

CFD ANALYSIS OF EXOTHERMIC REACTIONS IN Al-Au NANO MULTI-LAYERED FOILS

CFD-ANALIZA EKSOTERMNIH REAKCIJ V VEČPLASTNIH NANOFOLIJAH Al-Au

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This work presents the possibility of numerical modelling using Computational Fluid Dynamics (CFD) in the field of nano-foils. The governing equations were solved using a Finite Volume Methodology (FVM). The computational domain was discretized using a uniform Cartesian grid with the appropriate mesh size along the x and y directions employing the corresponding number of grid points. The field variables were discretized at the cell centres and the spatial, as well as the time, derivatives were approximated using the second-order accurate numerical scheme. The time-evolution of the temperature and concentration fields, as well as the atomic diffusion coefficient, will be presented for the appropriate Al-Au nano-foil geometry and boundary conditions.

Key words: Au-Al nano-foils, finite volume method, temperature and concentration transfer

V delu je predstavljena možnost numeričnega modeliranja z uporabo računske dinamike fluidov (CFDC) v primeru nanofolij. Ustrezne enačbe so bile rešene z uporabo metodologije končnih prostornin (FVM). Področje izračunov je bilo opredeljeno z uporabo enakomerne kartezijske mreže s primerno velikostjo vzdolž smeri x in y z ustreznim številom mrežnih točk. Spremenljivke polj so bile opredeljene za središče celic in prostorske ter časovne derivate ter aproksimirane z uporabo numerične sheme z drugim redom velikosti natančnosti. Časovni razvoj temperaturnih in koncentracijskih polj in koeficient atomske difuzije sta predstavljena za ustrezno geometrijo Al-Au-nanofolij in mejne pogoje.

Ključne besede: Al-Au nanofolije, metoda končnih elementov, prenos temperature in koncentracije

1 INTRODUCTION

Combustion synthesis, also called reaction synthesis, is a process in which two or more materials with large exothermic heats of mixing, known as reactants, are combined together. When the reactants are heated sufficiently, they begin to spontaneously intermix on the atomic scale, releasing heat in the process. A large body of literature exists on this process, and it has been reviewed extensively.¹

In general, combustion synthesis has been accomplished using mixed reactant powders that are pressed into a pellet of a certain green density. The combustion is initiated by an external energy source to heat the compact. Once combustion begins, the energy released by intermixing contributes to an increase of the local temperature, resulting in faster intermixing and combustion. A large amount of materials are reported to have been synthesized with this general method, including carbides, borides, silicides, nitrides, sulfides, hydrides, intermetallics, and complex composites. Combustion synthesis has many advantages, such as the ability to create high-melting-temperature materials in a low-temperature

process, near-net-form fabrication, and rapid material synthesis.

The ignition of combustion synthesis can be subdivided into two different modes. The first mode, important for present research, is termed self-propagating high-temperature synthesis, or SHS. In this method, the compact is heated locally using an external heat source. The local heating initiates the reaction locally, releasing heat that drives the reaction forward. The reaction moves across the compact in a self-propagating manner, driven by its own heat. The other method of ignition is termed thermal explosion, or simultaneous ignition.

However, powder compacts have several limitations when used for combustion synthesis. The density of the final product is often limited by the green density of the powder compact. If the combustion synthesis reaction is solid state, the particles can only inter diffuse with each other where they are in physical contact. The size of the powder particles is often large compared to the characteristic diffusion distance for a given system, making it difficult to achieve full intermixing. Particles of highly reactive components will often have a passivating outer

coating that acts as a barrier to the diffusion with other reactants. Moreover, voids between the powder particles limit thermal diffusion through the compact, reducing the ability of the reaction to be driven by its own heat.

With modern thin-film deposition techniques, fully dense multilayer materials with similar exothermic reactions can be fabricated (**Figure 1**). Such reactive multilayer foils consist of hundreds or thousands of nanometer-scale alternating layers of two or more materials, known as reactants, which can mix exothermically. When heat is applied locally to the foil, it undergoes SHS in a fashion similar to powder compacts. Structures of reactive multilayer foils offer several potential improvements over powder compacts.

The individual layers in a reactive foil are usually on the scale of tens of nanometers, which significantly decreases the diffusion distances involved in interatomic diffusion between the reactants. The thickness of the individual reactant layers can be controlled during fabrication, allowing significant control over the properties of the foil. The individual layers are in intimate contact with each other, which increases both thermal and atomic diffusion in the system while also eliminating voids in the system. Moreover, having thin layers of reactants that are in intimate contact with each other makes the SHS reaction propagate faster in most reactive foils than in a powder compact with the same components.

In an A-B multilayer system (here Al-Au), when one end of the foil is ignited with a small thermal pulse (such as a spark) at room temperature, local atoms will diffuse normal to the layers with A-A and B-B bonds being exchanged for A-B bonds, as shown in **Figure 1**.

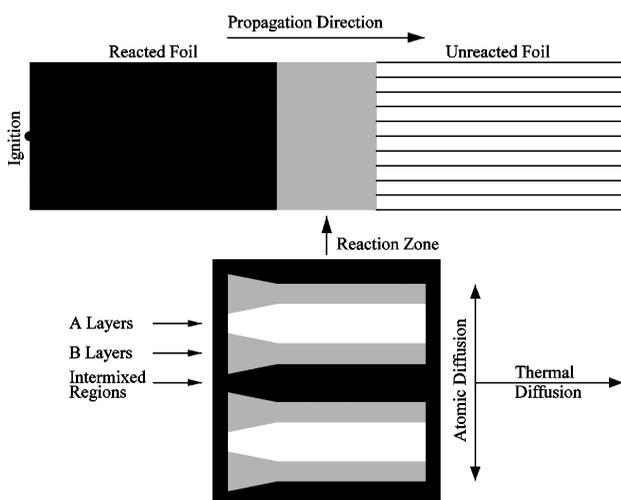


Figure 1: Schematic representation of a self-propagating reaction in a multilayer foil that is propagating from left to right. The unreacted foil consists of alternating layers of elements A and B with an intermixed region between the layers.

Slika 1: Shematična slika samonapredujuće reakcije z leve na desno v večplastni foliji. Nezreagirana folija je iz izmeničnih plasti elementov A in B s področjem mešanja med plastmi.

In this process, the heat is released and conducted parallel to the layers. If the atomic diffusion and energy release are sufficiently fast, then the reactions are self-propagating. These reactions can travel as fast as 25 m/s and can reach temperatures above 1500 °C.

Numerous studies of interface reactions have been performed in different multilayer systems. A common feature of these multilayers is the apparently large free energy, typically several tens of kJ/mol, available for the phase formation. This leads us to conclude that all possible product phases should be able to form from the very beginning of the reaction.

The aim of the present work is to support the development of novel reactive Au-Al nano-multilayered material that enables the rapid bonding of similar and dissimilar materials by using a Computational Fluid Dynamics (CFD) analysis. The advantage of numerical modelling is that, once the model is set up and established, a wide range of scenarios can be investigated with relatively little effort, and complex two-dimensional² as well as three-dimensional³ problems may be solved using numerical models. The potential use for the reactive Al-Au nano-foils is as a controllable, localised heat source for joining applications^{4,5}. To optimize the performance of the Al-Au foils it is necessary to have a clear understanding of the physical processes that dictate the reaction temperature, the rate of heat generation and the velocity at which the reaction propagates along a foil.

2 NUMERICAL MODELLING

The formulation of the mathematical model used to describe the self-propagating reactions in multi-layered Al-Au nano-foil is based on the following assumptions: (1) the effects of phase changes in the foil (e.g., melting of the reactants and/or products) are neglected; (2) the atomic diffusion is represented by a single binary diffusion coefficient D ; (3) the physical properties of the foil (e.g., density ρ , thermal conductivity λ and specific heat c_p) are assumed to be dependent on the composition. Last but not least, the width of the foil (z) is large in comparison to the thickness (y) and small in comparison to the length (x). As a result, it is possible to treat the thermal and atomic diffusion in the foil as a two-dimensional problem.

2.1 Governing equations

Under the assumptions above the atomic mixing is described using a time-dependent, conserved scalar field $C(x,y,t)$, defined such that $C = 1$ for pure Au and $C = 0$ for pure Al. The evolution of C is governed by:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x_j} \left(D \frac{\partial C}{\partial x_j} \right) \quad (1)$$

The atomic diffusivity is assumed to be independent of the composition and to follow the Arrhenius dependence on the temperature, according to:

$$D = D_0 \exp\left(-\frac{E}{RT}\right) \quad (2)$$

where D_0 is the Arrhenius pre-exponent, E is the activation energy and R is the universal gas constant. The values and $E = 25.20$ kJ/mol as used in the present study are taken from the work of Fouracre⁶.

The time-evolution of the concentration field is coupled with the temperature equation:

$$\frac{\partial T}{\partial t} = \frac{\partial}{\partial x_j} \left[\frac{\lambda}{\rho c_p} \frac{\partial T}{\partial x_j} \right] + \frac{\partial}{\partial t} \left[\frac{Q(C)}{\rho c_p} \right] \quad (3)$$

where the physical properties (thermal conductivity λ , density ρ and specific heat c_p) in any control volume of the domain are given by the corresponding fraction of the phase (Au and Al) in that volume. They are mathematically obtained as:

$$\begin{aligned} \lambda &= \lambda_{Au} C + \lambda_{Al} (1-C) \\ \rho &= \rho_{Au} C + \rho_{Al} (1-C) \\ c_p &= c_{p,Au} C + c_{p,Al} (1-C) \end{aligned} \quad (4)$$

The rate at which the heat is generated ($\partial Q(C)/\partial t$) is assumed to be proportional to the rate at which the composition of the foil changes. In the simplest case, the linear relationship between the composition and the energy released can be assumed⁷. However, a parabolic function may be closer to the real situation.^{8,9}

$$Q(C) = -\rho c_p (T_{f0} - T_0) C^2 \quad (5)$$

and this was used in the present study.

T_{f0} and T_0 are the adiabatic temperature of the reaction and the initial temperature of the foil.

2.2 Geometry, boundary and initial conditions

The laminar and time-dependent thermal and species transport in a two-dimensional nano-foil was considered as depicted in **Figure 2**.

At the inlet we prescribed the constant temperature $T = 1000$ K, while the convective heat flux condition (for natural convection along the vertical plate) was used at

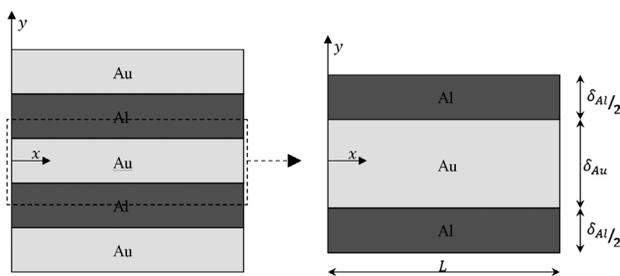


Figure 2: Schematic of multi-layered Au-Al nano-foil. $L = 2000$ nm, $\delta_{Au} = \delta_{Al} = 100$ nm

Slika 2: Shema večplastne Au-Al nanofolije. $L = 2000$ nm, $\delta_{Au} = \delta_{Al} = 100$ nm

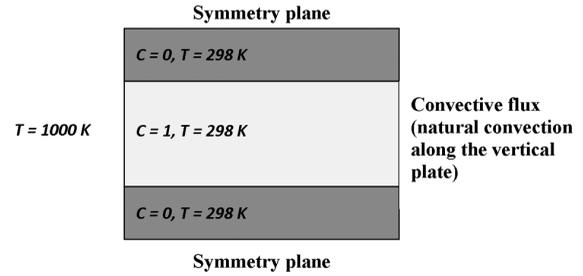


Figure 3: Boundary and initial conditions

Slika 3: Začetni in mejni pogoji

the outlet of the nano-foil. The initial conditions were as follows:

- Temperature $T = 298$ K in the whole nano-foil.
- Concentration $C = 1$ for Au and $C = 0$ for Al, see **Figure 3**.

For the nano-foil, the following physical properties were used for Au:

- Density $\rho = 19320$ kg/m³
- Specific heat $c_p = 130$ J/kgK
- Thermal conductivity $\lambda = 318$ W/mK

and Al:

- Density $\rho = 2700$ kg/m³
- Specific heat $c_p = 910$ J/kgK
- Thermal conductivity $\lambda = 237$ W/mK

2.3 Numerical procedure

The governing equations were solved using with the Ansys CFX numerical code which employs a standard finite-volume methodology¹⁰ with all the variables defined at the centre of control volumes populating the physical domain being considered. Each equation is integrated over the control volume to obtain a discrete equation which connects the variable at the centre of the volume with its neighbours.

The computational domain was discretized using a uniform Cartesian grid of mesh size $\Delta x = \Delta y$ along the x and y directions, respectively. The corresponding number of grid points is denoted by $N_x = 800$ and $N_y = 80$. For a spatial discretization of diffusive terms, the second-order accurate scheme based on central-differences was used, while temporal derivatives were approximated with a second-order accurate backward differences for a fixed time step $\Delta t = 10^{-9}$ s.

3 RESULTS AND DISCUSSION

The time-evolution of both the temperature and concentration field is presented in **Figure 4**. It can be seen that that the temperature (i.e., thermal) transport is essentially unidirectional (mostly taken in the axial direction) and occurs parallel to the layering.

In contrast to other studies^{7,8,9,11} where the atomic diffusion was modelled as a one-dimensional pheno-

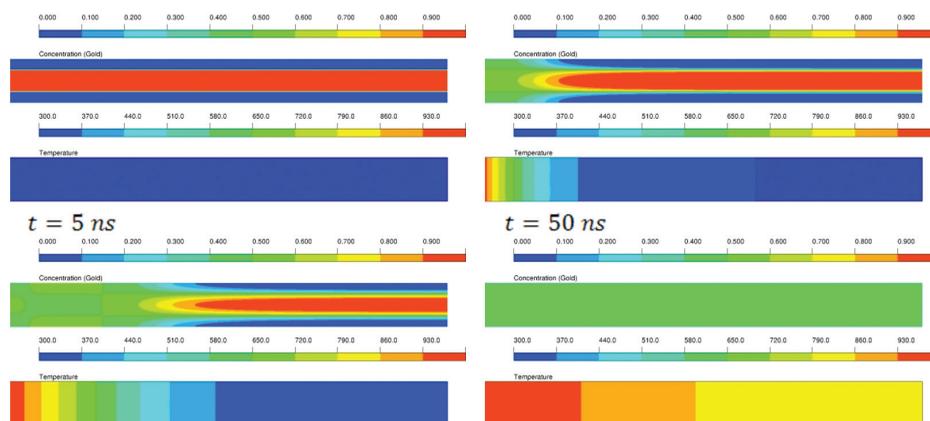


Figure 4: Time evolution of the temperature and concentration field
Slika 4: Časovni razvoj temperaturnega in koncentracijskega polja

menon (occurring normal to the layering) the present study shows that the atomic diffusion is actually a two-dimensional phenomenon (see **Figure 4**). This is due to the fact that the diffusion coefficient D , as defined by Eq. (2), varies with the temperature (as a matter of fact it increases as the temperature increases) in the axial direction. Of course, if the diffusion coefficient is kept constant, the concentration diffusion would occur only in the streamwise direction (due to the concentration gradient).

Finally, in the present study the concentration diffusion is a much faster process due to the higher value of the diffusion coefficient D in comparison to the value of the thermal diffusion coefficient α .

4 CONCLUSIONS

In the present study, the time-dependent temperature and concentration diffusion in the multi-layered Al-Au nano-foil was studied numerically using mathematical modelling and a finite-volume-based CFD code. The atomic diffusion coefficient D was taken to be independent of the composition and to follow the Arrhenius dependence on the temperature while the physical properties (and therefore the thermal diffusion) in any control volume of the domain were taken to be dependent on the corresponding fraction of the phase (Au and Al) in that volume.

The present numerical results show that the temperature diffusion is essentially unidirectional (in the axial direction) while the atomic diffusion occurs in both (i.e., axial and streamwise) directions due to the variable diffusion coefficient and the concentration gradient. The latter is an important fact (influencing the thickness of the intermixed region between layers as well as the heat

release in the process) which has been ignored in most of the existing analytical as well as computational models.

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