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RESEARCH ARTICLE

Development of an encapsulation process for toxic waste and hazardous chemicals in a fluidized bed

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Abstract

Objectives. *This paper presents research results on the encapsulation of a fluidized bed of liquid and solid toxic waste containing chemicals with a hazard class of 1–3.*

Methods. *Soils contaminated with hexachlorobenzene and hexachlorocyclohexane were used as the seed material. Ceresin was selected as the encapsulant, which was sprayed onto the fluidized bed through a pneumatic nozzle at a temperature of 135°C. Before the spraying of the ceresin, binders were introduced into the fluidized bed of the seed material through pneumatic nozzles in the form of a melt of high-temperature coal-tar pitch and wastewater containing sodium and arsenic salts as well as heavy metal oxides. The experiments were carried out using a modified GLATT AGT-150 laboratory unit.*

Results. *The results demonstrate that the mechanism for granule formation is a mixed mechanism. The binding of the seed material is carried out by both the pitch and salting out. In this case, the cavities in the agglomerates are partially filled with salt deposits, which increases the strength and integrity of the final product's structure. Ranges for the process parameter values were established at the point at which there was no unwanted agglomeration in the fluidized bed, and dust formation did not exceed 5%. When the ratio of the bed mass to the mass of ceresin is equal to unity, a moisture-resistant free-flowing product of hazard class 5 is obtained, which is suitable for transportation and long-term storage. The average diameters of the initial particles and encapsulated granules were 0.5 and 1.5 mm, respectively.*

Conclusions. *The present study demonstrates a potential process for the granulation–encapsulation of toxic waste and hazardous substances with a hazard class of 1–3 in a single fluid-bed apparatus, resulting in the formation of a moisture-resistant hazard class-5 granular product suitable for transportation and long-term storage. The results obtained can be used in the development of an industrial large-scale process for encapsulating waste of hazard classes 1–3.*

Keywords: *persistent organic pollutants, heavy metals, toxic wastes, wastewater, encapsulation, coal-tar pitch, ceresin*

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НАУЧНАЯ СТАТЬЯ

Разработка процесса инкапсуляции токсичных отходов и опасных химических веществ в псевдоожиженном слое

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Аннотация

Цели. *Исследование процесса инкапсуляции в условиях псевдоожиженного слоя жидких и твердых токсичных отходов, содержащих химические вещества 1–3 классов опасности.*
Методы. *Для исследований в качестве затравочного материала использовался грунт, загрязненный гексахлорбензолом и гексахлорциклогексаном. Инкапсулянтом выступал церезин, который при температуре 135 °C распылялся в псевдоожиженный слой через пневматическую форсунку. Эксперименты осуществлялись на модифицированной лабораторной установке GLATT AGT-150. Перед распылением церезина в грунт через пневматические форсунки вводились связующие – плав высокотемпературного каменноугольного пека и сточные воды, содержащие соли натрия, мышьяка, а также оксиды тяжелых металлов.*
Результаты. *Показано, что механизм гранулообразования носит смешанный характер. Связывание исходных частиц загрязненного грунта осуществляется как пеком, так и за счет высаливания. При этом полости в агломерате частично заполняются отложениями солей, что увеличивает прочность и целостность структуры конечного продукта. Установлены диапазоны значений управляющих параметров процесса, при которых в слое отсутствовала нежелательная агломерация, а пылеобразование не превышало 5%. При отношении массы слоя частиц (гранул) к массе поданного церезина, равном единице, получен влагоустойчивый сыпучий пригодный для транспортировки и длительного хранения продукт 5 класса опасности. Средние диаметры исходных частиц и капсулированных гранул соответственно составляют 0.5 мм и 1.5 мм.*
Выводы. *Выполненные исследования показали принципиальную возможность проведения в одном аппарате процесса сушки-грануляции-капсулирования токсичных отходов и опасных химических веществ 1–3 классов опасности с образованием сыпучего продукта 5 класса опасности, обладающего влагоустойчивостью, а также пригодного для транспортировки и длительного хранения. Полученные результаты могут быть использованы при разработке промышленного крупнотоннажного процесса инкапсуляции отходов 1–3 классов опасности.*

Ключевые слова: *стойкие органические загрязнители, тяжелые металлы, токсичные отходы, сточные воды, инкапсуляция, каменноугольный пек, церезин*

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INTRODUCTION

According to the official data of the Ministry of Natural Resources and Environment of the Russian Federation, there are currently more than 200 sites of accumulated environmental damage in Russia where toxic substances are stored or buried, including substances of hazard class 1 or 2 (in particular, persistent organic pollutants, POPs). The total volume of these substances is estimated to be in the millions of tons. It is also known that there are numerous unauthorized dumps and burial sites containing industrial waste.[1,2]

For the destruction of toxic substances classified as hazard class 1 or 2, the thermal high-temperature neutralization method has gained the greatest global popularity. Despite this method's high level of efficiency, its main drawback is the need to create an expensive gas cleaning system. At the same time, it is necessary to address the issue of wastewater disposal associated with this method [1, 2].

At the regional level, as a rule, the options considered for making relatively small landfill sites containing toxic substances/waste safe are either to establish a landfill for the appropriate hazard class in the immediate vicinity of the general-waste landfill site or to transfer the hazardous chemicals/waste to a specialized organization for further processing, disposal, or dumping.

The primary objective, both during the long-term storage and transportation of toxic waste, hazardous chemicals, and wastewater, is to minimize the likelihood of their penetration into the environment.

One of the industrial methods used for this purpose is encapsulation in a solid matrix. Concrete and bitumen are usually used as encapsulants [3]. A common disadvantage of both matrices is a significant increase in the weight and volume of the encapsulated waste as well as the observed leaching of toxic contents, the percentage of which depends both on the composition of the encapsulated substances and the encapsulator as well as on the storage conditions [4]. For example, the leaching of arsenic from concreted arsenic-containing waste over a period of 406 days under constant water exposure (at 10% of the maximum achievable arsenic content in the concrete matrix) has been calculated at 0.34% [5, 6]. In addition, experiments on the bituminization of ash from the incineration of toxic waste have demonstrated the leaching of heavy metals over 90 days at a level of 0.1–0.3%; the heavy metal concentration in the ash was 5–11 mg/kg and the ash content of the bitumen was up to 60% by weight [7]. The leaching of polycyclic aromatic hydrocarbons directly from bitumen can reach 1×10^{-4} mg/L over 64 days [8].

In the present study, ceresin, widely used as an insulating material in radio and electrical engineering, was proposed as an alternative encapsulant. Ceresin is a solid under normal conditions and is a water-insoluble and environmentally inert mixture of marginal hydrocarbons. Due to its relatively low melting point, density, and viscosity, ceresin can be easily sprayed through a mechanical or pneumatic nozzle into a fluidized bed of solid material to form a film on its surface.

To reduce the consumption of ceresin, the possibility of increasing the size of fluidized particles through granulation was investigated, which, in turn, would significantly reduce the total area of the encapsulated surface. Two mixtures were selected as binders: a high-temperature coal-tar and wastewater containing heavy metals and soluble sodium salts (including arsenite). This selection is based on the fact that these binders are also disposable and recyclable as waste, and therefore their use as raw materials is economically justified.

Thus, the aim of this study was to develop a process for encapsulation of toxic waste and hazardous chemicals using solid hydrocarbon waste and wastewater containing heavy metals and soluble salts as a binder in a product suitable for transportation and long-term storage without significantly increasing the initial weight and volume.

This study focused on hazard class-2 POP-contaminated soil removed from the Bolshie Izbishchi landfill site (Lipetsk oblast, Russia) and a hazard class-3 reaction mass, obtained by laboratory means from the wet cleaning stage of flue gases during the research of the thermal neutralization of hazard class-2 sludge sampled from the territory of the former *Srednevolzhsky Chemical Plant* (Chapaevsk, Russia).

The hazard class was determined using bioassay methods on hydrobionts in accordance with R 52.24.566-94.[3]

[1] Ministry of Natural Resources of Russia: The state register of objects of accumulated environmental damage (accessed April 13, 2021) (in Russ.). https://www.mnr.gov.ru/docs/docs/svedeniya_soderzhashchiesya_v_gosudarstvennom_reestre_obektov_nakoplennogo_vreda_okruzhayushchey_sr/?special_version=Y

[2] Rosprirodnadzor. Rosprirodnadzor reports on illegal landfills. Russia, Moscow; 2019 (published July 09, 2020, accessed Oct 8, 2020) (in Russ.). https://rpn.gov.ru/news/156/

[3] R 52.24.566-94. Recommendations. Methods for toxicological assessment of pollution of freshwater ecosystems. St. Petersburg: Gidrometeoizdat; 1994. 136 p.

MATERIALS AND METHODS

The composition of the POP-contaminated soil, sampled from the Bolshie Izbishchi landfill site, is presented in Table 1.

Table 1. The composition of the contaminated soil*

Component name	mass %
Water	17.45
Inert filler (talc, pyrophyllite, kaolin)	15.37
Hexachlorobenzene	0.004
Hexachlorocyclohexane (mixture of isomers including lindane)**	0.002
Soil	67.18
Total:	100.00

* Average particle diameter 0.5–0.6 mm.
** Lindane is classified as hazard class 1.

The wastewater composition is shown in Table 2.

High-temperature coal-tar with a melting point of ~116–120°C (CAS no. 65996-93-2)[4] was used as a binder to model the properties of waste from the production of coke and coal-tar pitch. Before use, the pitch was crushed in a laboratory ball mill to a particle size of less than 200 μm. The encapsulant was ceresin-75 (according to GOST 2488-79)[5], and air was used as a fluidizing agent. A schematic diagram of the laboratory unit developed for this study, based on GLATT AGT-150 (*Glatt GmbH*, Germany), is shown in Fig. 1.

The main geometric parameters of the installation are as follows: the diameter of the working chamber is 160 mm, the height of the cylindrical part of the working chamber is 450 mm, and the area of the gas distribution grid is 0.02 m^2.

The pitch spray nozzle (7) is installed at a height of 335 mm above the gas distribution grid (5), and the pneumatic spray nozzle for wastewater and ceresin (6) is installed at a height of 40 mm (nozzle direction up).

The design of the unit provides the ability for the selective discharge of the product into the collector (4)

Table 2. Wastewater composition

Component name	mass %
H_2O	77.55
PbO	0.02
ZnO	0.01
CdO	0.003
CoO	0.001
CuO	0.02
As_2O_3	0.02
Na_2CO_3	0.79
NaCl	20.69
Na_2SO_4	0.50
$NaNO_2$	0.33
Na_3AsO_3	0.07
Total:	100.00

through a sifter (3), in which the particles are segregated by weight by controlling the flow rate of the ascending air flow. The area of the slotted gap for the supply of air to the nozzles is 6.28×10^{-6} m^2.

The unit has a control panel for setting and controlling the operating parameters: the flow rate of the "processed" air and its temperature, the air pressure in the injectors, the flow rate and temperature of the supplied substances, and the pressure in the sifter of the finished product.

The laboratory unit's technological and design parameters were determined as the following:

– wastewater and ceresin consumption (G_{ww}, G_c): 0.6–3.0 kg/h;
– fluidizing agent consumption (G_{FA}): 40–400 m^3/h;
– pitch consumption (G_p): 0.5 kg/h;
– fluidizing agent temperature (T_{FA}): up to 260°C;
– spray air pressure in the injectors (P_{AA}): 1.4–3.6 bar;
– diameter of the spray nozzle of the injectors: 0.5 mm.

The experimental procedure is as follows. A specified amount of contaminated soil is loaded into the drying chamber (8). Then, using a fan (10) in the drying chamber (8), a fluidizing agent (air) of a specified temperature is introduced through the unit's

[4] CAS No. 65996-93-2. Coal-tar pitch, high temperature. Summary risk assessment report (accessed April 13, 2021). URL: https://echa.europa.eu/documents/10162/13630/trd_rar_env_netherlands_pitch_en.pdf
[5] GOST 2488-79. Geresin. Specifications. Moscow: Izd-vo standartov; 1980. 6 p.

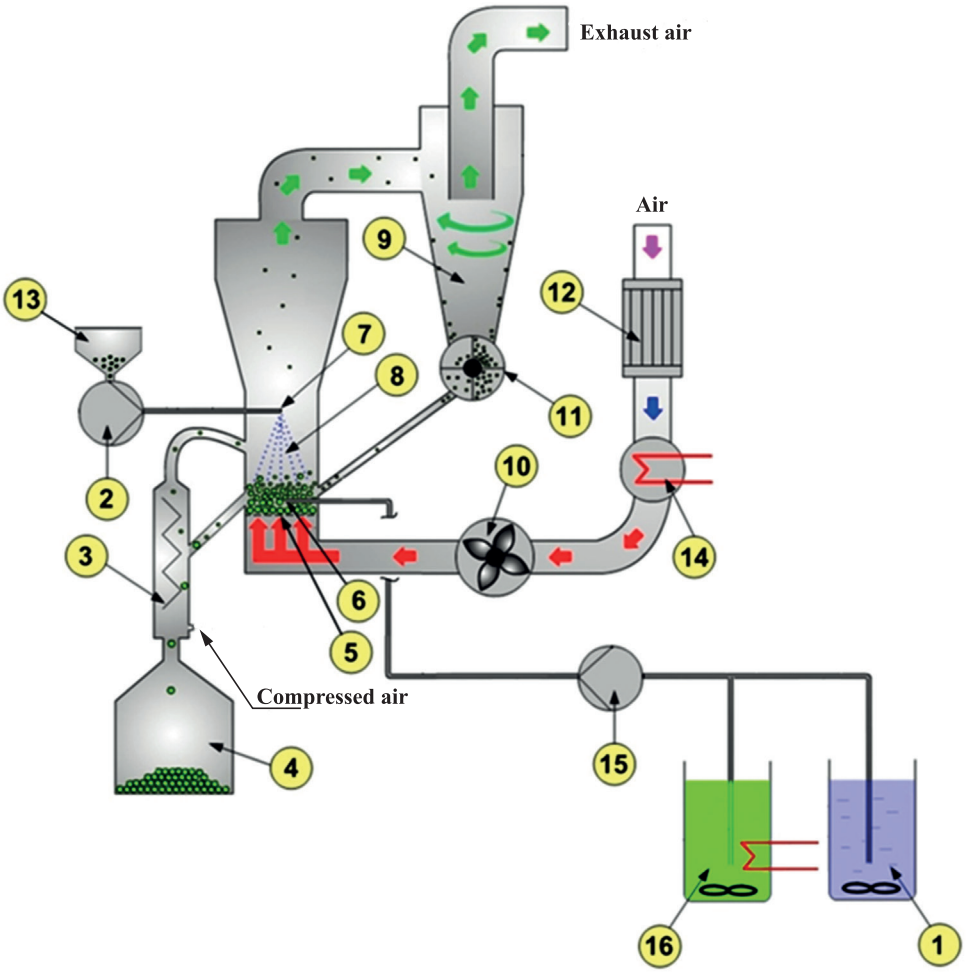


Fig. 1. Schematic diagram of the laboratory unit:
(1) tank with wastewater; (2) screw dispenser; (3) sifter; (4) product collector; (5) gas distribution grid; (6) nozzle for spraying wastewater and ceresin; (7) pitch feed nozzle; (8) drying-encapsulation chamber; (9) cyclone; (10) fan; (11) drum gateway; (12) refrigerator; (13) bunker with crushed pitch; (14) heater; (15) pump; (16) vessel with ceresin melt.

gas distribution grid (5), changing the contaminated soil/granulate bed into a fluidized state.

Wastewater (1), which is continuously agitated, is sprayed by a gerotor pump (15) through the pneumatic nozzle (6) into the drying chamber (8), where the moisture evaporates and the solids settle on the surface of the contaminated soil. At the same time, crushed pitch is fed from the hopper (1) via a screw dispenser (2) to the working chamber of the pneumatic nozzle (7). This chamber (7) is equipped with an electric heating element that converts the powdered pitch into a molten state (temperature ~200–210°C). The pitch melt is then fed from the working chamber to the nozzle for spraying using an integrated screw pump.

The exhaust air ascends to the top of the device and, after additional dust separation in the cyclone (9), is discharged into the supply and exhaust ventilation system. The fine particles separated in the cyclone are continuously discharged through the cellular rotary valve (11) back to the working area of the machine.

After the accumulation of granules of a given size in the bed, the temperature of the fluidizing agent is reduced to room temperature, and by means of a pneumatic nozzle (6), the molten ceresin is fed from the heated container (16) at a temperature of 135°C with constant stirring. As the molten ceresin is sprayed onto the fluidized bed, the previously formed granules form capsules covered with an inert solid shell.

The operating time of the single experiment is 1 h. At the end of the experiment, the unit is cooled to room temperature and the product is discharged from the drying chamber (8).

The experiments were carried out in three stages:

Stage 1: determination of the operating parameters of the dry granulation of wastewater on the surface of the contaminated soil,

Stage 2: determination of the operating parameters of the dry granulation of wastewater on the surface of the contaminated soil with the simultaneous spraying of pitch, and

Stage 3: determination of the operating parameters of the ceresin encapsulation of the granules obtained in Stage 2.

The resulting product was subjected to a sieve analysis. In addition, at Stage 3, the product was tested for moisture resistance by placing it in water (volume 2 L) at room temperature for 96 h and performing a chemical analysis on the extract, and biotesting on hydrobionts was conducted to determine the product's hazard class.

RESULTS AND DISCUSSION

The flow rate of the fluidizing agent was estimated using the calculation method described by N.A. Shakhova [9]. Thus, the minimum flow rate of the fluidizing agent at a temperature of 100°C, which provides fluidization of particles in the size range d_p = 0.5 mm (density 2165 kg/m^3), was 65 m^3/h (operating speed w_{FA} = 0.9 m/s).

It is known [10] that the values of the working speed of w_{FA} in the range of 0.8–1.4 m/s are routinely related to drying in a fluidized bed, whereas, in dry granulation, the speed of the fluidizing agent is usually 1.0–2.0 m/s. Therefore, it was also advisable to use the fluidizing agent flow rate of 110 m^3/h (w_{FA} = 1.53 m/s) during the experiments.

The calculation of the mass of layer M, which contains the particles d_p = 0.5 mm located in the nozzle flare zone, and the maximum height of layer l, which should not exceed the height of the working chamber (0.45 m), under conditions in which the operating speed of the fluidizing agent changes from 0.9 m/s to 1.53 m/s (a flow rate change from 65 m^3/h to 110 m^3/h), was also carried out according to the Shakhova method [9, 11].The results of the mass calculation are presented graphically in Fig. 2.

Based on the results of the calculated data presented in Fig. 2, we assumed that the amount of contaminated soil used as a seed with d_p = 0.5 mm can be in the range of M = 0.5–2.4 kg (w_{FA} = 0.9 m/s) and M = 0.4–1.7 kg (w_{FA} = 1.53 m/s).

The temperature of the fluidizing agent and the wastewater flow rate are among the main control parameters that regulate the moisture content in the layer and affect the nature of the interaction of the particles with each other [12].

The calculation of the dependence of the temperature of the fluidizing agent T_{FA} on the wastewater flow rate G_{ww} (in the range of 0.6–3.0 kg/h) while maintaining a constant temperature in the bed of 100°C (the highest moisture intake during drying) and fluidizing agent G_{FA} flow rates of 65 m^3/h and 110 m^3/h was carried out according to the heat

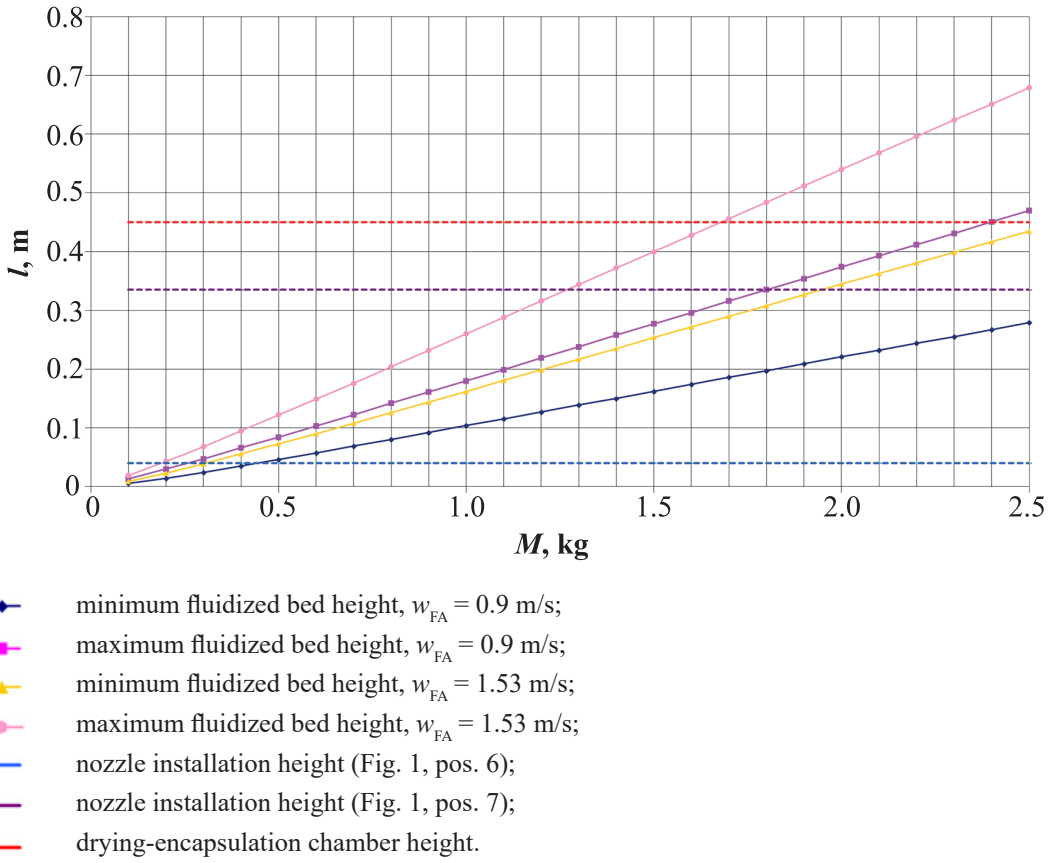


Fig. 2. Calculated dependence of the fluidized bed height on its mass (d_p = 0.5 mm).

balance equation given in P.G. Smith's monograph [13]. The values presented in Table 3 were used for the calculation.

The results of this calculation in graphical form are shown in Fig. 3.

As can be seen in the graph, with a fluidizing agent flow rate of 110 m^3/h, the potential capacity of the wastewater drying plant is significantly higher. However, it should be noted that an increase in the speed of the fluidizing agent increases the probability of particle abrasion, which, in turn, results in an increase in dust formation.

Thus, based on the calculated data, the initial temperature of the fluidizing agent in the laboratory experiments was T_{FA} = 118.5°C. The effect of pitch supply on the heat content of the layer was not taken into account since the main influence on the temperature of the latter is the amount of water supplied (and evaporated). In the ceresin spray mode, the fluidizing agent heating was turned off, and its temperature corresponded to room temperature.

The average diameter of wastewater droplets was calculated using the empirical K. Masters ratio [11, 14]. Recommended by Masters for pneumatic injectors, the "flow rate of the spraying agent/flow rate of the sprayed liquid" ratio is in the range of 0.1–10. In addition, the size of the droplets d_d, which is determined by this ratio, should be at least 10 times

Table 3. Values of constants for calculating the temperature of the fluidizing agent T_{FA} and determining the amount of moisture removed from wastewater G_{ww}

Parameter name	Dimension	Value
Heat capacity of the fluidizing agent (at 100°C)	kJ/(kg·K)	1.01
Heat capacity of wastewater (at 20°C)	kJ/(kg·K)	2.88
Wastewater supply temperature	°C	20.0
Salinity of wastewater	–	0.23
Heat loss	%	5.0

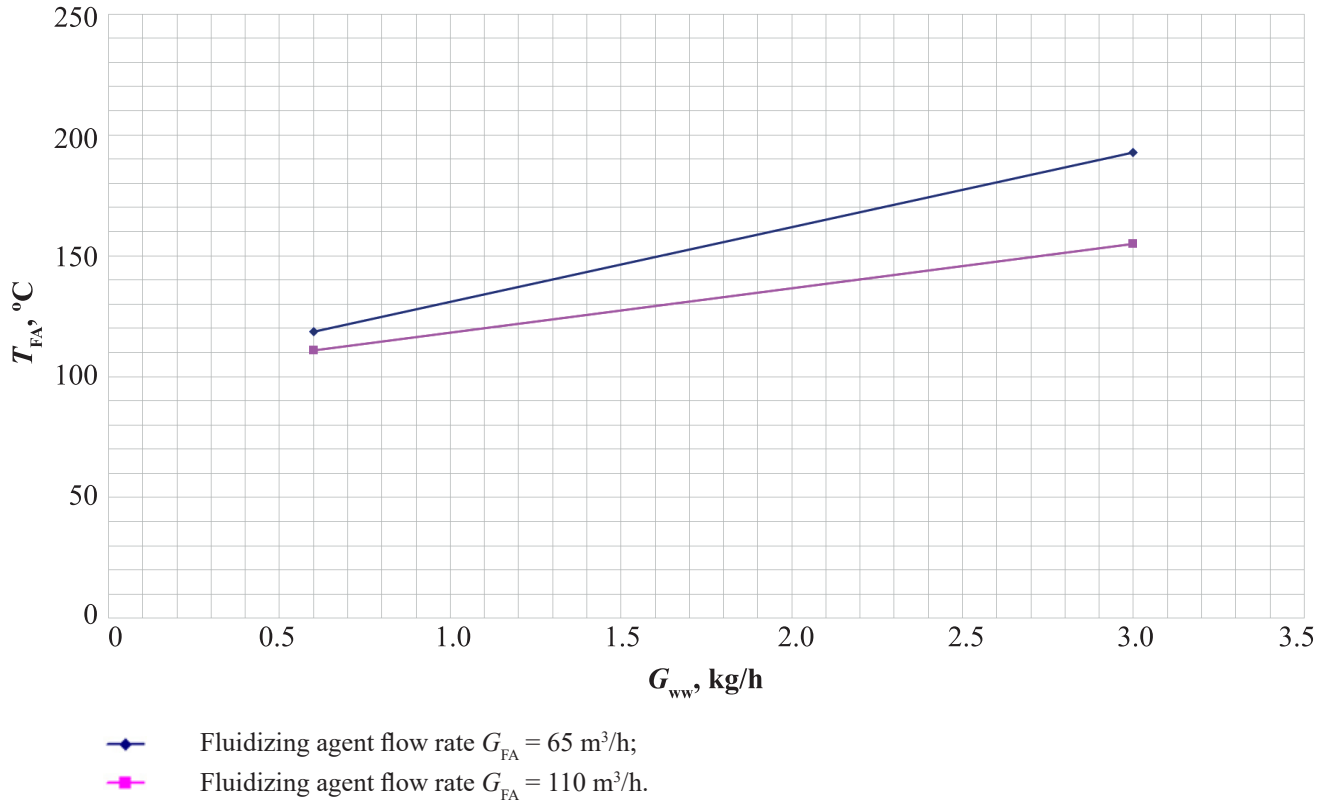


Fig. 3. Calculated dependence of the fluidizing agent temperature on the wastewater flow rate while maintaining a constant temperature in the bed 100°C.

smaller than the diameter of the particles on the surface being sprayed [15–18]. Since during the dry granulation–encapsulation process there is a constant growth of granules (i.e., the diameter of d_p increases), the maximum value of d_d was also assumed to be 0.2 mm (200 μm). The graph in Fig. 4 shows the calculated dependence of the size of the droplets of sprayed wastewater and pitch on the pressure of the atomizing agent.

Thus, the pressure range of the atomizing agent was assumed to be 1.4–2.4 bar at a wastewater flow rate of 0.6 to 3.0 kg/h and d_p = 0.5–2 mm. At d_p = 2 mm, a pressure of 1.4 bar is sufficient to spray the pitch, but, at d_p = 0.5 mm, the minimum pressure of the atomizing agent is at least 3 bar.

Taking into account the fact that the values of the physical properties of ceresin at a temperature of 135°C, which affect the nature of the droplet spraying, are comparable to wastewater [19], 1.4 bar was taken as the initial pressure value for the ceresin spray.

Tables 4, 5, and 6 set out the results of the experimental studies in which the results of the calculations are used as the initial operating parameters of the process.

In Experiment 2, dust formation was noted, which was visually observed, as well as discrepancies between the values of the expected amount of discharged granulate and the actual amount of more than 10%. An increase in the amount of seed material allowed the removal of unwanted dust formation and achieved the required performance for wastewater due to the increased contact surface between the liquid and solid phases (Experiment 3).

The values of the process parameters presented in Table 4 (Experiment 3) were used as the initial values at Stage 2 of the research.

In Experiment 4, intensive formation of large agglomerates in the bed was observed. An increase in the temperature in the bed and the pressure of the atomizing agent did not lead to a noticeable improvement in the results (Experiment 5).

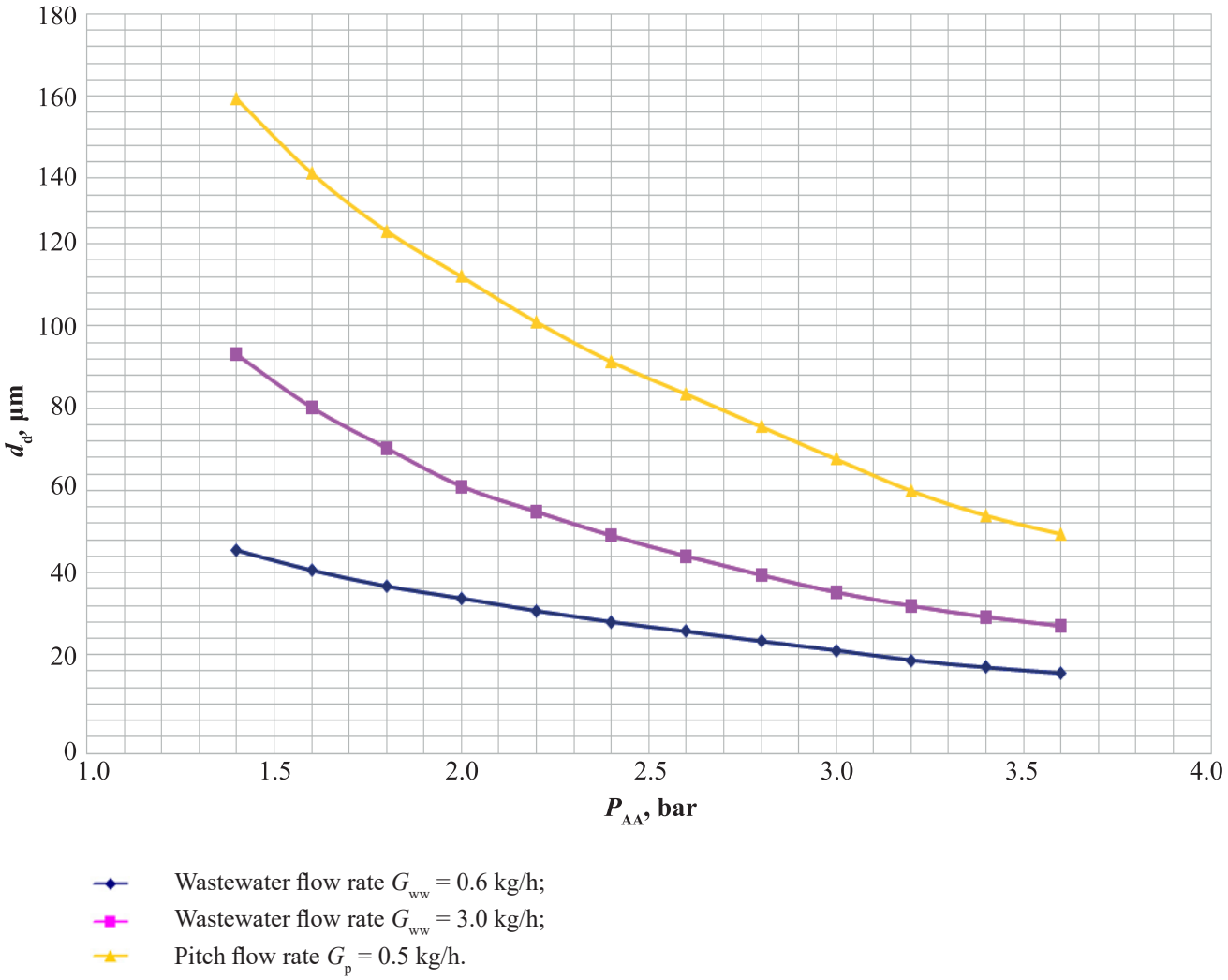


Fig. 4. Calculated dependence of the droplet size of sprayed wastewater and pitch on the pressure of the atomizing agent.

It is clear that one of the reasons for the formation of agglomerates was the insufficient velocity of the particles relative to each other, i.e., cohesive interparticle interaction forces were significantly greater than the crushing and grinding forces acting on the part of the fluidizing bed, which is probably primarily due to the sputtering of the pitch melt. In this regard, in subsequent experiments, the flow rate of the fluidizing agent was increased to 110 m^3/h.

Table 4. Summary table of the experimental results on the dry granulation of wastewater on the surface of contaminated soil (Stage 1)

No. exp.	Parameters							Notes
	M, kg	G_{FA}, m^3/h	T_{FA}, °C	T_B,** °C	T_{EA},** °C	P_{AA}, bar	G_{ww}, kg/h	
1	0.4	64–67	118–188	95–102	75–83	1.4–2.2	0.6–2.1	+*
2	0.4	63–68	187–192	90–101	77–82	2.2–2.4	2.1–3.0	Dust formation
3	0.5	61–67	185–198	88–104	76–84	2.2–2.4	1.5–3.0	+

* No unwanted dust formation and agglomeration.
** Temperature in the bed (T_B) and temperature of the exhaust gases (T_{EA}), respectively.

Table 5. Summary table of the experimental results on the dry granulation of wastewater on the surface of contaminated soil with simultaneous spraying of pitch (Stage 2)

No. exp.	Parameters									Notes
	M, kg	G_{FA}, m^3/h	T_{FA}, °C	T_B,** °C	T_{EA},** °C	$P_{AA(ww)}$*, bar	$P_{AA(p)}$*, bar	G_p, kg/h	G_{ww}, kg/h	
4	0.5	62–70	121–192	97–106	82–93	2.2–2.4	3.0–3.1	0.5	1.5–1.8	Unwanted agglomeration
5	0.5	63–65	117–190	95–103	81–90	2.4–2.8	3.3–3.6	0.5	1.5–1.8	Unwanted agglomeration
6	0.5	107–115	118–133	92–103	71–85	2.4–2.8	3.3–3.6	0.5	1.5–1.8	Dust formation
7	0.6	108–112	129–135	95–100	78–81	2.0–2.4	3.3–3.6	0.5	1.5–2.1	+
8	0.6	106–114	134–144	92–100	70–81	1.4–2.0	3.3–3.6	0.5	1.5–2.5	Dust formation
9	0.8	107–114	131–154	91–98	73–81	1.4–2.0	3.3–3.6	0.5	1.5–2.5	+
10	1.0	107–113	135–153	88–95	74–82	1.4–2.0	3.3–3.6	0.5	1.5–3.0	+
11	1.0	106–115	143–154	92–99	77–85	1.4–2.0	–	–	2.1–2.2	+
12	1.0	107–113	138–145	84–93	75–78	–	3.3–3.6	0.5	–	+

* Atomizing agent pressure for wastewater ($P_{AA(ww)}$) and pitch ($P_{AA(p)}$) spraying, respectively.
** Temperature in the bed (T_B) and temperature of the exhaust gases (T_{EA}), respectively.

In Experiment 6, the number of particles less than 0.5 mm in the unloaded product increased from 3.43 to 8.58 wt % in comparison with the experiments in Stage 1. At the same time, ~37% of the dust fraction was made up of pitch particles. It is likely that the growth of the dust fraction was associated with both the increased velocity of the fluidizing agent and with a reduction in the number of contacts between the solid particles in the bed and those sprayed with pitch (in the form of melt) due to the intensification of heat removal in the supra-bed space.

A gradual increase in the amount of contaminated soil to be loaded up to 1 kg (and, consequently, the minimum bed height), while reducing the pressure of the atomizing agent in the nozzle (Fig. 1, pos. 6) up to 1.4 bar at a wastewater flow rate of up to 3 kg/h, allowed a stable dry granulation process to be achieved without agglomerate formation (Experiment 10).

Comparative experiments were carried out feeding only wastewater into the bed (Experiment 11) and only pitch (Experiment 12). In both cases, the quantities of the loaded contaminated soil and the supplied solid fraction were 1 kg and 0.5 kg, respectively. The results of the sieve analysis of the product from Experiments 11–13 are shown in Table 6, and Fig. 5 shows the appearance of the granules. Images were taken using a Leica DM2500 microscope (*Leica Microsystems*, Germany).

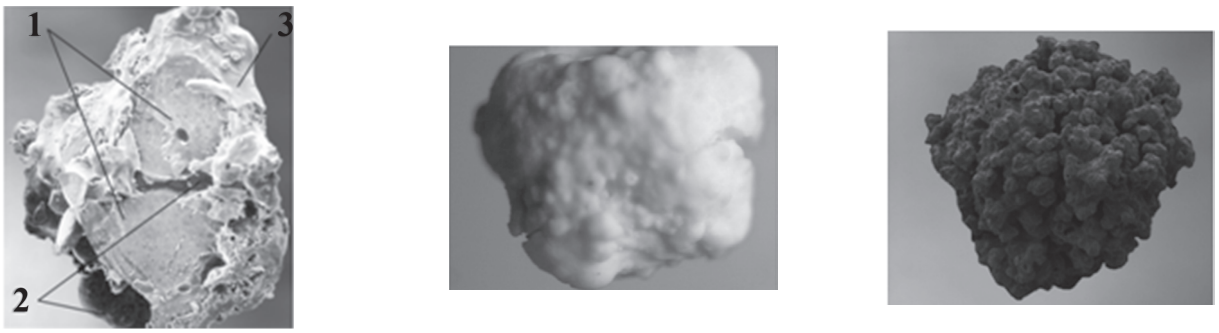
Based on the results of the analysis of the granulometric composition of the product, we assumed that the predominant mechanism for granule growth in Experiments 10 and 12 was agglomeration. This agrees with M. Hemati, *et al*. [20], who note that the growth of granules in a fluidized bed is due to agglomeration if the number of granules with a diameter two times larger than the diameter of the initial particles is more than 15% of the total number of particles in the bed during the process for 1 h.

Although the values of the surface tension and wetting angle of the pitch melt are comparable to water [21], the cohesive–adhesive interaction and wettability are mainly determined by the polarity of the substances in contact and the dispersion forces, and therefore the mechanisms for granule growth can differ markedly. In addition, the surface of the pellets during the spraying of wastewater is formed by salting out, and the size of the particles deposited on the surface of the contaminated soil is an order of magnitude smaller than that of the pitch particles. Figure 5c clearly shows the cavities, while the surface structure of the granule in Fig. 5b looks much more uniform.

Figure 5a shows a photo of the granules obtained in Experiment 10. Based on its internal structure, we can imagine that the granulation mechanism is of a

Table 6. Results of sieve analysis of the product were obtained by spraying wastewater and pitch melt into a fluidized bed of contaminated soil

Fraction, mm	Exp. 10, %	Exp. 11, %	Exp. 12, %
<0.25	0.17	0.12	2.42
0.25–0.49	1.88	2.54	3.33
0.50–0.99	70.51	94.29	76.47
1.00–1.99	26.42	2.87	17.41
≥2.00	1.02	0.18	0.37



a b c

Fig. 5. Fraction 1.00–1.99 mm: (a) granule from Experiment 10; (b) granule from Experiment 11; (c) granule from Experiment 12. At Fig. a: 1 – contaminated soil, 2 – pitch, 3 – deposits of salts containing heavy metals.

mixed nature. The binding of the initial particles of the contaminated soil is carried out by both pitch and salting. In this case, the cavities in the agglomerate are partially filled with salt deposits, which increases the strength and integrity of the structure of the final product. This can be seen by increasing the number of granules of 1.00–1.99 mm by more than 10% (Experiments 10 and 12) and by reducing dust formation.

At Stage 3 of the studies, a fraction of 1.00–1.24 mm was used for encapsulation with ceresin, which was previously produced at the specified technological parameters (Experiment 10).

We have made an approximate calculation of the minimum amount of ceresin to be fed into the bed under the following assumptions: ceresin is evenly distributed over the surface of the spherical particles; particle mass is 1 kg; bulk particle density is ~1250 kg/m^3; ceresin density is ~900 kg/m^3). The calculation results are shown in the graph in Fig. 6.

It should be noted that the amount of theoretically required ceresin strongly depends on the size of the particles in the bed. So, for example, for the coating of particles d_p = 1 mm with a ceresin film with a thickness of 200 μm, the ratio of the layer mass and the mass of the encapsulator is 1.0 : 0.9, and for d_p = 1.5 mm it is 1.0 : 0.6.

The results of Stage 3 of the experimental studies are presented in Table 7. When the consumption of ceresin G_c was 0.6 kg/h and 0.8 kg/h (Experiments 13 and 14), the resulting product did not pass the waterproofness test because chloride ions were found in the water extract as well as trace amounts of organic compounds that make up the pitch. This indicates an incompleteness/heterogeneity of encapsulation.

An increase in the flow rate to 1 kg/h (Experiment 15) made it possible to obtain a sealed, moisture-resistant product, which, according to the results of the bioassay on hydrobionts, was assigned to hazard class 5.

Unloading was carried out through a sifter, the pressure in which was set to 1.1 bar, which allowed particles less than 1 mm to be filtered out. The difference between the expected and actually unloaded product quantity was ~5%.

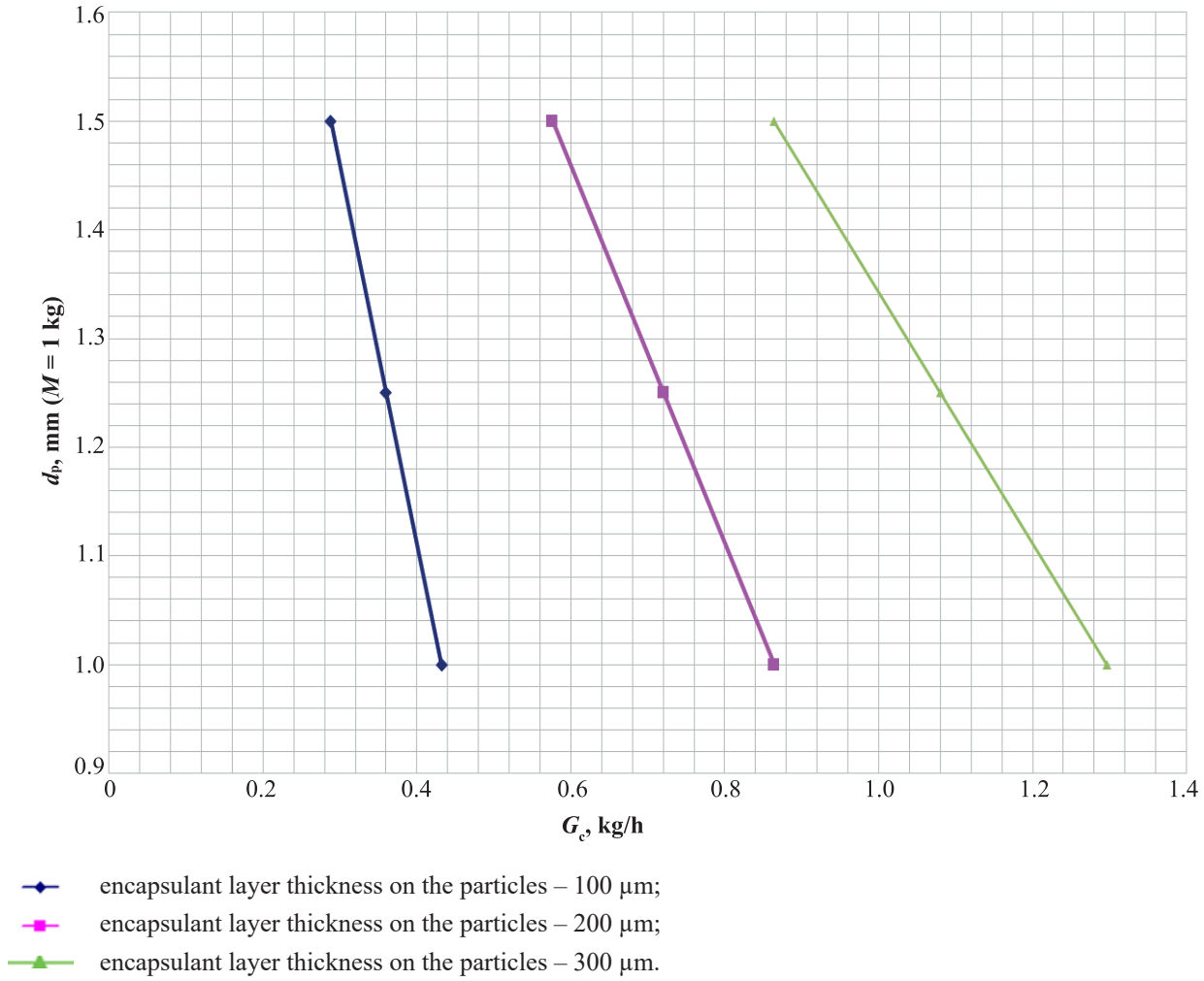


Fig. 6. Calculated dependence of the required encapsulant (ceresin) consumption on the initial particle size in the fluidized bed (the mass of the bed is 1 kg).

Table 7. Results of experiments on encapsulation of granulate with ceresin

No. exp.	Parameters							Notes
	M, kg	G_{FA}, m^3/h	T_{FA}, °C	T_B,* °C	T_{EA},* °C	P_{AA}, bar	G_c, kg/h	
13	1.0	107–114	18–23	18–30	15–24	1.4–2.0	0.6	Water resistance test failed
14	1.0	105–113	18–23	17–33	16–26	1.4–2.0	0.8	Water resistance test failed
15	1.0	103–115	18–23	17–35	15–29	1.4–2.0	1.0	Water resistance test passed

* Temperature in the bed (T_B) and temperature of the exhaust gases (T_{EA}), respectively.

Figure 7 shows the appearance of the resulting product. Image was taken using a Leica DM2500 microscope.

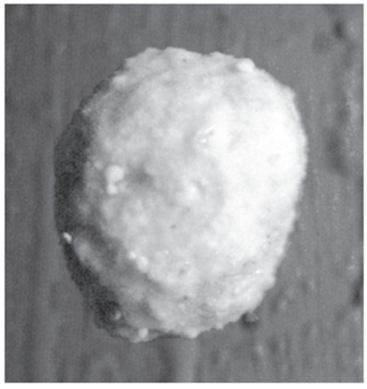


Fig. 7. Encapsulated waterproof hazard class-5 product (Experiment 15, fraction 1.25–1.99 mm).

CONCLUSIONS

In conclusion, the present study demonstrates the potential for a drying–granulation–encapsulation process for toxic waste and hazardous chemicals with a hazard class of 1–3 in one device, producing a bulk product of hazard class 5 that is moisture-resistant and suitable for transportation and long-term storage.

The technology for encapsulating toxic waste and wastewater can be implemented, for example, in an industrial continuous drying–granulation unit of the GLATT-GFG-500 type, which has been successfully used for the dry granulation of reaction masses obtained as a result of the alkaline hydrolysis of lewisite [11]. In industrial implementation, the ratio of the bed mass to the mass of the encapsulant is expected to change through a reduction in the amount of ceresin sprayed since, according to the experience with the GLATT-GFG-500 unit, the size of the salt particles produced and discharged reached 5–6 mm.

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Authors' contribution

Yu.A. Eleev – formulation of research goals and objectives, development of experimental methods, analysis of the results.

Yu.S. Bogoyavlenskaya – experimental research and primary processing of the obtained experimental data.

E.N. Glukhan – general management of the research process and preparation of materials for publication.

V.F. Golovkov – scientific support and assistance in the analysis of research results.

V.V. Afanasyev – scientific and technical support for the modernization and preparation of the experimental unit.

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