

ON THE MODELLING OF SOLID-STATE REACTIONS. SYNTHESIS OF YAG

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The composition $3\text{Y}_2\text{O}_3 : 5\text{Al}_2\text{O}_3$ commonly called as yttrium aluminium garnet ($\text{Y}_3\text{Al}_5\text{O}_{12}$, YAG) is an important solid-state laser material widely used in luminescence systems and window materials for a variety of lamps [1; 2; 3; 4; 5; 6]. In view of the high-temperature chemical stability and an extremely high creep resistance, YAG is a promising fiber material for the preparation of ceramic composites [7; 8; 9; 10; 11; 12; 13; 14; 15; 16]. The electrical conductivity of YAG is also reported to be lower than any other polycrystalline oxide [17]. Owing to such wide and diverse application potential of YAG-based ceramics, new routes for the synthesis of pure and homogeneously doped yttrium aluminium garnet are highly desirable.

The solid-state synthesis of YAG ceramic from Al_2O_3 and Y_2O_3 powders usually requires extensive mechanical mixing and lengthy heat treatments above 1600°C [18; 19]. Several wet-chemical techniques such as polymerized complex route [20], metal-organic preceramic processing [18], coprecipitation methods [21] or yttrium carboxylate-alumoxane route [3] have been used to produce YAG phases. Most of these methods suffer from the complex and time consuming (long refluxing times, gelation periods of several days, etc.) procedures and/or mismatch in the solution behaviour of the constituents.

Recently, for the preparation of nanocrystalline yttrium aluminium garnet we have developed a new sol-gel process using mixtures of inorganic salts of the respective elements [22; 23]. This research have demonstrated the versatility of the sol-gel method to yield monophasic YAG samples at lower sintering temperature 1000°C when compared to the temperature required for the solid-state synthesis ($>1600^\circ\text{C}$). The successful synthesis of crystalline YAG phase at 1000°C is the one of the lowest reported temperature for the crystallisation of this material. The sol-gel method of preparation of YAG in aqueous media is inexpensive and thus appropriate for the large scale production of YAG ceramics.

Thus, it is clear that the conditions for the formation of monophasic YAG are dependent largely on the synthesis method used. By changing solid-state method to the sol-gel chemistry approach, the YAG formation temperature decreases from 1600°C upto 1000°C . However, the important question concerning the reasons for the observed changes in the preparation temperature by changing synthesis method remains to be answered. Such a situation has initiated the present work, motivating us to elucidate the reasons responsible for the low-temperature formation of $\text{Y}_3\text{Al}_5\text{O}_{12}$ using sol-gel technique. The optimization of synthesis parameters of sol-gel processes have been determined mostly in an experimental way, i.e., according to the desired properties of the final ceramic material. To overcome these difficulties, the pathway of chemical reactions should be performed according to the possible computational modelling [24]. However, no model has yet been constructed that provides quantitative agreement of the reaction mechanisms with the experimental data of process parameters and desired structural, morphological and physical properties of the final ceramic material.

In this article we present several physical models for YAG synthesis. In general case, reaction

mechanism can be described by the reaction–diffusion system

$$\begin{aligned}\frac{\partial c_1}{\partial t} &= \sum_{j=1}^3 D_1 \frac{\partial^2 c_1}{\partial x_j^2} - 3k c_1 c_2, \\ \frac{\partial c_2}{\partial t} &= \sum_{j=1}^3 D_2 \frac{\partial^2 c_2}{\partial x_j^2} - 5k c_1 c_2, \quad x \in V, \quad t > 0, \\ \frac{\partial c_3}{\partial t} &= \sum_{j=1}^3 D_3 \frac{\partial^2 c_3}{\partial x_j^2} + 2k c_1 c_2.\end{aligned}$$

$$c_i(x, 0) = c_i^0, \quad i = 1, 2, 3, \quad x \in \bar{V} = V \cup \partial V,$$

$$\left. \frac{\partial c_i}{\partial x} \right|_{x \in \partial V} = 0, \quad i = 1, 2, 3, \quad t \geq 0.$$

where $c_i = c_i(x, t)$, $i = 1, 2, 3$, is the concentration of the i th reaction component at a point x of the reaction space V at time t . In this article we studied one- and two-dimensional cases of the system.

Equations were solved numerically using finite difference technique. In one-dimensional case, the problem was approximated by symmetric implicit scheme, and in two-dimensional case—by alternating direction scheme. Both schemes were solved using stream sweeping method from [25].

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