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Testing of the Granulation Process for the Preparation of a Mixture with the Chemical Composition of a Heavy Tungsten Alloy

Olgierd Janusz GOROCH[*], Zbigniew GULBINOWICZ,
Katarzyna BARCZ, Paweł SKOCZYLAS,
Mieczysław KACZOROWSKI

Warsaw University of Technology, Faculty of Production Engineering,
Institute of Mechanics and Printing
85 Narbutta Str., 02-524 Warsaw, Poland
[*]Corresponding author's e-mail address and ORCID:
o.goroch@wip.pw.edu.pl; https://orcid.org/0000-0002-9852-8289

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Abstract. The subject of this paper are the results of testing the process of preparation of a granulate made of a mixture of powders with the chemical composition of a heavy tungsten alloy. Different methods of granulation are discussed which reveal how widely they vary in physical and chemical phenomena. It is a testament to how vast the research field of granulation of matter is. The importance and significance of granulation methods were proven with the distinguishing features of the granulated form of the stock materials and products compared to their powdered (dusty) form.

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Granulation is applied to produce convenient forms of products or intermediates which are acceptable to end users, and to facilitate or even enable the application of products and intermediates in appropriate technologies.
Keywords: material engineering, granulates, heavy tungsten alloys, sub-projectiles.

1. BACKGROUND AND ORIGINS

The Laboratory of Heavy Alloys at the Institute of Mechanics and Printing of the Warsaw University of Technology (Poland) routinely researches heavy tungsten alloys (HTAs), materials used in the production of firearm ammunition. HTAs, [1, 2] are characterized by high density (17.2 to 18.6 Mg/m^3) and the dependency of mechanical properties on the content of tungsten, alloy additives and the processing technology, which includes sintering, heat treatment and plastic working [3]. The as-sintered HTA structure comprises hard, spheroidal grains of tungsten, between 20 and 60 μm in diameter, embedded in a plastic matrix called the binding phase.

Because of their very high density, good mechanical properties, and high hardness, HTAs are applied in the defence industry as sub-projectile penetrators [4]. Note that the penetrators are made in two versions: the APDS (*Armour Piercing Discarding Sabot*) penetrators and FAPDS (*Frangible Armour Piercing Discarding Sabot*) penetrators. The tests and their results presented in this paper enabled an analysis of the forming process based by powder granulation of HTAs in the production of APDS penetrator blanks.

Granulation is a concept of fabrication of solid particles with a specific shape, dimensions, and physical and chemical properties. The ambiguity of the concept definition makes it a blanket term for various (and often significantly different) methods of fabricating the products. The methods can be related to other unit operations, including crystallisation and drying. This is the reason for the difficulty in providing a simplified classification of granulation methods.

The stock material for the fabrication of granules [5] can be molten, followed by spraying into droplets of a suitable size, which are cooled to crystallise and become more or less regular pellets. This method is applied, for example, in the production of crystalline sulphur; the production process is performed in pelleting towers. The upper section of a pelleting tower houses the alloy spraying process, while air is forced at a pressure from the bottom section and up the tower. The process of granulation from a suspension can be performed in a similar manner, using similar equipment units and combined with drying and agglomeration of the fine particles formed in the first stage of the process (the fusion of small pellets into larger ones by impact).

However, the most popular method of granulation consists in the use of fine solids (powder or liquid) which are processed into larger granules [6], or agglomerations of the initial solids. The process and equipment required for the method may vary.

A common distinction is made between pressureless and pressurized granulation. Pressureless granulation is viable in a fluidized bed setup or in a free-flowing layer of the granulated stock (in a drum, disc, or vibrating screens). Pressurized granulation (like pelletizing, brick-forming, or compression) requires an elevated pressure which results in the mechanical strengthening of a specific number of initial particles. The strengthening occurs when the initial particles are brought close to one another, usually by plastic strain. Certain applications exist in which the pressure-granulated material is heated if reasonable due to the processing requirements or the service requirements for the finished product. If powder stock is granulated, a binding liquid is used at the agglomeration stage both in pressureless and pressurized granulation. Binding liquids can help attract the initial particles to one another during the granulation process only or generate permanent cohesion forces which improve the strength of bonding between the initial particles also when the product is dry.

The different methods of granulation discussed above reveal how widely they vary in physical and chemical phenomena. It is a testament to how vast the research field of granulation of matter is. The importance and significance of granulation methods were proven with the distinguishing features of the granulated form of the stock materials and products compared to their powdered (dusty) form.

2. TEST MATERIALS

The Institute of Mechanics and Printing Technology at the Warsaw University of Technology operates a full complement of specialised equipment for the fabrication of large series of products from tungsten composites. This capacity enables the proprietary fabrication of test specimens in compliance with the required processing parameters. The process of powder mixing requires considerable time (20 h) [7] and dedicated rotary ball mills.

Table 1 presents the composition of the mixture used to fabricate the granulate. Table 2 lists the granulation fabrication parameters. The sintering applied at the specified temperatures was intended to soak the solid material. Once poured into a mould, the granulate was sintered for 45 min in a hydrogen gas atmosphere inside the furnace illustrated in Fig. 1b. The process was followed by grinding and vibration sifting of the sintered material. The granulate produced in this way enabled better compaction of the powder. The preliminary tests revealed that 1,100°C was the optimum temperature for the granulate preparation process. At 1,000°C, the produced mixture would spill and generate heavy dust while sifting, whereas 1,200°C caused a heavy sinter, which prevented sifting.

Table 1. Granulate mixture composition [1]

Elements	Chemical analysis [%]	Theoretical density [g/cm^3]
W	96.4	18.44
Ni	2.6	
Fe	0.8	
Co	0.2	

Table 2. Granulate production parameters

Temperature [°C]	Sintering time [min]
1,000	45
1,100	45
1,200	45

3. TEST METHODOLOGY

During the testing performed at the Institute of Mechanics and Printing of the Warsaw University of Technology, the stock material was screened with a vibration sifter. The efficiency of screening defined the severity of screening (i.e. the assessment of screening precision of the fine particles) of the components (the quantity of the stock in a single batch fed to the chemical processor) and the flow rate of the fines. A particle size distribution analysis was applied to determine the mean particle size, the median, and the uniformity index.

It was followed by density tests, one of the fastest and straightforward non-destructive methods applied in the testing of materials. Density tells the mass of a specific volume of the material of interest; when determined, the parameter is very often used in further analysis of the material, including spectrometry, particle size measurements, and other methods. The determination of density is a relatively inexpensive and fast method of uniform control of materials.

One of the most important physical-mechanical properties which characterise flowing materials is the angle of repose. The angle of repose depends on the degree of the mutual mobility of particles: the higher the mobility, the lower the angle of repose. The angle of repose is determined by pouring a granular charge onto a level plane with a low flow rate of the particles. When rolling down into a cone, the angle between the slope of the particles and the level plane is angle α (Fig. 1).

For a dry charge, angle α is approximate to the angle of internal friction; by this, and thanks to the ease of measurement of the angle, it is often determined as a characteristic of the material of interest. In most materials, the angle of repose depends on the moisture content of the material and increases with the latter [8, 9, 10].

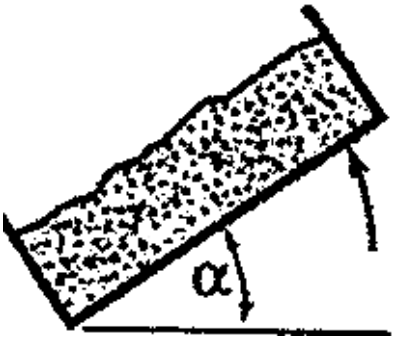

Fig. 1. Angle of repose of a flowing material

Another important characteristic of a mixture of powders is the particle size distribution, determined by laser light diffraction with the following values: D(0.1), the diameter of 10% of the particles below its specified value; D(0.5), the median of diameter referenced to volume (V); and D(0.9), the diameter of 90% below its specified value.

The structural tests were performed using a traditional metallographic method to reveal the microstructure of the fabricated granulate. The metallographic examination was performed on specimens prepared by conventional grinding and polishing on a buffing grinder with adjustable feed pressure, with the rotation of the specimen relative to the rotating disc. An Olympus IX-70 metallographic microscope was used for the examination.

4. TEST RESULTS

4.1. Measurement of particle size by dry screening

The material [11] was sintered at 1,100°C for 45 minutes in a hydrogen gas atmosphere furnace, cooled down, reduced in size, and screened on vibration sifters. Two mixtures of powders screened into two fractions were selected for further testing, a fraction of ≥ 80 μm and a fraction of $40\ \mu m \leq x < 80\ \mu m$. The granulated HTA was prone to breaking up (Fig. 2).

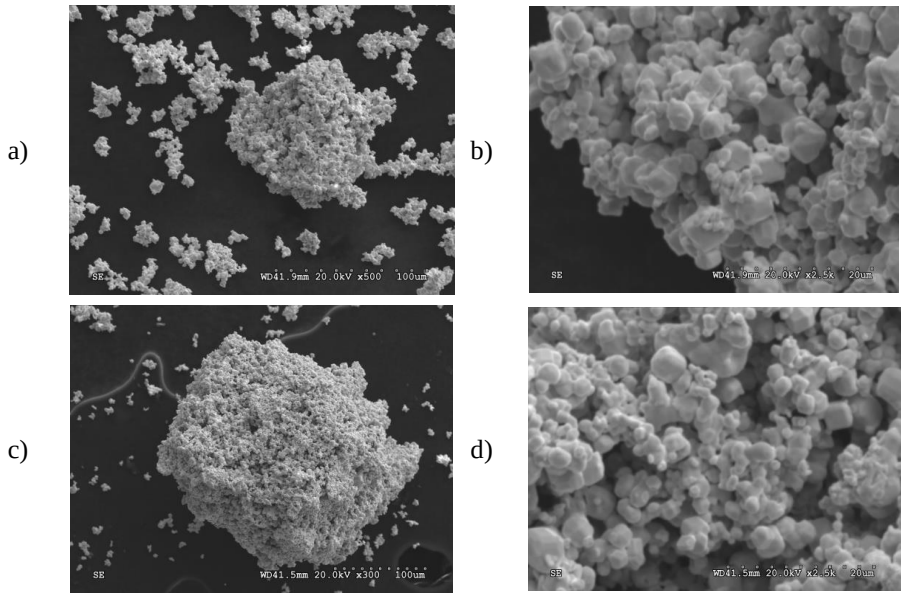

Fig. 2. SEM imaging of the granulates: a) & b) – 40 μm ≤ x < 80 μm granulate fraction; c) & d) – ≥ 80 μm granulate fraction

4.2. Bulk density measurement

The first method used to determine the bulk density involved a Hall flowmeter. The test was performed by pouring the granulate through a graduated cylinder inside a metering funnel. High molecular friction was observed in each of the tested granulated mixtures (fraction of ≥ 80 μm and < 80 μm) and rendered the measurement infeasible.

The second method used to determine the bulk density involved pouring a specific powder specimen through an arrangement of oblique panels (called Scott's volumeter). Table 3 shows the test results for the loose poured powder with 25 cm^3 of volume and 47.7 g of mass, which accurately filled the cylinder.

Table 3. Bulk density test results

Granulate particle fraction ≥ 80 μm				**Granulate particle fraction 40 μm ≤ x < 80 μm**			
#	Gross weight [g]	Granulate weight [g]	Bulk density [g/cm^3]	#	Gross weight [g]	Granulate weight [g]	Bulk density [g/cm^3]
1	109.84	62.14	2.49	1	110.82	63.12	2.52
2	108.84	61.14	2.45	2	110.78	63.08	2.52
3	107.90	60.2	2.41	3	111.74	64.04	2.56
Mean		**61.16**	**2.45**	**Mean**		**63.1**	**2.54**

The bulk density was determined with the ratio of the poured mixture mass to the cylinder volume per PN-EN 23923-2. The bulk density was reduced with the increase of the particle size (Table 3). The cause was that the finer fraction mixture featured a considerable share of dust, which was an unfortunate by-product of vibration screening. This increased the packing density [12] of the powder and adversely increased the granulate's angle of repose.

4.3. Flow rate test results

Another method applied to analyse the fabricated granulate was a powder flow rate test. The angle of repose depends on internal friction between the particles, or the resistance of motion between the particles. It was demonstrated that the angle of repose largely depended on the test method. Practical challenges resulted from the separation of material and the compaction or aeration of the powder while the cone was being formed.

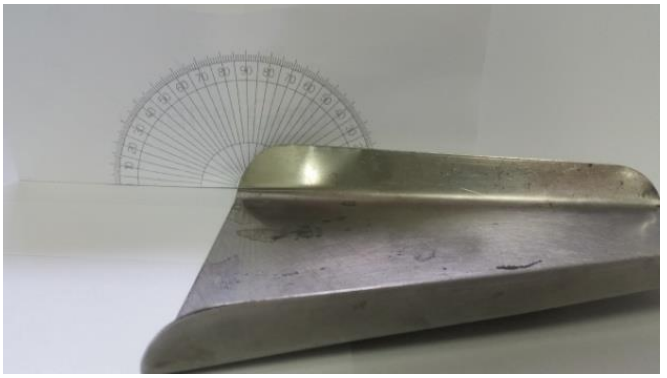

Fig. 3. Photographic image of the granulate angle of repose test

The selected method consisted in measuring 100 g of the powder into a metallic cylinder, followed by spreading the contents of the cylinder on a flat metal sheet (Fig. 3). The flat metal sheet was then lifted by one edge until the powder began sliding along it; the lifting motion was stopped, and the angle of inclination was measured between the underside of the flat metal sheet and the level surface below it. Table 4 lists the angle of repose test results. The graph shows that the larger the fraction of the granulate, the lower the angle of repose. Lower angles of repose improved the compressibility of the powder. It seemed reasonable to apply granulation in powder sintering, the more so that the angle of repose for the non-granulated mixture was 25°.

Table 4. Angle of repose test results

Granulate particle fraction ≥ 80 µm	14°
Granulate particle fraction 40 µm ≤ x < 80 µm	23°

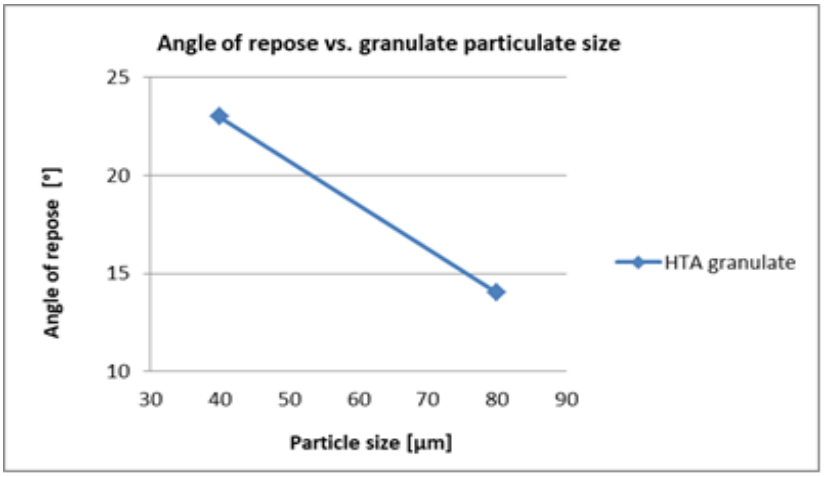

Fig. 4. Graph of variation in the angle of repose

4.4. Particle size and shape analysis results

There are many methods of particle / grain size measurement. A general classification includes direct and indirect methods. The direct methods use the grain mesh size, Martin's diameter or Feret's diameter, for example. The indirect methods are based on physical phenomena with certain conversion techniques and include measurement of float viscosity, diffraction of light in a suspension, photo-electrical spatial scanning, and centrifugal grain segregation. It must be made known that each test method generates different information about the particle size distribution. The results of the determination are primarily affected by the test method. The test methods vary in the material properties, which include geometric characteristics, density, surface characteristics (porosity), and more.

The particle size method applied in the research discussed in this paper used laser light diffraction. Here, the significant effect on the test results originated from the particle shape, porosity, and particle size range in the specimen.

Table 5 lists the particle size analysis results in ranges d(0.5), d(0.1), and d(0.9), determined by laser light diffraction.

Table 5. Malvern test particle size results

Granulate	d (0.5) μm	d (0.1) μm	d (0.9) μm
Granulate fraction ≥ 80 μm	31.95	7.60	166.62
Granulate fraction 40 μm ≤ x < 80 μm	19.03	7.76	39.97

4.5. Compaction of the granulate

Compaction is one of the simplest procedures in which a flowing powder complies with the action of uni- or multi-directional external forces which cause the material to become compacted. To produce a highly dense material after sintering, compaction pressure must be applied to produce a blank with the maximum density possible and the best possible density uniformity within the blank volume. Table 6 lists the results of blank forming. An approximately 30 g batch of the granulate was compacted in a dia. 16 mm die.

Table 6. Blank forming test results

Granulate fraction ≥ 80 μm				
#	Batch [g]	Compaction pressure [MPa]	As-compacted height [mm]	As-compacted specimen density [g/cm^3]
1	30	294.19	14.40	10.46
2	29.98	392.26	13.35	11.17
3	29.88	196.13	15.30	9.71
Granulate fraction 40 μm ≤ x < 80 μm				
#	Batch [g]	Compaction pressure [MPa]	As-compacted height [mm]	As-compacted specimen density [g/cm^3]
1	30.02	294.19	14.15	10.55
2	30.00	392.26	13.60	10.97
3	29.98	196.13	15.50	9.62

The as-compacted specimens were sintered in a hydrogen gas atmosphere furnace following a processing formula developed for the materials from which sub-projectile penetrators are made.

Following the sintering process, the actual density of the specimens was determined, the results of which are listed in Table 7. The assumed theoretical density was 18.4 g/cm^3.

Table 7. Density test results

HTA granulate fraction ≥ 80 µm			**HTA granulate fraction ≥ 80 µm**		
Compaction pressure [MPa]	#	Actual density [g/cm^3]	Compaction pressure [MPa]	#	Actual density [g/cm^3]
294.19	1	18.04	392.26	1	17.97
	2	17.89		2	17.94
	3	17.83		3	17.90
Mean		**17.92**	**Mean**		**17.94**
HTA granulate fraction ≥ 80 µm			**HTA granulate fraction 40 µm ≤ x < 80 µm**		
Compaction pressure [MPa]	#	Actual density [g/cm^3]	Compaction pressure [MPa]	#	Actual density [g/cm^3]
196.13	1	17.87	294.19	1	17.69
	2	17.86		2	17.97
	3	17.81		3	17.86
Mean		**17.85**	**Mean**		**17.84**
HTA granulate fraction 40 µm ≤ x < 80 µm			**HTA granulate fraction 40 µm ≤ x < 80 µm**		
Compaction pressure [MPa]	#	Actual density [g/cm^3]	Compaction pressure [MPa]	#	Actual density [g/cm^3]
392.26	1	17.85	196.13	1	18.01
	2	17.90		2	17.94
	3	17.89		3	17.97
Mean		**17.88**	**Mean**		**17.97**

4.6. Structural tests

The structural tests included traditional metallographic examination and fractography with SEM [13]. The metallographic examination was performed with an Olympus IX-70 metallographic microscope, at various powers of magnification, and with bright field and NIC (Nomarski interference contrast). The fractographic examination was performed with a Zeiss LEO 1530 SEM.

4.6.1. Light metallographic microscopy

All specimens were examined by optical metallographic microscopy. Figure 7 illustrates an example of the structure in specimen # 2 (the HTA granulate ≥ 80 μm), compacted as sintered at 392 MPa.

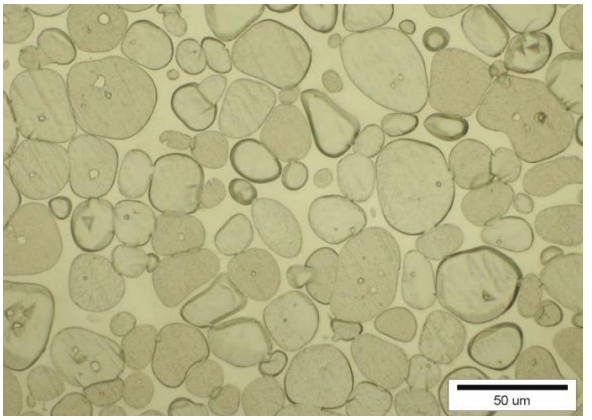

Fig. 5. Microstructure of specimen # 2 (HTA granulate ≥ 80 μm), compacted at 392 MPa

The photograph in Fig. 5 reveals a specific structure of HTA. The tungsten grains were distributed within a matrix which was an Ni-W-Fe-Co solution [14, 15]. The findings revealed that irrespective of the direction of cutting the metallographic sections (in parallel or perpendicular to the bar centreline), the tungsten grains had a nearly spheroidal geometry directly after the sintering process.

4.6.2. SEM structural tests

Figure 6 illustrates the image of the microstructure of specimen # 2 (the HTA granulate ≥ 80 μm) as sintered. The image was generated with the SEM in a SE (secondary electrons) application. Figure 6 reveals spherical tungsten grains uniformly distributed in the matrix with large plates which were precipitates of the binding phase.

In some locations, the binding phase formed a continuous envelope around the tungsten grains. It could be surmised that the binding phase was the root cause of the extreme brittleness in high-cobalt alloys.

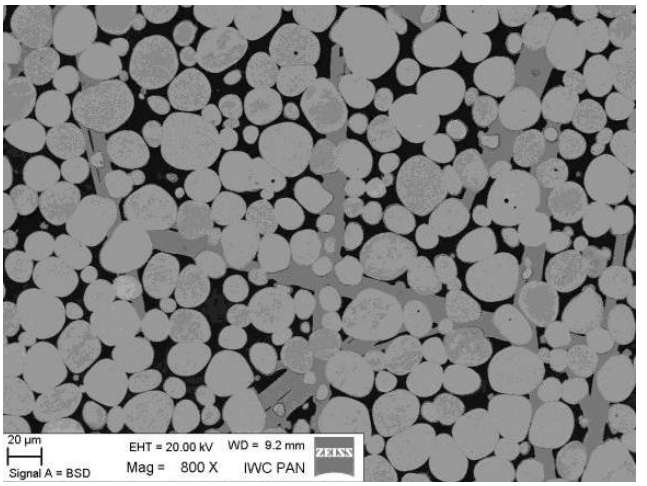

Fig. 6. Image of specimen # 2 (HTA granulate ≥ 80 µm), compacted at 392 MPa

5. SUMMARY AND CONCLUSIONS

The results presented in this paper demonstrated the requirements to be met when producing granulates from HTAs (the optimum preliminary sintering temperature) and the criticality of the selection of a suitable granulate fraction followed by its correct characterisation. It was demonstrated that testing of the granulates (the mixtures and the fabrication method), including the bulk density and angle of repose tests, is important to downstream processing of the material. A significant challenge identified at the stage of granulate preparation was the separation of the granulate particles from dust during vibration screening (sifting). The dust was an obsolete by-product which increased the mass and the bulk density while reducing the angle of repose, a phenomenon adverse to the mixture compaction.

However, the results of the preliminary tests presented here, and their analysis permitted the following final conclusions:

1. It is feasible to fabricate a granulate from a powder which meets the strength specifications defined for the barstock intended for the production of armour-piercing sub-projectile penetrators.
2. The preliminary tests proved that the granulate mass increased with the bulk density in a steady volume.
3. It was proven that the angle of repose of the granulate (Fig. 6) was reduced as the grain size increased.
4. The produced granulate satisfied the requirements for actual density (Section 1).

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Badanie procesu przygotowywania mieszanki o składzie chemicznym wolframowego stopu ciężkiego metodą granulacji

Olgierd Janusz GOROCH, Zbigniew GULBINOWICZ,
Katarzyna BARCZ, Paweł SKOCZYLAS,
Mieczysław KACZOROWSKI

Politechnika Warszawska, Wydział Inżynierii Produkcji,
Instytut Mechaniki i Poligrafii
ul. Narbutta 85, 02-524 Warszawa

Streszczenie. Przedmiotem pracy są wyniki badań procesu przygotowania granulatu wykonanego z mieszaniny proszków o składzie chemicznym wolframowego stopu ciężkiego. Omówione różne sposoby granulacji pokazują wyraźnie jak bardzo odmienne są mechanizmy i zjawiska fizyko-chemiczne jakie występują w poszczególnych przypadkach. Świadczy to o obszerności pola badawczego ogólnie rozumianej granulacji. O jej znaczeniu i ważności świadczą cechy wyróżniające formę granulowaną surowców i produktów w porównaniu z formą proszkową (pylistą). Granulacja jest więc stosowana, aby uzyskać wygodną, akceptowalną przez użytkowników końcową formę produktu bądź półproduktu oraz aby ułatwić, bądź nawet umożliwić ich stosowanie w odpowiednich technologiach.
Słowa kluczowe: inżynieria materiałowa, granulaty, wolframowe stopy ciężkie, amunicja podkalibrowa