MICROPOWDERS PRODUCTED BY DISINTEGRATOR MILLING

P. Peetsalu, D. Goljandin, P. Kulu, V. Mikli, H. Käerdi

Abstract

A theoretical model for size reduction of ductile materials by collision is proposed. A disintegrator milling system with a centrifugal-type classifier is developed; milling of ductile nickel and cobalt-based alloy powders is described. The developed disintegrator milling system of high separative sensitivity enables to produce the metallic micropowders with size below 5 μm. The main characteristics of the powders produced (particle size, powder morphology, oxygen content and specific surface area) are determined. Results of different testing methods are compared. Resulting from the studies of the powders granularity and morphology, the used methods of laser particle granulometry and image analysis describe the characteristics of powders adequately.

Keywords: milling by collision, disintegrators, micropowders, characterization of powders, granularity and morphology of particles

INTRODUCTION

Metal powder from recycled metal – a raw material for powder metallurgy – is produced by different technologies [1]. The most widely used technology is the atomizing of melted metal. An alternative technology for producing metal powder is the milling of metal chips. One of the methods for producing metal powders offered here is grinding by collision. This method has some advantages:

- nothing will be lost in the chemical composition of the alloy,
- the quality of the material will increase, as the microstructure of the material improves due to the intensive impact stresses and mechanical activation,
- the retreatment of chips solves two problems: the utilization of chips, and production of raw materials for powder metallurgy.

The first systematic experimental research concerning grinding of brittle materials by collision at a certain velocity was conducted by Rumpf [2] and his school.

The reducing mechanism of a ductile metal, such as steel chips, is completely different from that of grinding a brittle one. In this investigation, main attention was paid to the mechanisms of breaking spherical particles, and to the characterization of milled product from a ductile metal. The model was verified and tested on stainless steel.

THEORETICAL MODEL FOR SIZE REDUCTION OF DUCTILE MATERIALS BY COLLISION

Stresses on collision

The stresses generated in the particle at collision may be estimated by two extreme models: either according to the quasistatic “Hertz model” applied to a spherical particle or

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to the “wave model”, where the particle with a plane side hits the target exactly with the same side. According to the Hertz model, the stresses σ_H of collision in a general case are

$$\sigma_H = 0.279 \cdot A^{-3/5} \cdot B^{1/5} \cdot C^{4/5} \cdot v^{2/5}, \qquad (1)$$

where v is the velocity of collision, A, B and C are the coefficients depending on the radii of the colliding bodies and on their Young’s modulus.

According to the wave model, the stresses σ_w in the contact surfaces are

$$\sigma_W = \rho_1 \cdot c_1 \cdot \rho_2 \cdot c_2 / (\rho_1 \cdot c_1 + \rho_2 \cdot c_2) \cdot v, \qquad (2)$$

where ρ_1 and ρ_2 are densities of the colliding bodies, c_1 and c_2 are velocities of the elastic waves and $c_i = (E_i/\rho_i)$, $i = 1.2$

Figure 1 shows the dependence of the stresses in a stainless steel particle with a diameter of 2 mm on the collision velocity and on the target (grinding body) material.

Since real particles differ from ideal spheres, and they do not collide precisely with plane surfaces, the actual real stresses are between these limit values (Fig.1.).

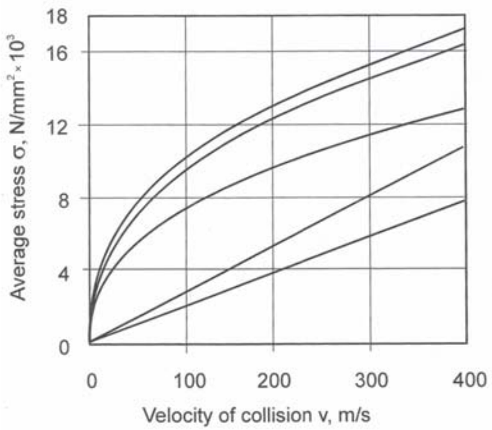

Fig.1. Dependence of the maximum of average stress on the velocity of collision. stainless steel particle (d = 2 mm) collision to: 1- hard metal WC-6Co plate; 2 – plate of the same steel; 3 – the same equal to another particle.

Collision of a particle with another particle or plate

When grinding of materials, e.g. stainless steel chips [3], initial metal chips are loose and the metal is work hardened. At collision, its behavior is nearly similar that of a brittle material. With reduction in size, a chip’s particles approach an isometric and even spherical form. Next, collision of two spherical ductile metal particles (Fig.2.) will be analyzed.

The distribution of pressure stresses σ on the contact area is

$$\sigma = \frac{3F}{2\pi r_k^2}\sqrt{1-\left(\frac{r}{r_k}\right)^2}, \qquad (3)$$

where F is the contact force at collision.

The maximum stress σ_m and the average stress σ_{av} can be expressed as follows:

$$\sigma_m = \frac{3F}{2r_k^2}, \quad \sigma_{av} = \frac{F}{r_k^2}, \quad \sigma_{av} = 2 \cdot \sigma_m / 3 \qquad (4)$$

Figure 1 shows the dependence of average stresses on the collision velocity. As the order of stresses is high, we can suppose that the radius of the plastic deformation area is equal to that of the contact area r_k.

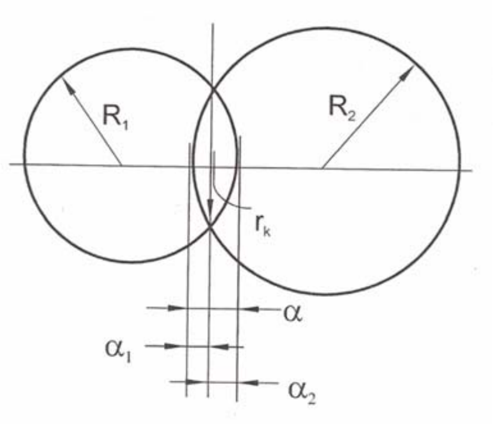

Fig.2 Collision of two spherical particles: α - approach of the centres, α_1 and α_2 – approach of each particle, r_k – contact area radius.

Size reduction model

After a certain number n_1 of loadings, the surface of the particles will be completely covered with plastic deformations

$$n_1 = \frac{4 \cdot \pi \cdot R^2}{\pi \cdot r_k^{\;2}} = \left(\frac{2 \cdot R}{r_k}\right)^2 = 1.15 \cdot \rho_1^{-2/5} \cdot C^{-2/5} \cdot v^{-4/5} \qquad (5)$$

Repeating such series many times (a great number of cycles), the area will be repeatedly deformed. As a result of repeated collision loading and fatigue breaking of the particle surface, small pieces of the size of δ will be separated. After a certain number of loading series n_2 a layer with a thickness of δ will be separated. The size of the particle will be decreased by 2δ. A simple differential equation and its solution can be written as

$$\frac{dR}{d\delta} = -\frac{n}{n_1 n_2}, \quad R = R_o - \frac{n \cdot \delta}{n_1 n_2} \qquad (6)$$

where R_o is the initial radius of the particle.

The particle will be ground when the radius R approaches the boundary size to be separated in a classifier. If the boundary size is equal to δ (7), the necessary number of collision loadings for grinding the particle of a size $2R_o$ is

$$n = n_1 \cdot n_2 \left(\frac{R_o}{\delta} - 1\right) \; or \; n \cong n_1 \cdot n_2 \cdot R_o / \delta \qquad (7)$$

The depth of plastic deformation is of the same order as the approach of $\alpha_1 = \alpha$. Supposing that

$$\delta = \alpha / k, \quad k = 2\text{-}8, \qquad (8)$$

The grinding of stainless steel particles of size d = 2-2.5 mm at the collision velocity v = 150 m/s, the approach α = 80 μm, the ground particle size in the product is in the order δ = 20 μm and so k = 4.

On the other hand, by separative grinding, stainless steel particles with the above described size will be fully ground at approximately n = 30.000-40.000 collision loadings. In our cases, from (5) and (7)

$$n_1 \cong 17 \text{ and } n_2 = \frac{n \cdot \delta}{n_1 \cdot R_o} \cong 20 \qquad (9)$$

It follows that by impact at the same surface spot, the particle of stainless steel must be loaded $n_2 \approx 20$ times before fatigue breaking takes place. That is low cyclic fatigue breaking, which occurs due to the high rate of intensity of stresses at high velocity collision.

Figure 3 illustrates the principal scheme of a disintegrator mill and formation of powder particles by the milling of brittle and ductile materials.

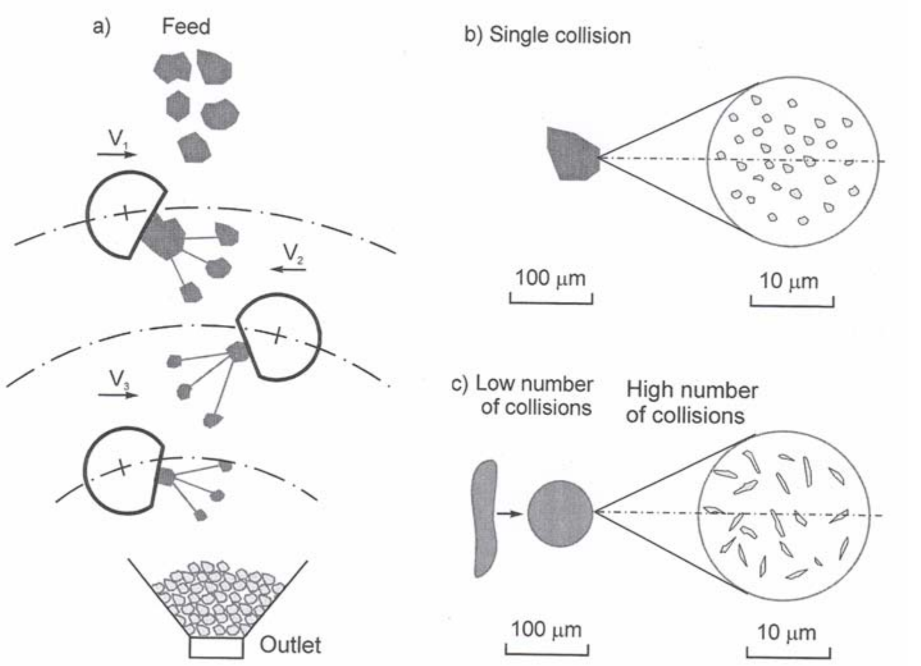

Fig.3. Principal scheme of size reduction by disintegrator milling (a) and mechanisms of fracture of brittle (b) and ductile materials (c).

GRINDABILITY OF DUCTILE ALLOYS

Disintegrator systems for materials processing

Multi-functional DS-series disintegrators [4], operating in three different modes: direct milling, separative milling, and selective milling, were developed. To process different materials, the series includes the mill DS-158 for preliminary milling of continuous chips, the semi-industrial device DSL-115 for intermediate milling and the laboratory separative milling system DSL-160 with inertial or centrifugal classifiers. The parameter of grinding – the specific treatment energy – was used to estimate grindability [5]. Depending on the type of the device and rotation velocities, the specific energy of milling of various machines was as follows: 1.8 kJ/kg for the preliminary mill DS-158, 17.7 kJ/kg for the semi-industrial mill and 19.4 kJ/kg for the laboratory disintegrator DSL-160 at the rotation velocity of 10000/10000 rpm. The special laboratory milling system DSL-175 was used to produce ultrafine powder (Fig.4). The parameters of a disintegrator mill are given in Table 1.

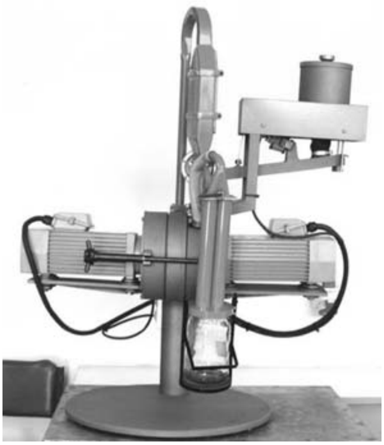

Fig.4. Laboratory disintegrator milling system DSL-175.

Tab.1. Parameters of disintegrator mill DSL-175

Rotation velocity of rotors, [rpm]	Maximum velocity, [m/s]	Specific energy of treatment E_s , [kJ/kg]
2000/2000	32	0.8
4000/4000	64	3.1
8000/8000	128	12.4
12000/12000	192	28.0

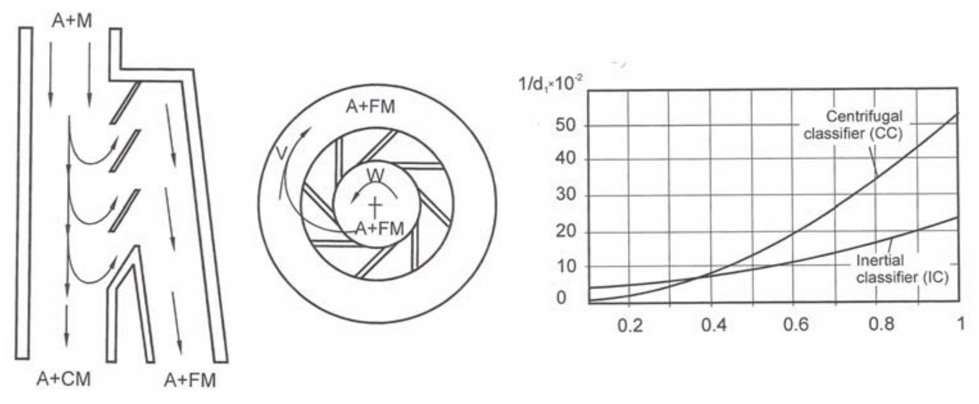

Fig.5. Principal schemes of (a) inertial and (b) centrifugal classifiers and (c) dependence of the separation effect on the classifying parameters (A – air, M – materials, CM – coarse material, FM – fine material, IC – inertial and CC – centrifugal classifier).

The disintegrator system DSL-175 for ultrafine milling of metallic powders was modified. On the basis of the experiments described, a new centrifugal-type classifier was designed (Fig.5), with its parameters shown in Fig.6.

The separation effect F_{sep} of the centrifugal classifier (CC) is

$$F_{sep} = 1/d_1 = f(Q, v_i, b, h)\ , \qquad (10)$$

where Q is the productivity of separative air aspirated through the grid, v_i is the velocity of particles ($v_i = v+v_c$, v – velocity of air flow in a disintegrator, v_c – velocity of blades of CC), b and h are the dimensions of CC.

An experimental classifier was designed and tested. Table 2 shows the results of granularity (laser analysis) of the powders classified by this device.

Tab.2. Characteristics of stainless steel powder ground by the disintegrator DSL-160 with a centrifugal classifier.

Classifying parameters – relative velocity[1]	Particle size and distribution %		
	d_m [μm]	$d_m < 5$ μm	$d_m < 3$ μm
0.125 (1/8)	3.1	60	50
0.25 (2/8)	1.2	93	80
0.375 (3/8)	1.1	88	85
0.5 (4/8)			

[1] v_i/v_{max} (ratio of particle velocity to the maximum possible velocity in the classifier)

[2] d_m – mean diameter

Production of micrometrical metal powders from prealloyed powders

To produce composite spray powders with higher corrosion resistance, prealloyed metal powders, such as a binder metal, were used.

The following metal powders as initial materials served as as a binder metal.

- Co-based powder Alloy 59, MBC Metal Powders
- Cr-Ni alloy powder Fukuda SX717, Fukuda Metal Foil & Powder Co. LTD.,
- Ni-based powder Anval Ultimet, Carpenter Powder Products.

The chemical composition and particle size of the alloy powders used are given in Table 3.

Tab.3. Selected metal powders and their composition

Powder type	Chemical composition [wt.%]	Initial particle size [μm]
Alloy	Ni- 23Cr-10Mo-1Fe-Mn-Si	45-150
Fukuda	Co-26Cr-9Ni-5Mo-2W-0.8Mn-0.3Si-0.08N-0.06C	53-150
Ultimet	Cr-42Ni-2.5Mo-1.0Si-0.5B	-45

To produce ultrafine powders (less than 5 μm), we used the disintegrator milling system DSL-175 with inertial and centrifugal classifiers (see Fig.4). Powders were milled in a protective environment – argon.

To characterize the ground product – ultrafine metallic powders – the following methods were used:

- specific surface area measurement, using the sorptometer KELVIN 1042,
- granulometric analysis, using the laser particle sizer ANALYSETTE 22 COMPACT and image analysis based on the light microscopy and SEM, using the Image-Pro Plus 3.0 system and the corresponding program,
- oxygen content measurement, using the oxygen analyzer Leco.

Figure 6 shows the particle shape of the powders ground. Figure 7 presents the particle size distribution histograms and cumulative distribution fucntions (both in percents by volume) of laser granulometry and image analysis.

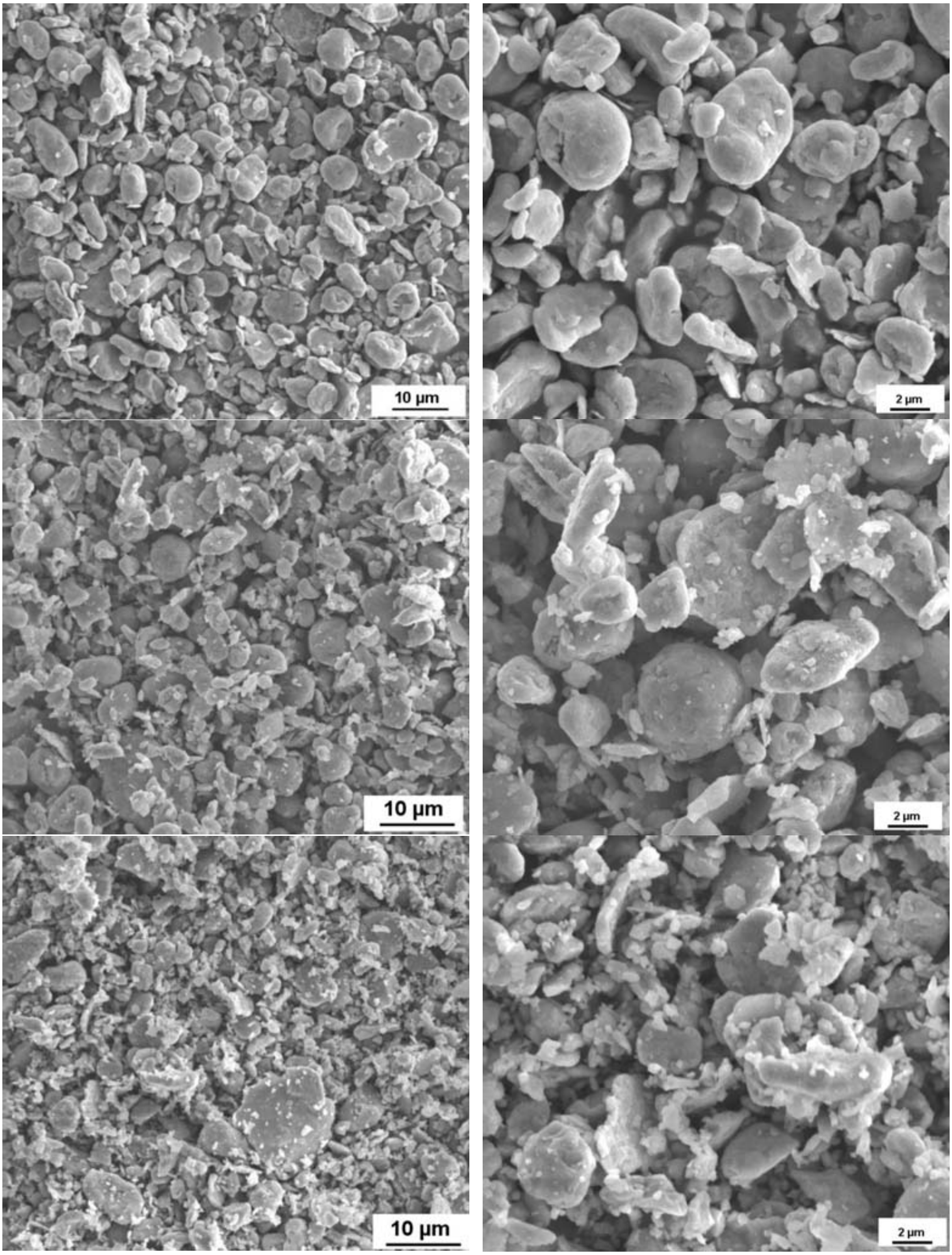

Fig.6. SEM of powder particles: a, b – Alloy; c, d – Fukuda; e, f – Ultimet.

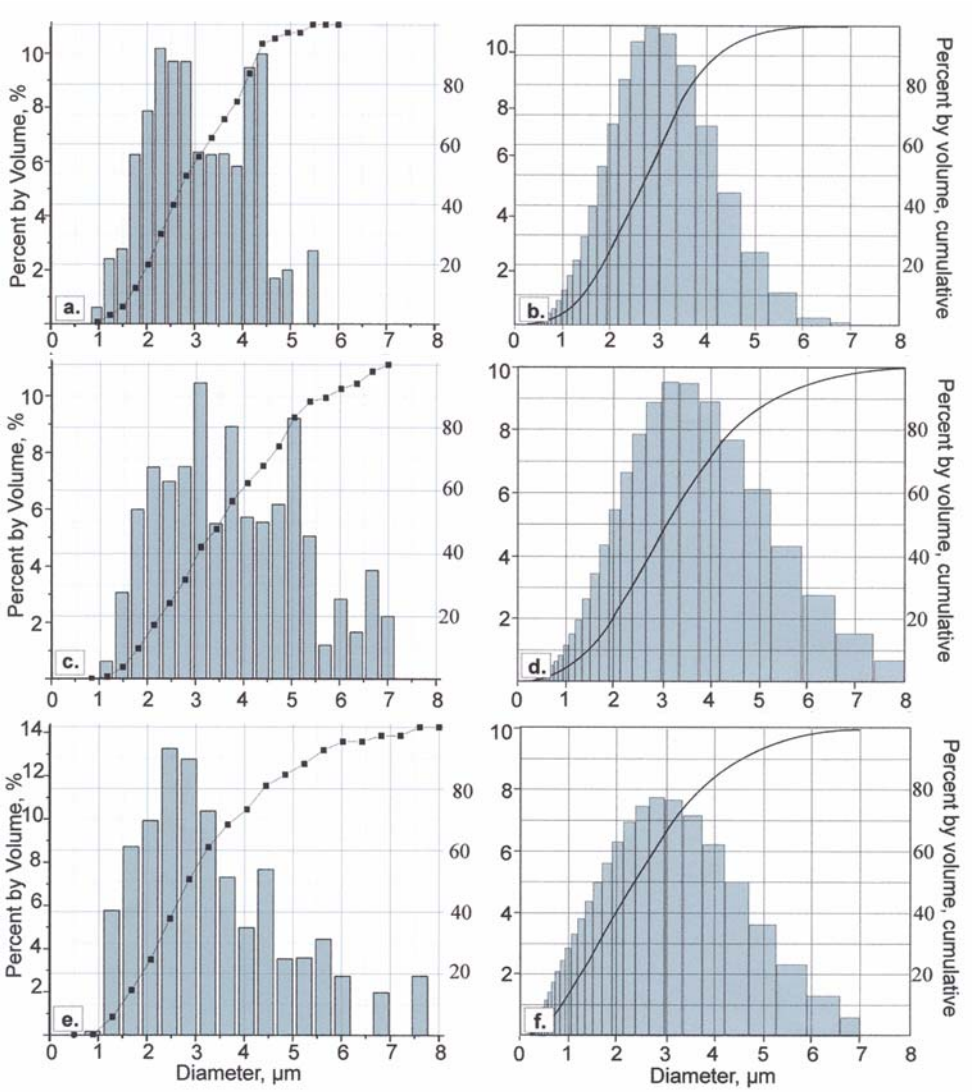

Fig.7. Particle size distribution (a, c, e – laser granulometry; b, d, f – image analyses) of powder particles: a, b – Alloy; c, d – Fukuda; e, f – Ultimet.

Oxygen measurements of disintegrator milled powders

To ascertain the influence of milling and powder particle size reduction on the oxygen content inthe final product, the oxygen content was measured by a LECO analyzer. Regardless of milling in the protective environment i.e. argon, due to the very high specific area of the powder after milling, the oxygen content of the powder increased catastrophically both at milling and during its handling in the air. It is in direct correlation with the increase in the specific surface area of the powder (Tab.4).

Tab.4. Specific surface area and O_2 content of initial and milled powders

Powder type	Initial		After milling	
	Specific surface area [m^2/g]	O_2 content [%]	Specific surface area [m^2/g]	O_2 content [%]
Alloy	0.016	0.06	0.651) / 2.6$^{2)}$	2.7
Fukuda	0.016	0.01	2.871) / 2.4$^{2)}$	5.8
Ultimet	0.044	0.13	3.131) / 3.4$^{2)}$	4.5

1) BET method

2) Laser granulometry

To decrease the O_2 content, oxides were then reduced in hydrogen at temperatures 650, 850 and 100 °C. As it follows from Fig.8, the decrease in of O_2 content is only 5...20 % (maximum by Ni-based powder Alloy-59).

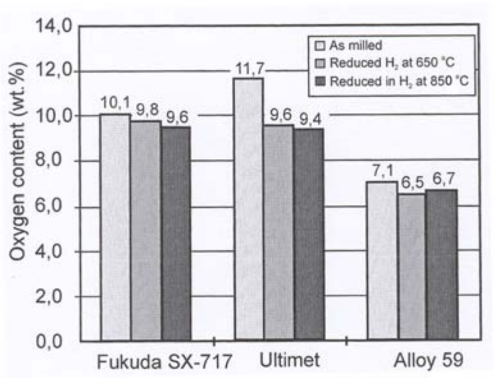

Fig.8. Results of oxygen measurements of milled powders.

Characterization of powder particle size by laser diffraction and image analysis methods

To characterize particle size of the ground product, i.e. ultrafine metallic powders, we used the following methods:

- granulometric analysis, using laser diffraction
- image analysis by the SEM.

As the powders used in this study had a maximum diameter less than 10 µm (micropowders), the traditional sieve analysis did not suit. Commonly, micropowder size is measured by the laser diffraction method [6] or indirectly through the specific surface area obtained from the BET method [7, 8].

To characterize particle size by image analysis, we studied different preparation techniques and compared the results obtained. First, the tablet from the powder was pressed (Fig.9a). Tables surfaces were examined by the SEM and the images obtained were analyzed by the image analysis system. Due to overlapping of powder particles that method was quite time consuming. The same quality results were obtained faster by help of cross-section polishes from the powder studied (Fig.9b). To prepare the cross-section polishes, the micron size particles were mounted into epoxy. Best results were obtained by using only the final steps of grinding (5 µm paper) and polishing (1 µm diamond suspension). The cross-section polishes were examined by the SEM JEOL JSM-840A. An experiment was conducted in the backscattered electron regime (COMPO) and at the magnification 2000x. To study the influence of the number of particles on the end results, different amounts (10 and 40) of analysis fields were used.

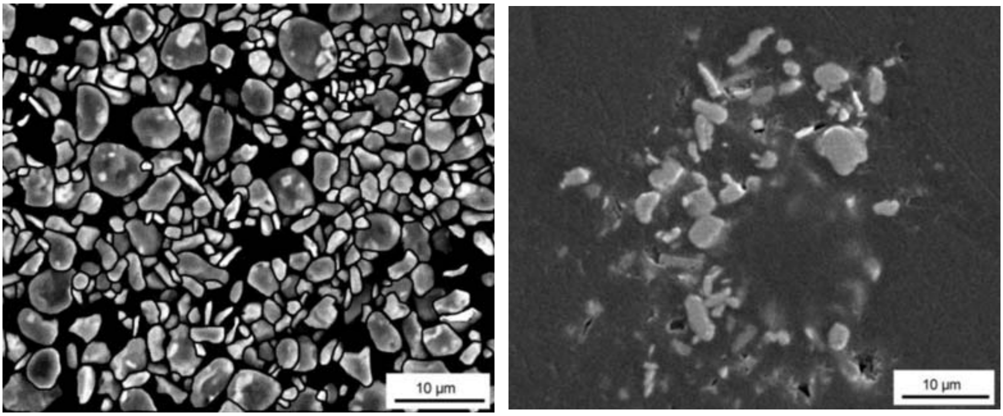

Fig.9 Micrographs of the Alloy powders: a – tablet surface; b – cross-section polish.

Laser diffraction was measured by the laser particle sizer ANALYSETTE 22 COMPACT. The results of laser granulometry and the image analysis method are given in Table 5 and Fig.7.

Tab.5. Results of powder particle size measurements µm

Powder type	Laser granulometry		Image analysis	
	d_m	d_{max}	d_m	d_{max}
Alloy	2.77	6.57	1.89	5.42
Fukuda	3.26	9.16	2.12	6.81
Ultimet	2.58	8.20	1.74	7.23

As it follows from Fig 6 and from our earlier studies [3, 9], the disintegrator milling of ductile materials produces spherical and plate-form particle micropowders (Fig.6.). According to the results presented in Fig.7, the image and laser diffraction analyses show similar results. The particles studied had nearly the same size distributions (especially cumulative) and the largest particles did not exceed 8 – 10 µm.

Characterization of powders particle shape

Particle shape was characterized by their elongation – the aspect ratio. The shape factor aspect ratio *AS* (similar to elongation in the literature) commonly used is calculated by

$$AS = \frac{a}{b}, \qquad (11)$$

where *a* and *b* are the axes of the Legendre ellipse [10, Fig.6].

The Legendre ellipse is an ellipse with the centre in the object's centroid and with the same geometrical moments up to the second order as with the original object area. In the case of circle AS = 1 and for all other shapes AS>1. Particle aspect was calculated by help of both preparation methods, described by Fig.9. No significant differences were found. Figure 10a shows the particle aspect distribution of the micropowders studied. Most of powder particles had a relatively large elongation (mainly close to 2), which is normal in the grinding of ductile materials by collision in the disintegrator mill. Alloy and Ultimet micropowder particles had practically the same aspect distribution, while the Fukuda powder aspect was slightly smaller. Figure 10b demonstrates the dependence of the aspect ratio on the mean diameter (d_m) of micropowder particles. As it follows from Fig.10b, the d_m = 2-3 µm size particles are elongated to a greater extent. At the same time, the aspect had the second smallest local maximum values between the size interval d_m = 5-6 µm. It is probably caused by the nature of the disintegrator grinding of plastic materials. A rise in the elongation of larger particles was caused on particle deformation and on the joining together of smaller particles.

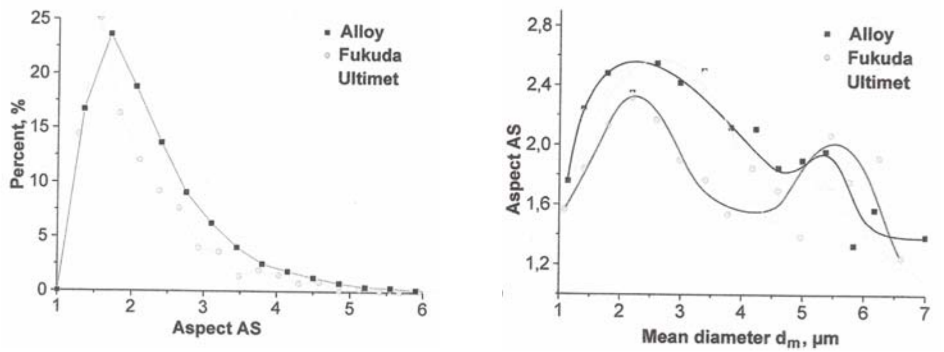

Fig.10. Particle shape factor – aspect distribution (a) and dependence of the aspect on the particle size (b).

CONCLUSIONS

- Based on our theoretical model for size reduction of ductile materials by collision, the possibility of ultrafine powder production from nickel and cobalt – based alloys was ascertained.
- The developed disintegrator milling system with a centrifugal classifier of high separative sensitivity enables us to reduce the size of metallic micropowders to below 5 µm.
- Resulting from our study of comparative powder characterization methods, the proposed powder characteristics for granularity and morphology can adequately describe the properties of powders.

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