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Numerical simulation of the homogenization process of a binary mixture in a ball mill

I.S. Polyakov[1,2,], O.V. Lapshin[1], E.N. Boyangin[1]*

[1]*Tomsk Scientific Center SB RAS, Tomsk, Russia*
[2]*Tomsk State University, Tomsk, Russia*
[*]*plkv.ivan@mail.ru*

Abstract. The process of mixing a mixture of various bulk substances is present in many technological processes. A ball mill is a widespread type of devices having a rotating axisymmetric chamber with the mixed material inside used for mixing processes. In this paper, a study of the homogenization process of a binary mixture in a ball mill using numerical modeling is carried out. The dependence of the number of contacts between particles on the mixing time has been studied. The dependence of the degree of homogenization on time, obtained using numerical modeling, is in excellent agreement with the results of processing experimental data.
Keywords: Homogenization, ball mill, binary mixture, numerical simulation, heterogeneous contacts.

1. Introduction

The process of mechanical mixing of powder mixtures in various kinds of mills is a widespread method to obtain a homogenized system [1]. It is known that the rate of chemical reaction of two substances largely depends on their mutual contact area, which, in the case of a powder mixture, directly depends on the homogeneity of the system [2].

It is obvious that during the transition of the system from a heterogeneous state to a homogeneous one with mechanical mixing, the contact area of the two substances increases. However, at present, in various technological processes, the assessment of this parameter is directly associated with significant difficulties or is impossible.

To estimate the area (or number) of contacts between particles of different material, the theory of mixing of solid particles is usually used [3–7]. According to the simplest model, mixing of a binary powder mixture consists of two stages – convective and diffusion. At the convective stage, particles are separated from the mass of a component of one grade and mixed with the same particles of a component of another grade. Due to this, the powder mixture is homogenized, which ensures surface contact of particles of dissimilar components with each other. Further, diffusion is carried out through this contact. Usually, the duration of the convective stage is short, its task is to form a contact surface through which mutual diffusion of components begins.

The resulting mixtures can belong to various types: random, partially ordered, ordered, interactive or non-interactive, cohesive, free-flowing, interacting with small particles and free-flowing with large ones particles, etc. The mixing process is significantly influenced by such factors as the method and speed of mixing, absolute and relative particle sizes, particle shape of components, surface charge, density, brittleness and plasticity of particles, the presence of fluid phases in the system, as well as solid additives affecting the rheology of the mixture.

In order to increase the efficiency of solid-phase thermally activated synthesis and increase the uniformity of the product in composition and structure, preliminary preparation of the initial reagents is practiced in order to achieve the best mixing, up to the atomic level. Thermal decomposition of correctly selected precursors [8], co-precipitation (for example, using the sol-gel method [9], homogenization of suspensions [10, 11] and other techniques are used. In the course of thermal solid-phase synthesis, it is often stopped in order to grind the sintered mass and free the surfaces of the partially reacted reagent particles from the product layer separating the reagents from each other and hindering further transformation.

Our work presents the results of numerical simulation of the mixing process of a binary mixture in the LIGGGHTS® opensource Discrete Element Method (DEM) package. The

assessment of the correspondence of numerical calculations to the real experiment was carried out using the methodology proposed in [12] for comparing the degree of homogeneity of a real mixture to its model analogue at fixed time points. Experimental data were obtained on the 3Ni + Al system.

2. Experimental procedure

Aluminum powders (ASD-4, average particle size ~ 5 μm) and nickel (PNK-UT4, average particle size ~4 μm) were used in the study. The powders, with a weight ratio of 13.3 Al / 86.7 Ni %, were mixed in the steel cylindrical drum of the IBMT-30 laboratory ball mill with certain time intervals, ranging from 10 seconds to 4 hours, at a speed of 75 rpm.

At each stage, samples weighing 2g were taken from the mixture to study the microstructure. The morphology and structure of the powders and synthesized specimens were studied using an Altami MET 1C optical microscope. The analysis of the homogeneity of the powder composition was carried out using optical images of powder mixture grinds using the "brightness histogram" function of the Adobe Photoshop. Since the nickel and aluminum particles had different colors and brightness, the total average brightness of the photo was first estimated, then the image was divided into 8 parts, and the average brightness of each part was measured. The standard deviation was calculated, and this was taken as the degree of homogeneity of the given composition of the mixture, mixed for a certain time.

3. Simulation and experimental results

To estimate the size of the reaction surface area, we built a numerical model using the LIGGGHTS® opensource discrete element method particle simulation code. The calculation of the number of contacts between particles was carried out using the *contact/atom* function, adapted by us to calculate heterogeneous contacts (Fig. 1). In the most general sense, LIGGGHTS® integrates Newton's equations of motion for collections of atoms, molecules, or macroscopic particles that interact via short- or long-range forces with a variety of initial and/or boundary conditions. For computational efficiency LIGGGHTS® uses neighbor lists to keep track of nearby particles. The lists are optimized for systems with particles that are repulsive at short distances, so that the local density of particles never becomes too large.

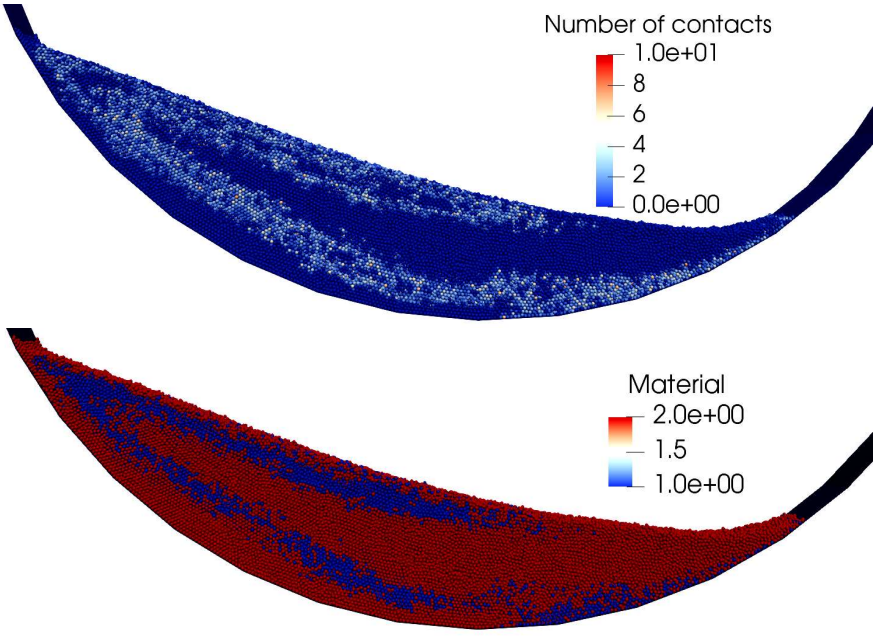


Fig. 1. Indication and counting of the number of contacts between simulated nickel (red) and aluminum (blue) particles.

Numerical simulation results (Fig. 2.) was presented in dimensionless parameters $n_c = N/N^*$ and $\tau = t/t^*$, where N – the current number of heterogeneous contacts, N^* – maximum number of heterogeneous contacts, t – time, t^* – time of reaching N^* during mixing. It was found that the rate of homogenization of the binary mixture increases with increasing particle size. In this case, the structure of the mixture becomes more evenly distributed, as a result of which the degree of its homogenization increases. It was also revealed that the rate of homogenization of the binary mixture increases with increasing particle size.

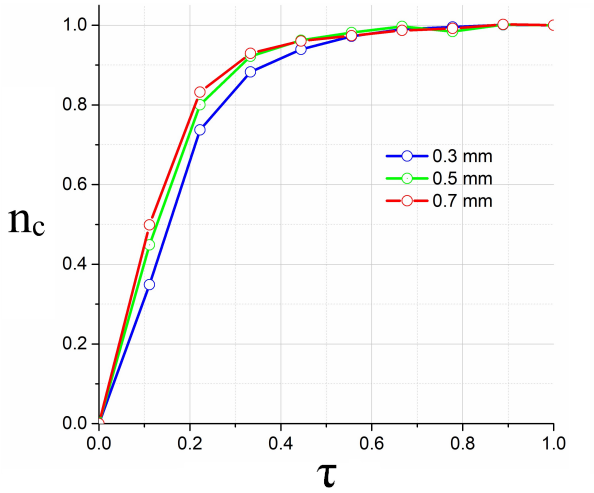
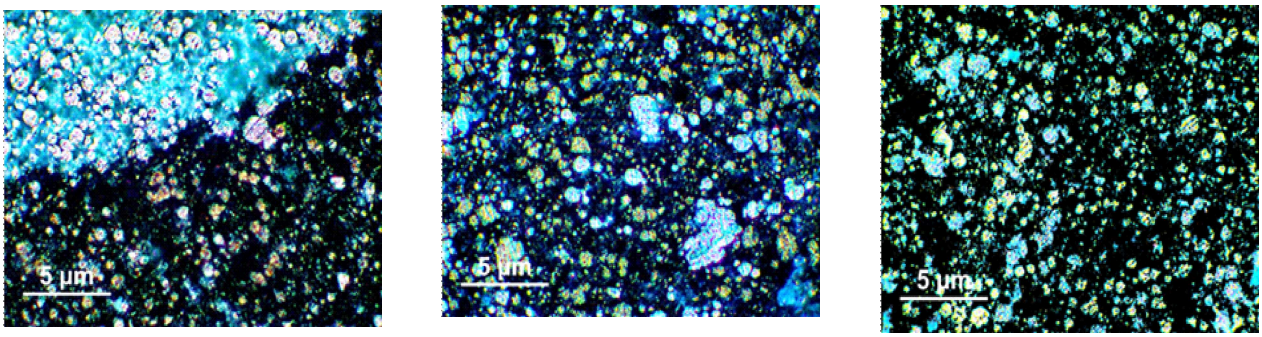


Fig. 2. The dependence of the degree of homogenization (the number of heterogeneous contacts) on time at different particle diameter.

Fig. 3 shows pictures of the microstructure of the 3Ni+Al powder mixture at various points of its mixing in the mill. It can be seen that with the prolongation of mechanical treatment, the redistribution of nickel and aluminum particles in the powder composition occurs, as a result of which the degree of homogenization of the mixture increases.



(a) **(b)** **(c)**

Fig. 3. Optical images of the grinds of the 3Ni+Al powder mixture from the mixing time in a mill without balls: a) 10 sec; b) 5 min; c) 1 hour.

A comparison of the number of contacts from the numerical experiment and the standard deviation of the average brightness of optical images of thin sections of the 3Ni+Al powder mixture from the mixing time showed their good agreement with each other (Fig. 4). From the figure it can be seen that the maximum number of heterogeneous contacts in the mixture is achieved in about the same mixing time, equal to 2 minutes, which and its complete uniformity, experimentally determined as the minimum value of the standard deviation of the average brightness of optical images.

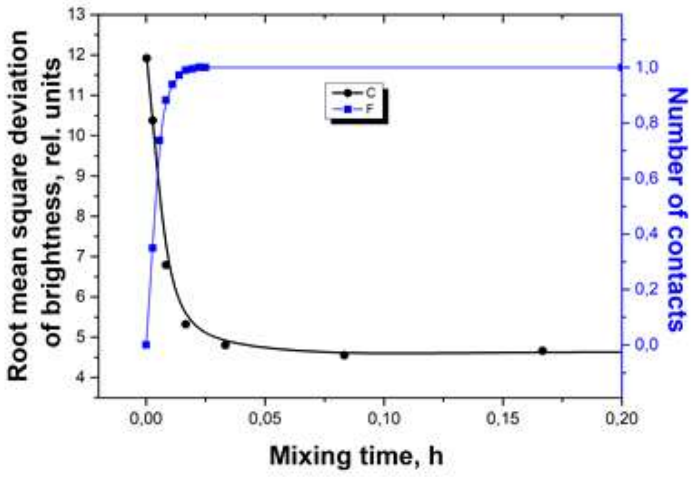


Fig. 4. Comparison of the number of contacts from a numerical experiment and the standard deviation of the average brightness of optical images of 3Ni+Al powder mixture grinds from the mixing time in a mill without balls.

4. Conclusion

Thus, the following results were obtained:

- an experimental study of the degree of uniformity of the powder composition 3Ni+Al from the time of machining in a low-energy mill was carried out. The degree of uniformity of the powder composition was analyzed using the "brightness histogram" function from optical images of powder mixture grinds using Photoshop. It was assumed that the complete uniformity of the composition corresponds to the minimum value of the standard deviation of the average brightness of optical images;

- numerical simulation of the mixing process of a binary mixture similar to the composition of 3Ni+Al in the LIGGGHTS® open source Discrete Element Method (DEM) package has been performed. It was found that with increasing mixing time of the mixture, the number of heterogeneous contacts increases to its maximum value, after which it ceases to change. It was also revealed that the rate of homogenization of the binary mixture increases with increasing particle size.;

- numerical simulation has shown that the maximum number of heterogeneous contacts in the 3Ni+Al mixture is achieved in about 2 minutes. A good agreement was obtained between the numerical calculation and the data of the physical experiment.

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