Modeling and comparative analysis of packing of the polyfractional mixture of B_4C powder

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Abstract. Modeling of the real polyfractional powder mixtures has been carried out using the data obtained by SEM, XRD and grain-size analysis. Parameters of the dense packing of powder mixture particles were defined. The influence of every fraction on the packing density of powder mixtures has been deduced.

1. Introduction

The main problem of powdered microstructure modeling, using dense packing of equivalent particles, is the shortage of initial data about the grain-size composition, i.e. knowledge incompleteness of the spatial positioning of microparticles and its geometric shape. Basically the real shape of the powder particles is idealized in a model by elemental bodies (usually by spheres) [1]. As a result of particle packing modeling, with satisfactory accuracy, imitating the microstructure, the task adds up to the search of the value of model input parameters responsible for the characteristics of grain-size composition [2, 3].

2. Experiments

High-clean boron carbide powder mixtures with 5-15 wt.% of nano additive content (B_4C-PIHT1, B_4C-PIHT2, B_4C-PIHT3, B_4C-PIHT4) were obtained by combination of mechanical and chemical methods [4]. The initial powder were milled, then purified of the impurities of enriched milled nanopowder particles, rinsed and dried. After that, the mixture was deagglomerated and finally high-clean powder with specified nanosized fraction was obtained.

Averaged values of powder mixture morphologic parameters, obtained by laser diffraction (LD), BET, SEM and XRD methods, are presented in the Table 1. According to SEM data, the powders consisted of equiaxed particles with a broad particle size distribution and practically did not form agglomerates.

According to the XRD pattern analysis, PIHT powder mixtures contained not more than 3 wt.% of H_3BO_4. The content of iron in the different samples varied from 0.16 wt.% to 0.51 wt.% with the mean value of 0.32 wt.%. There were not less than 79 wt.% of boron content in the powders.

The analyses of XRD diffractograms with the separation of peak broadenings on the components of different boron carbide fractions (Figure 1) showed the average size of coherent-scattering region of only submicron powder fraction. These results satisfactorily correlate with the results of nanosize fraction contents obtained by laser diffraction method (Table 1).

The summary results of the powder materials study are presented in the Table 1.

Table 1.Summary results of the powder materials study

	PIHT-1(1)	PIHT-2(2)	PIHT-3(3)	PIHT-4(2)
Specific surface area, S_{ss}, m^2/g	3.71	5.94	5.11	5.20
Particle size (BET) d, μm	0.64	0.40	0.47	0.46
Particle size (SEM) d, μm	2.6	2.9	4.2	2.7
Particle size (LD) D, μm	1.87	2.74	3.81	2.56
Agglomeration degree, nD	2.93	6.86	8.10	5.56
B$_4$C content, wt.%	97.3	99.8	97.8	98.9
H$_3$BO$_3$ content, wt.%	2.7	0.2	2.2	1.1

3. Results and discussion

Grain-size composition and SEM images of the particle of synthesized powders are presented in Figure 2.

Based on the experimental data of grain-size analysis, the modeling of optimal particle packing has been carried out in order to optimize the process of PIHT powder consolidation. The modeling objective of the densest polyfractional PIHT powder particles, when the fine particle fractions effectively fill the spaces between larger particles, was being solved.

According to SEM analysis, since PIHT powder particles have equiaxed shape, it is possible to formalize them in a packing model as a class set of spherical particles of appropriate grain-size composition. This fact had been taken into account in order to perform the modeling.

$$Qv = \frac{A}{w \cdot r \cdot \sqrt{2\pi}} \cdot exp\left(\frac{-\left[\ln\frac{r}{<r>}\right]^2}{2w^2}\right) \tag{1}$$

where Qv – percent fraction content of particles with radius r, A – amplitude value of approximating class, w – class standard deviation, $<r>$ - mean values of particle class radius.

Quantitative variables of modeled objects, obtained by discrete element method using S3D PorouStructure software (Ichikawa's algorithm, central packing), were used as an evaluation criterion of powder possibility in forming dense powder packing. Multimodal behavior of synthesized powder particle grain-size distribution has been taken into consideration for model construction of packings (Figure 2). In order to describe experimental data of granulometry, the sum of normal logarithmical distributions with the profile, described by equation (1), has been used.

Data treatment of the grain-size distribution results, obtained by LD, showed, that investigated powders consist of 4-5 particle classes with logarithmically normal and quite broad size range (from tens of nanometer to tenths of microns, Figure 2). Let us denote these fractions for all powders by class serial number from 1 to 5, with the increase of mean grain size (Table 2). In addition, parameters of the constituent classes of this powder distribution function (mean particle radius r (μm), standard deviation w) were defined. The transition from diameter to radius of the particles is governed by the format of input parameters, which are used in discrete element packing model.

Table 2. Resulting parameters of synthesized powder packings

Parameter	Powder	Class 1	Class 2	Class 3	Class 4	Class 5	mixture
Mean coordination number N_c	PIHT-1(1)	6.0	244.1	4659.7	88554.5	541091.1	6.2
	PIHT -2(2)	5.0	25.6	86.3	2174.7	459.0	6.1
	PIHT -3(3)	4.7	20.5	234.8	-	1337.0	6.2
	PIHT -4(2)	5.3	34.7	1231.0	-	95.3	6.2
Relative packing density	PIHT -1(1)	0.138	0.081	0.108	0.529	0.045	0.901
	PIHT -2(2)	0.054	0.098	0.141	0.488	-	0.779
	PIHT -3(3)	0.052	0.253	0.381	-	0.066	0.752
	PIHT -4(2)	0.123	0.137	0.520	-	-	0.780

Comment. Class color corresponds to the color of the presented below plots and images of packing models.
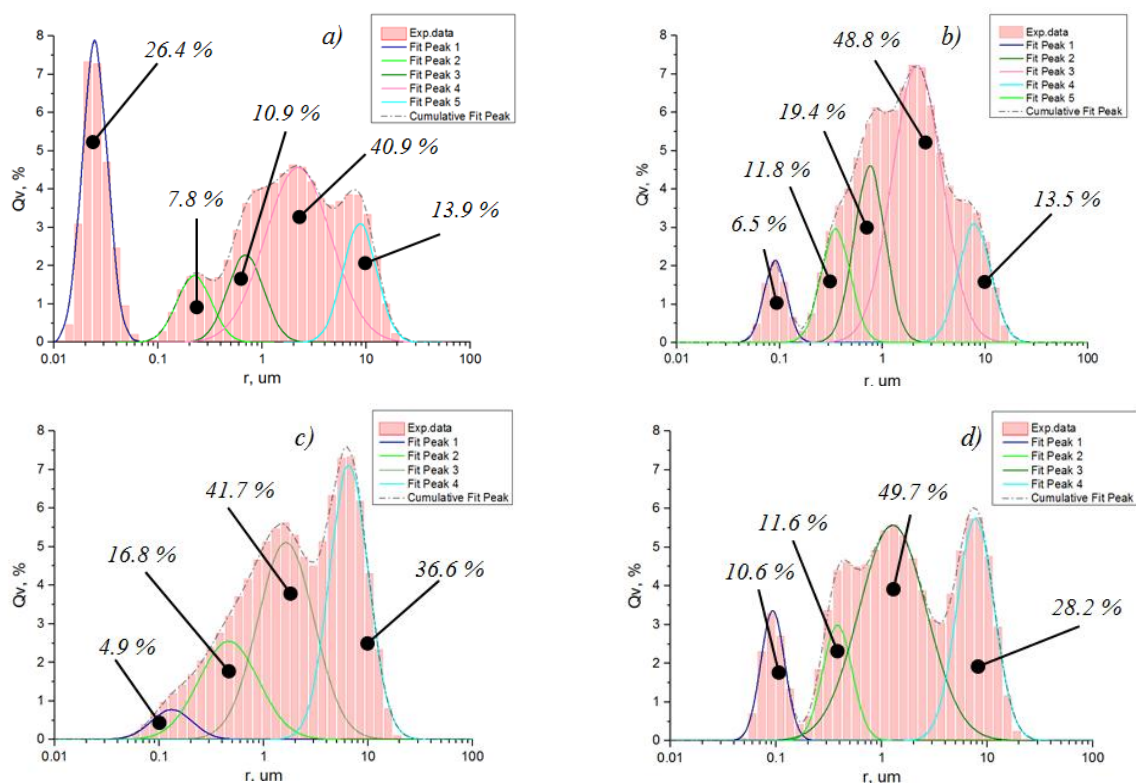

Figure 2. Results of grain-size analysis data expansion of the powder particles on the class fractions of logarithmically normal distribution: *a)* − mixture PIHT-1(1); *b)* − mixture PIHT-2(2); *c)* − mixture PIHT-3(3); *d)* − mixture PIHT 4(2).

PIHT-1(1) powder have distinctly more submicron and nanofraction content and they are shifted more to the smaller particle size (Figure 2 a). The behavior and distribution ranges of the PIHT-2(2) larger (micron) fractions are sufficiently close to the PIHT-1(1). In addition, the PIHT-2(2) micron fractions are considerably overlapped, forming close to normal distribution in a range from 0,2 to 20 μm. There are two factors which can favor the process of powder packing and consolidation. First, the relatively high individual content of submicron fraction (from 6 to 20%) in PIHT-1(1) and PIHT-2(2), PIHT-4(2) (Figure 2 a,b,d). Second, the presence of large particle distribution close to uniform in a range of two orders of magnitude. At the same time, fine fraction acts not only as a filler of the free space between large particles, but also forms a buffer active layer on the boundaries between large particles. This buffer layer eases large grain intermotion (imitation of plastic deformation of powder body) under the prepressing pressure and provides activation processes of high-temperature sintering due to the well-known effect of nanosized state.

Packing models, build by discrete element method using obtained parameters of distribution functions, consisted of representative set of particles in amount from 600000 to 200000. The images of typical packing sections, where each particle class has its own color, are presented in Figure 3.
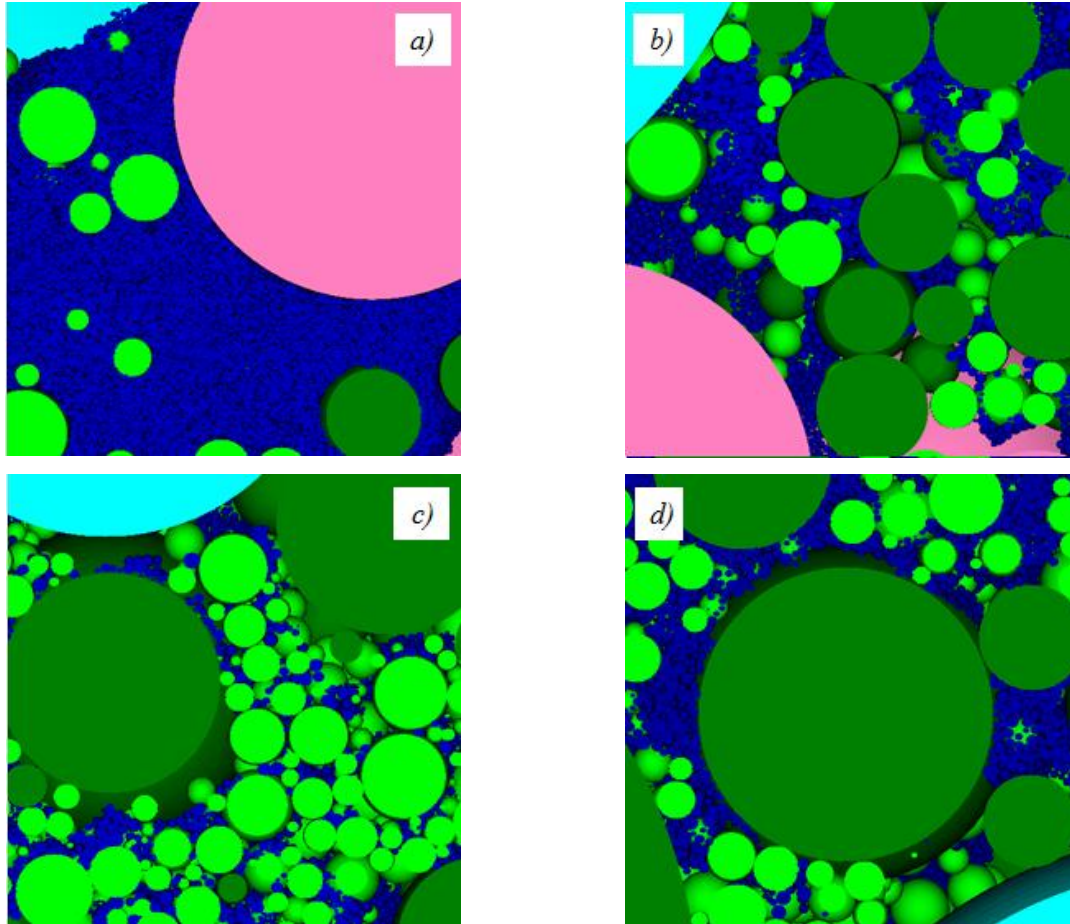

Figure 3. Section fragments of particle packings of different PIHT powder classes, obtained by discrete element modeling using data of granulometric analysis: *a)* − mixture PIHT-1(1); *b)* − mixture PIHT-2(2); *c)* − mixture PIHT-3(3); *d)* − mixture PIHT-4(2).

Comparative analysis of the obtained results (quantitative parameters of packings, Table 2) allows to make several conclusion regarding the optimization of grain-size composition of boron carbide powders, produced from low-quality and waste products by developing methods. These conclusions are of a great importance for further production of high-quality consolidated articles. One clear example of the influence of grain-size composition and parameters of powder particle classes on the packing characteristics is the accumulation curve of contribution of these classes to the compaction, presented in Figure 4. In particular, PIHT-1(1) sample, where nano fraction effectively fills the spaces between large particles of wide and continuous fraction composition, has the most dense packing (other conditions of the model being equal).
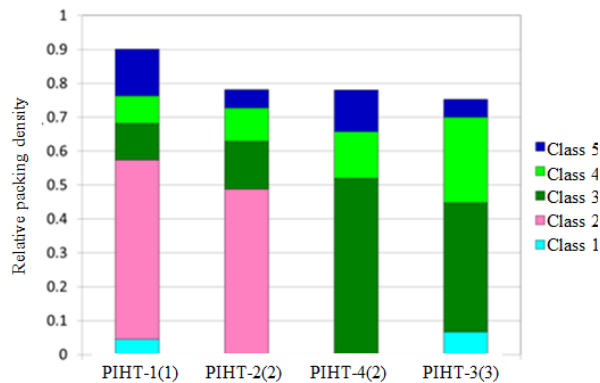
Figure 4. Accumulative density plot of PIHT powder particle packing

The obtained results allow to make several conclusions regarding optimization study.

1. Particle classes with highest content make the main contribution to the packing density. At the same time, class 5 with the maximum particle size influences the less effectively on the density increase.

2. The contribution of the finest particle fraction (class 1) to densification of all powder considerably depends on the difference of mean size and the size of the next class. The higher this difference, the most effectively submicron and nanoparticles of class 1 fills the spaces between particles of large fractions.

3. The interruption of continuous distribution (mainly the absence of class 4 in the PIHT-3(3) and PIHT-4(2) samples) negatively affects the packing density, which can not necessarily be compensated by the presence of individual finely dispersed fraction. Moreover, some trend of the directly proportional dependence of every class contribution to the densification on its percentage content in the packing is observed (Figure 5).
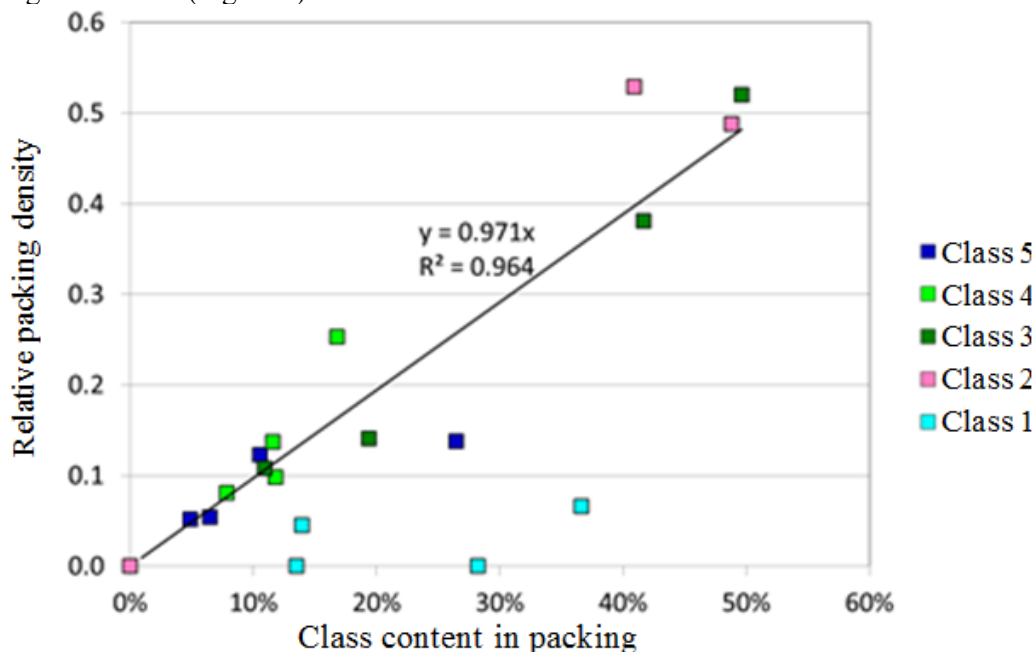

Figure 5. Correlation between relative class density and its content in model packing.

Considerable deviation from such dependence is observed only for the large particle class (class 5), which can be described by its relatively small content in the model packing (up to 100 units versus

half a million particles in the other classes). However, the mentioned feature can be used in order to preliminary evaluation of particle contribution of different fractions to the density of consolidated powders.

Acknowledgements

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