y_3 = 0; \quad Y_4 = 0.
\]

The position of mobile coordinate \( \xi(t) \), connecting with the plunger movement, are calculated during numerical solution of the problem. Numerical algorithm of mechanical part of the problem and thermal conductivity problem in the area with variable size are described in [24-26]. Kinetical equations were solved with Euler method.

3. Problem formulation

This model allows investigating the evolution of the temperature, velocity, stresses, density, porosity and composition during sintering assisted by external mechanical loading. We can vary the size of the reagent, the initial density distribution, initial composition of the powder mixture, limiting stages for the reacting scheme, mechanical load and times, when the heating rate changes and external load acts. As a result we will obtain various composition of the final material. These calculations can be classified as numerical experiment.

Because the comparison of porosity evolution with experiment for chemically inactive system were demonstrated in [25, 26], here we demonstrate only the possibilities of the model that appear when chemical reactions are taken into account.

The figures 2 illustrate the composition change for powder mixture with the initial state \( Y_1 = 0.5; \ Y_2 = 0.5 \). The concentration surfaces correspond to times \( t = 1. - 3.5 \) c; \( 2 - 4 \) c; \( 3 - 4.5 \) c; \( 4 - 5 \) sec.

![Figure 2. Evolution of the Ti and TiNi in time and in space.](image-url)
For used here parameters, the chemical reactions proceed quite not uniformly, and concentrations values. Due to the conjugate heat exchange, heat losses to the walls retard the reaction and temperature growth. Chemical reaction rates change with time not monotonically. In this case it is not possible present the concentrations in the same scale, and the pictures have only qualitative character. After the sintering, the material contains basically TiNi$_3$ and Ti$_2$Ni. Other phases stay because the conditions were irreversible and temperature field was not uniform.

The average composition in time is shown in the Fig.3. We see really that TiNi concentration growths firstly, and then decreases. Additionally note that stresses in the material rises when porosity and composition change. Then, during cooling, stresses relax because external load does not apply.

![Figure 3. Average composition of the material in time during reactive sintering](image)

**Conclusion**

Thus, the model of sintering process assisted by mechanical loading in closed volume was suggested. The stages of chemical conversion are taken into account. It was demonstrated that the composition of the final product can be non uniform due to irreversible conditions with not uniform temperature field. The model can predict the composition change with technology parameters variation. However the stresses in the reaction zone can appear due to temperature gradient and composition change. In this case the mechanical sub problem will two-dimensional, and one can expect new features appearance in the process course.

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