

RESEARCH INTO THE EFFECT OF PARTICLE SIZE DISTRIBUTION OF NEUTRON-ABSORBING POWDER MATERIALS ON THE COMPACTION IN ABSORBING ROD CLADDINGS

I.O. Chernov, M.M. Belash, V.O. Romankov, V.M. Yevseyev, Y.S. Legenkyi
“Nuclear Fuel Cycle” Science and Technology Establishment
of National Science Center “Kharkov Institute of Physics and Technology”,
Kharkiv, Ukraine
E-mail: chernov@kipt.kharkov.ua

The results of research into the effect of the granulometric compositions of neutron-absorbing powder materials such as boron carbide, dysprosium titanate and dysprosium hafnate and their mixtures in the initial state on the characteristics of backfills in the claddings of both “short” and full-size absorbing rods are presented. It was revealed that the achievement of the maximum backfill density is facilitated by providing the content of a fine fraction of about 30%. The maximum backfill density values achieved for boron carbide, dysprosium titanate, and dysprosium hafnate were 1.82, 5.1, and 5.82 g/cm³ respectively. The optimal modes of vibration compaction (type and frequency of oscillations), which ensure the achievement of the required values of the backfill density in absorbing rods in a short time were defined.

INTRODUCTION

In the actual design reactor absorber elements of WWER-1000, dysprosium titanate and boron carbide are used as neutron-absorbing materials in the form of powder fills. The use of dysprosium hafnate powder or pellets, as well as the use of combined heterogeneous powder compositions of boron carbide and titanate/dysprosium hafnate with a volume fraction from 10 to 50% is considered promising [1, 2].

It is believed that one of the most effective ways to form homogeneous, dense, and unstressed structures of dispersed powder material is the process of vibration compaction [3]. Vibration compaction technology has also been used in the manufacture of nuclear reactor fuel elements [4, 5]. One of the technological tasks in the development of powder backfill technology is to determine the effect of the particle size distribution of powders on the maximum achievable backfill density.

The process of vibration compaction is considered to be one of the most effective ways to form homogeneous, dense, and unstressed structures of dispersed powder material [3]. Vibration compaction technology has also been used in the manufacture of nuclear reactor fuel rods [4, 5]. One of the technological tasks in the development of powder backfill technology is to determine the effect of the particle size distribution of powders on the maximum achievable backfill density.

Guiding the known principles of packaging polydisperse materials, according to which, to achieve maximum packaging and backfill density, it is recommended to use powders of two and three fractions when selecting the fractional composition of powders B₄C, Dy₂TiO₅, Dy₂O₃·HfO₂ for testing the process of their vibration compaction in absorbing rod claddings [6]. The following conditions are recommended: the average particle size of the first (coarse) fraction D₁ = 1/10 of the inner diameter of the absorbing rod cladding (D), and the particle size of additional fractions (second and third) is determined from the following relations D₂ ≤ 0.15 · D₁, D₃ ≤ 0.15 · D₂. Thus, the packing factor (δ)

of the powder system of a 2-fraction composition is 0.867, and for a 3-fraction composition it is 0.951. In this case, the mass content of coarse, medium, and fine fractions should be 67:24:9.

The main technical requirements for backfill densities of absorbing rods with absorbing materials are as follows:

- dysprosium titanate: not less than 4.9 g/cm³;
- boron carbide: not less than 1.7 g/cm³.

The objective of this study is to research the effect of the particle size distribution of boron carbide, dysprosium titanate, dysprosium hafnate powders and their mixtures on the bulk density in the initial state and the density of backfill in the absorbing rod claddings.

1. MATERIALS AND METHODS

Powders with a size of -1+0.315 mm were used as a coarse fraction of boron carbide, and as a fine fraction – F240 grit with a size of less than 60 μm, manufactured by the Zaporizhzhya Abrasive Plant according to TU U 24.1-00222226-047:2005.

Powders of dysprosium titanate and dysprosium hafnate were obtained in the laboratory of the NFC STE NSC KIPT. Powders with a size of -0.9+0.1 mm were used as a coarse fraction, and powders less than 0.1 mm were used as a fine fraction.

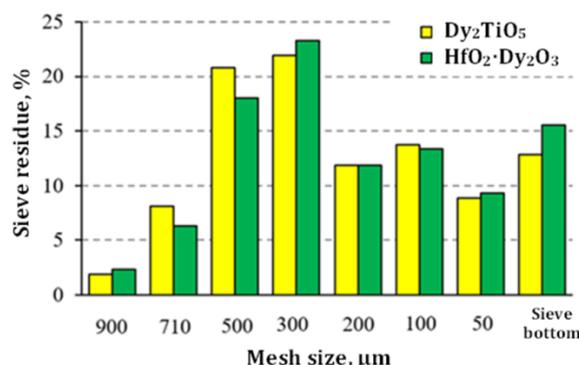


Fig. 1. The particle size distribution of dysprosium titanate and hafnate powders

The technological scheme for the manufacture of dysprosium titanate and hafnate powders is based on the grinding of sintered briquettes of synthesized absorbing materials Dy_2TiO_5 and $HfO_2 \cdot Dy_2O_3$ with subsequent measurement and adjustment of the particle size distribution by sieving the powders (Fig. 1).

Table 1
Powder compositions of dysprosium titanate (hafnate) and boron carbide

No. of composition	Material	Volume fraction, %	Mass fraction, %
1	Dy_2TiO_5	10	24.5
	B_4C (-1 + 0.315) mm	80	75.5
	B_4C (F240)	20	
2	Dy_2TiO_5	30	55.6
	B_4C (-1 + 0.315) mm	70	44.4
3	Dy_2TiO_5	40	66.0
	B_4C (-1 + 0.315) mm	60	34.0
4	$HfO_2 \cdot Dy_2O_3$	10	27.2
	B_4C (-1+0.315) mm	80	72.8
	B_4C (F240)	20	
5	$HfO_2 \cdot Dy_2O_3$	30	59.0
	B_4C (-1 + 0.315) mm	70	41.0
6	$HfO_2 \cdot Dy_2O_3$	40	77.1
	B_4C (-1 + 0.315) mm	60	22.9

70 vol.% B_4C + 30%vol. $HfO_2 \cdot Dy_2O_3$



60 vol.% B_4C + 40%vol. $HfO_2 \cdot Dy_2O_3$



Fig. 2. Combined powder mixtures of dysprosium hafnate and boron carbide

The particle densities of the powders measured by the pycnometric method [7] were 7.23 and 8.47 g/cm³ for dysprosium titanate and dysprosium hafnate, respec-

tively. Combined powder mixtures were formed from the obtained powders of dysprosium titanate and dysprosium hafnate, as well as boron carbide. The characteristics are given in Table 1 and the appearance in Fig. 2.

The study of the vibration compaction process and the characteristics of backfills of combined absorbing materials was carried out on "short" tubes made of stainless steel 08Cr18Ni10Ti, 1 m long with an internal diameter of Ø7.0 mm and full-length absorbing rods, which were assembled claddings (a cladding with a welded lower end plug – a cone) with a length of ~ 4095 mm. Vibration compaction was carried out on the VEDS-100 MK vibration machine at the following modes: oscillation frequency was 120 Hz, type of output signal was sine, vibration acceleration was 125 m/s².

2. RESULTS AND DISCUSSION

The average values of the bulk density of absorbing material powders with a fine fraction in the range of 10...50 % vol. are given in Tables 2, 3, and 4.

Table 2
Bulk density and backfill density of boron carbide powder mixtures

Fractions ratio (-1+0.315) mm/ F240 (-0.06) mm	Bulk density, g/cm ³	Backfill density, g/cm ³
50/50	1.29	1.70
55/45	1.31	1.77
60/40	1.32	1.82
65/35	1.32	1.82
70/30	1.32	1.81
75/25	1.31	1.77
80/20	1.29	1.70

Table 3
Bulk density and backfill density of dysprosium titanate powder mixtures

Fractions ratio (-1+0.1) mm/ (-0.1) mm	Bulk density, g/cm ³	Backfill density, g/cm ³
50/50	3.5	4.69
60/40	3.62	4.97
70/30	4.0	5.1
80/20	3.96	5.03
90/10	3.85	4.68

Table 4
Bulk density and backfill density of powder mixtures of dysprosium hafnate

Fractions ratio (-1+0.1) mm/ (-0.1) mm	Bulk density, g/cm ³	Backfill density, g/cm ³
50/50	4.63	5.48
60/40	4.69	5.79
70/30	4.73	5.82
80/20	4.56	5.78
90/10	4.46	5.74

It has been established that the highest bulk density of ~ 1.3 g/cm³ and the backfill density of ~ 1.82 g/cm³ for boron carbide powders in the absorbing rod cladding

are provided at the coarse fraction content of (0.9+0.315) mm in the range of 60...70% (Fig. 3).

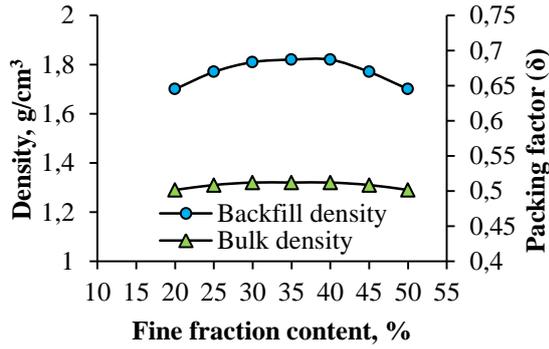


Fig. 3. Dependence of bulk density, backfill density and packing factor (δ) of B_4C powder on the content of fine fraction F240

A similar trend is observed for dysprosium titanate and dysprosium hafnate powders. Thus, the maximum bulk density and backfill density for these materials were observed in the fractional composition of (-0.9+0.1) mm – 70% and (-0.1) mm – 30%.

These values for dysprosium titanate powder were 4.0 and 5.09 g/cm³, respectively (see Table 3).

The maximum values of bulk density and backfill density for dysprosium hafnate powder were 4.73 and 5.82 g/cm³, respectively (see Table 4).

The results of the study of B₄C powder compaction in full-length absorbing rods at varying the particle size distribution, type and frequencies of vibrations generated by the vibration machine are shown in Figs. 4–6.

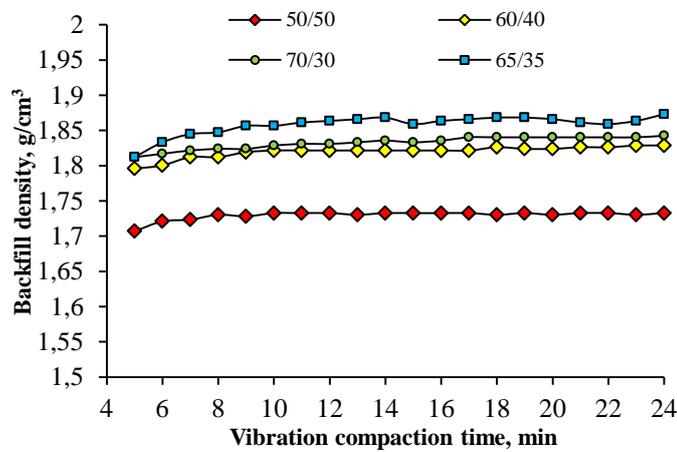


Fig. 4. Dependence of backfill density B_4C on the ratio of fractions in the mixture

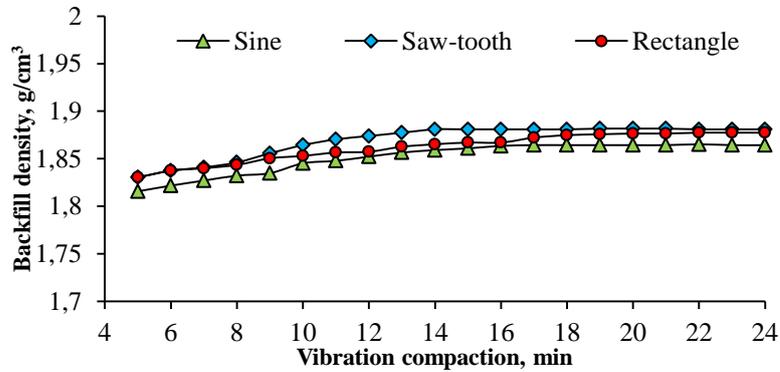


Fig. 5. Dependence of B_4C backfill density on the oscillation type (fraction ratio 65/35%)

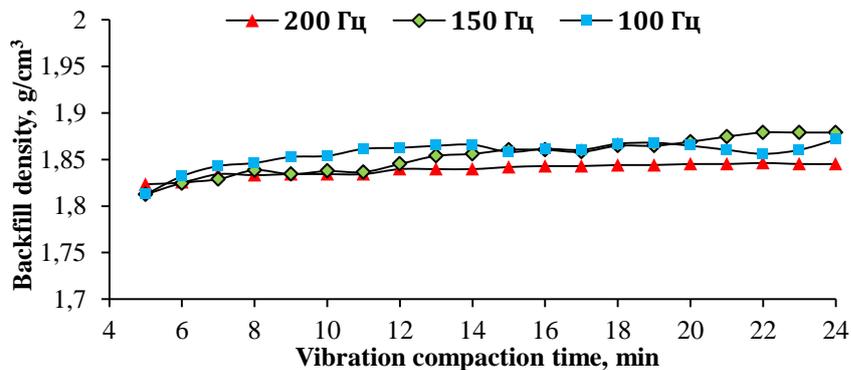


Fig. 6. Dependence of B_4C backfill density on the oscillation frequency (fraction ratio 65/35%)

It has been established that the optimal ratios between the coarse and fine fractions of boron carbide powder are 60...70 and 30...40%, respectively, for the manufacture of full-size absorbing rods. The mode of vibration compaction was the following: sinusoidal type of oscillations and a frequency of 100^{+20} Hz. It ensured the achievement of the required value of $> 1.8 \text{ g/cm}^3$ of the B_4C backfill density in the absorbing rod claddings.

The results of study into the compaction of Dy_2TiO_5 powder in the absorbing rod claddings with a design backfill height of 300 mm under varying particle size distributions (the ratio of coarse to fine fractions) are shown in Fig. 7.

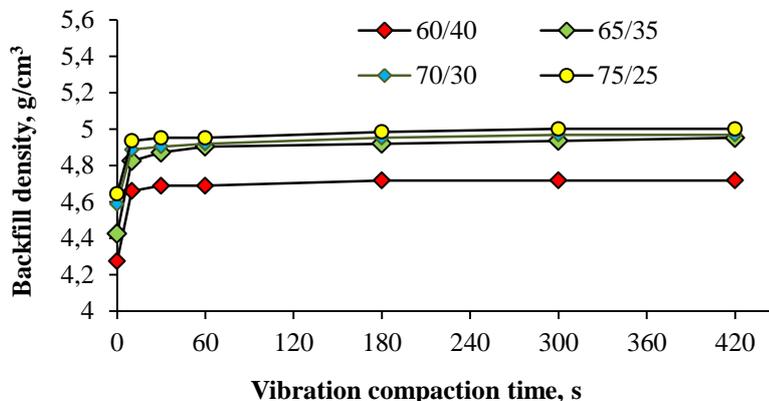


Fig. 7. Dependence of the density of dysprosium titanate powder of different particle size distributions on the time of vibration compaction

It has been established that to obtain a backfill density of at least 4.9 g/cm^3 , the ratio between coarse and fine fractions of dysprosium titanate powder when using two fractions of $(-0.9+0.1) \text{ mm}$ and of $(-0.1) \text{ mm}$ should be 60...75 and 25...40%, respectively.

The backfill density increases to almost 5.2 g/cm^3 when using three components of the powder composition: $(-0.9+0.1) \text{ mm} - 67\%$, $(-0.1) \text{ mm} - 24\%$, and $(-0.05) \text{ mm} - 7...9\%$.

The process of vibrocompaction of dysprosium titanate powder weighing about 63 g, designed for a column height of 300 mm with the vibration machine turned on, is quite fast under optimal operating conditions. The backfill density increases to 4.9 g/cm^3

in the first 10...20 s, and then increases slightly and slowly after the powder flows into the absorbing rod cladding.

Thus, the process of vibration backfilling of dysprosium titanate powder causes an increase in the backfill density from the bulk density of 4.0 to 4.9 g/cm^3 , which is about 20%.

The results of preliminary studies of the backfill characteristics of the dysprosium titanate + boron carbide and dysprosium hafnate + boron carbide combined mixtures carried out in "short" claddings 1 m long with an internal diameter of $\text{Ø}7.0 \text{ mm}$ are presented in Tables 5 and 6, respectively.

Table 5

Characteristics of combined powder fills of dysprosium titanate and boron carbide

No. of mixture	Combined powder composition	Bulk density, g/cm^3	Backfill density, g/cm^3	Max. density of the composition, g/cm^3	Packaging efficiency
1	70% vol. B_4C $(-1+0.315) \text{ mm}$ + 20% vol. B_4C (F240)+ 10% vol. Dy_2TiO_5	1.55	2.24	2.98	0.75
2	70% vol. B_4C +30% vol. Dy_2TiO_5	1.89	2.96	3.94	0.74
3	60% vol. B_4C +40% vol. Dy_2TiO_5	2.11	3.36	4.46	0.75

Table 6

Characteristics of combined powder fills of dysprosium hafnate and boron carbide

No. of mixture	Combined powder composition	Bulk density, g/cm^3	Backfill density, g/cm^3	Max. density of the composition, g/cm^3	Packaging efficiency
4	70% vol. B_4C $(-1+0.315) \text{ mm}$ + 20% vol. B_4C (F240) + 10% vol. $\text{HfO}_2 \cdot \text{Dy}_2\text{O}_3$	1.6	2.44	3.1	0.79
5	70% vol. B_4C +30% vol. $\text{HfO}_2 \cdot \text{Dy}_2\text{O}_3$	2.0	3.08	4.27	0.72
6	60% vol. B_4C +40% vol. $\text{HfO}_2 \cdot \text{Dy}_2\text{O}_3$	2.3	3.69	5.45	0.68

The study of the vibration compaction process and the characteristics of backfills of combined absorbing materials was also carried out on full-size absorbing

rods. The general characteristics of backfills are given in Table 7.

Table 7

Characteristics of backfills of combined mixtures in full-size absorbing rods

No. of mixture	d , mm	Dy ₂ TiO ₅ powder weight, g	B ₄ C powder weight, g	Dy ₂ O ₃ ·TiO ₂		Backfill density B ₄ C, g/cm ³	Total length of the column, mm
				Column height, mm	Backfill density, g/cm ³		
1	7.01	27.2	226	365	1.93	1.84	3551.0
2	7.00	34.8	223.9	339	2.66	1.84	3509.5
3	7.00	40.4	224.1	336	3.12	1.83	3512.0
4	6.98	27.9	222.6	330	2.19	1.83	3513.0
5	6.99	35.2	223.6	295	3.09	1.81	3509.0
6	6.99	42.6	223.3	306	3.61	1.82	3508.0

The study of the kinetics of vibrocompaction of combined powder compositions revealed that the time to reach the maximum density is at least 90 s. This maximum value can be characterized by the values of packing factor in the range from 0.68 to 0.79 and calculated basing on the maximum density of the combined mixture of dysprosium titanate (dysprosium hafnate) and boron carbide, which in turn depends on the volume (mass fraction) and density of each of the mixture components.

Studies have revealed that the packing factor, defined as the ratio of the backfill density to the compact material density (theoretical material density), did not depend on the type of materials in the combined powder compositions, but was determined by the granulometric composition of the backfill (the content of the fine fraction).

It was found that an increase in the vibration frequency to 250 Hz leads to an intensification of the vibration compaction process and an increase in the density of backfill by about 3...5%. The installation of a weighting rod also leads to an increase in backfill density. Analyzing the data obtained, it should also be noted that the backfill density of combined powder mixtures in full-length absorbing rods is 2...10% lower compared to the backfill density in "short" tubes. This phenomenon may be related to the possible redistribution of coarse fraction (boron carbide) and fine fraction (dysprosium titanate or dysprosium hafnate) particles during backfilling, moving or falling from heights of more than 4 m.

CONCLUSIONS

1. The influence of the granulometric compositions of boron carbide, dysprosium titanate, and dysprosium hafnate powders and their mixtures in the initial state on the characteristics of backfills in the claddings of both "short" and full-size absorbing rods was researched.

2. It has been established that the highest bulk density of ~ 1.3 g/cm³ and the backfill density in the absorbing rod cladding of ~ 1.82 g/cm³ are provided at the content of a coarse fraction of (0.9+0.315) mm in the range of 60...70% for boron carbide powders.

3. The maximum values of bulk density and backfill density were 4.0 and 5.09 g/cm³, respectively, for dysprosium titanate powder.

4. The maximum values of bulk density and backfill density were 4.73 and 5.82 g/cm³, respectively, for dysprosium hafnate powder.

5. Vibrocompaction modes (type and frequency of oscillations) are set to achieve the required value of the absorbing rod backfill density: sinusoidal type of oscillations, frequency of 100^{±20} Hz.

REFERENCES

1. V.D. Risovany, A.V. Zakharov, E.M. Muraleva. New advanced absorbing materials for thermal reactors // *Problems of Atomic Science and Technology. Series "Physics of Radiation Effect and Radiation Materials Science"*. 2005, N 3(86), p. 87-93.
2. A.V. Zakharov, V.D. Risovany, E.P. Klochkov, D.N. Suslov, O.G. Sidorenko, et al. *Absorbing core of a nuclear reactor control rod*: RF patent No. 2119199 of 20.09.1998, G21C7/10, G21C7/24.
3. I.G. Shatalova, N.S. Gorbunov, V.I. Lichtman. *Physical and chemical bases of vibration compaction of powder materials*. M.: "Nauka", 1965.
4. A.A. Maershin. *Fuel elements with vibrocompacted oxide fuel*. Dimitrovgrad: SSC "NIIAR", 2007, 327 p.
5. R. Herbig, K. Rudolf, A. Maershin. Vibrocompacted fuel for the liquid metal reactor BOR-60 // *Journal of Nuclear Materials*. 1993, v. 204, p. 93-101.
6. I.I. Loktev. Model of packing of polydisperse materials // *Proceedings of the Seventh Russian Conference on Reactor Materials Science* (Dimitrovgrad, September 8-12, 2003). Dimitrovgrad: SSC RF NIIAR, 2004, v. 2, p. 153-165.
7. GOST 22662-77. *Metal powders. Methods of sedimentation analysis. Methods of sedimentation analysis*.

Article received 07.06.2023

ДОСЛІДЖЕННЯ ВПЛИВУ ГРАНУЛОМЕТРИЧНИХ СКЛАДІВ ПОРОШКІВ НЕЙТРОНОПОГЛИНАЮЧИХ МАТЕРІАЛІВ НА ЇХ УЩІЛЬНЮВАНІСТЬ В ОБОЛОНКАХ ПЕЛ

І.О. Чернов, М.М. Бєлаш, В.О. Романьков, В.М. Євсєєв, Є.С. Легенький

Представлено результати досліджень впливу гранулометричних складів порошоків нейтронопоглинаючих матеріалів: карбїду бору, титанату диспрозію і гафнату диспрозію та їх сумішей у вихідному стані на характеристики засипок у оболонках як «коротких», так і повнометражних пел. Встановлено, що досягненню максимальної щільності засипки сприяє забезпечення вмісту дрібної фракції біля 30%. Досягнуті максимальні щільності засипок карбїду бору, титанату диспрозію і гафнату диспрозію відповідно: 1,82; 5,1 і 5,82 г/см³. Встановлено оптимальні режими віброущільнення (тип та частота коливань), що забезпечують досягнення необхідних значень щільності засипок пел за короткий час.