Dry Powder Compaction under Ultrasonic Action to Shape Nanostructured Bulk Materials

O. Khasanov, Yu. Pokholkov

Tomsk Polytechnic University

Tomsk, 634050, Russia

khasanov@tpu.ru

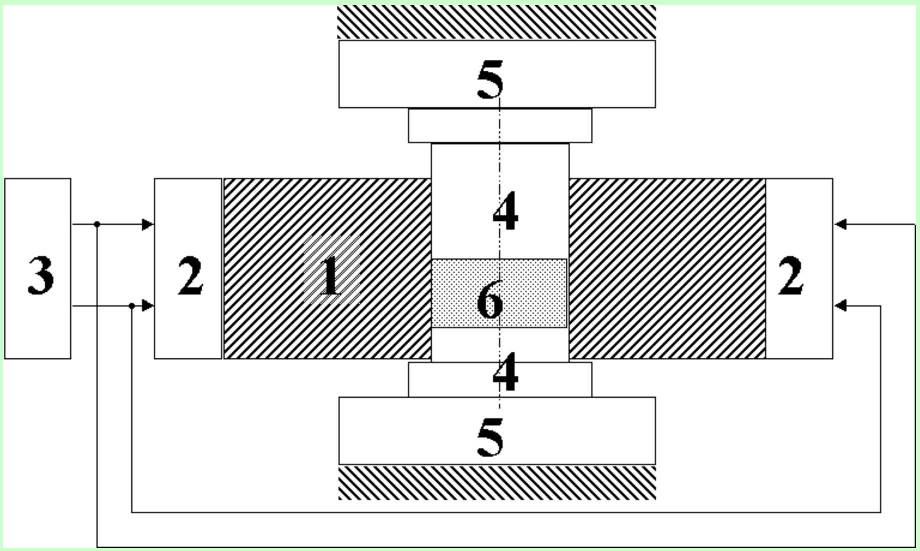
www.tpu.ru/eng/spectr.htm

Important problems of manufacturing nanostructured ceramic products

- Shaping of dust-like nanopowder into required shape with uniform density, without gradients of internal stresses
- High friction forces at particle-particle and particle-die interaction due to high specific area of the nanopowder
- Preventing of grain growth and warping, distortions during green compact sintering to achieve nanoscale structure
- Providing required purity and density of the product
- An economy

The method of dry nanopowder compacting under powerful ultrasonic action

The powerful ultrasonic (PU) action was applied to the die and powder simultaneously with uniaxial compaction pressure **P**:



1 - die-waveguide;

2 - two powerful magnetostrictive transducers converting the electromagnetic oscillations with frequency about ***f*** = 20 kHz and generator power of ***W*** = 1, 2 or 3 kW into ultrasonic vibrations;

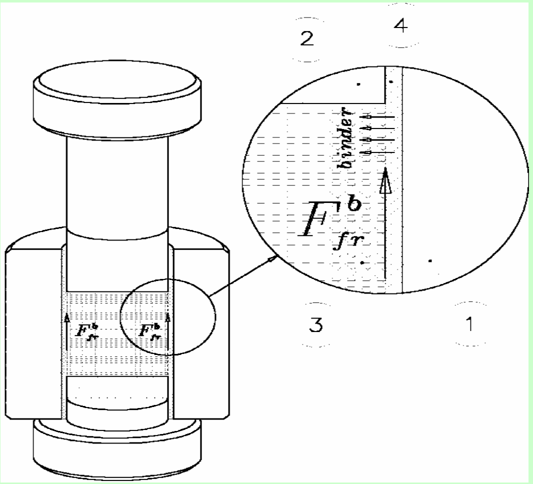
3 - HF-generator;

4 - punches;

5 - hydraulic press;

6- powder

Ultrasonic action during nanopowder compaction decreases the wall friction, interparticle friction



The case of common uniaxial compaction using *a binder*

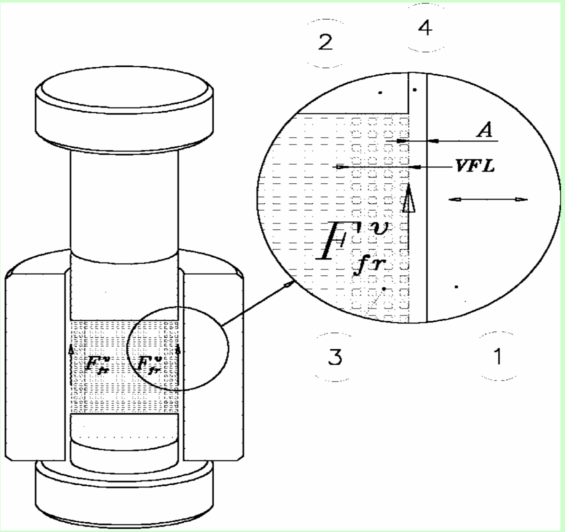
A wall friction force F_{fr}^{b} is less than at dry compaction due to lubrication of die wall layer

1 - die

2 - punch

3 - nanopowder

4 - binder



The case of *dry* uniaxial compaction *under ultrasonic vibrations*

A wall friction force F_{fr}^{v} is less than at compaction using binder due to vibrofluidization layer near a die wall.

No any organic impurities inside of the nanopowder, as a result no residual porosity, which is caused by their burnout during sintering.

4 - vibrating die wall surface with an amplitude **A ~ W**

VFL - vibrofluidized layer

The uniform packing of nanoparticles occurs when acoustic flows are absent,

i.e. when vibratory displacement of nanoparticles ***a*** does not exceed their average size ***d:***

$|\boldsymbol{a}| \leq \boldsymbol{d}$

The critical intensity (specific capacity) of PU oscillations I_c at which this condition is satisfied:

$\mathbf{I_C = (\rho c\, \omega^2 d^2)}/2$

ρ - density, **c -** sound velocity of the green compact; $\omega = 2\pi f$ - oscillation frequency.

The powder is a gas-dispersive medium when compaction pressure $\mathbf{P < P_c}$.

$\mathbf{P_c}$ **is critical compaction pressure** at which interparticle bonds become stronger and conditions of sound propagation through the powder as in solid porous body are created

[Khasanov et.al., MRS Symp. Proc., V.520, 1998, p. 77- 82].

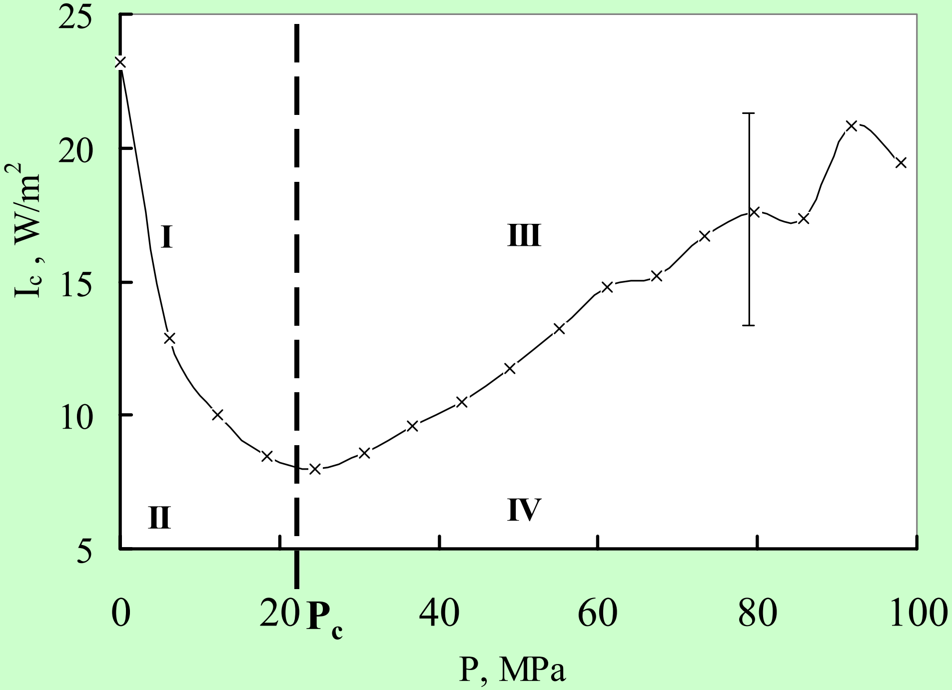
PU-action on nanopowder during compacting can be characterized by 4 types defined by parameters P_C, I_C:

I) $P < P_C$, $I > I_C$ - agglomerate crushing, nanoparticle activation under the conditions of acoustic flows;

II) $P < P_C$, $I < I_C$ - dense packing of nanoparticles on the initial compacting stages;

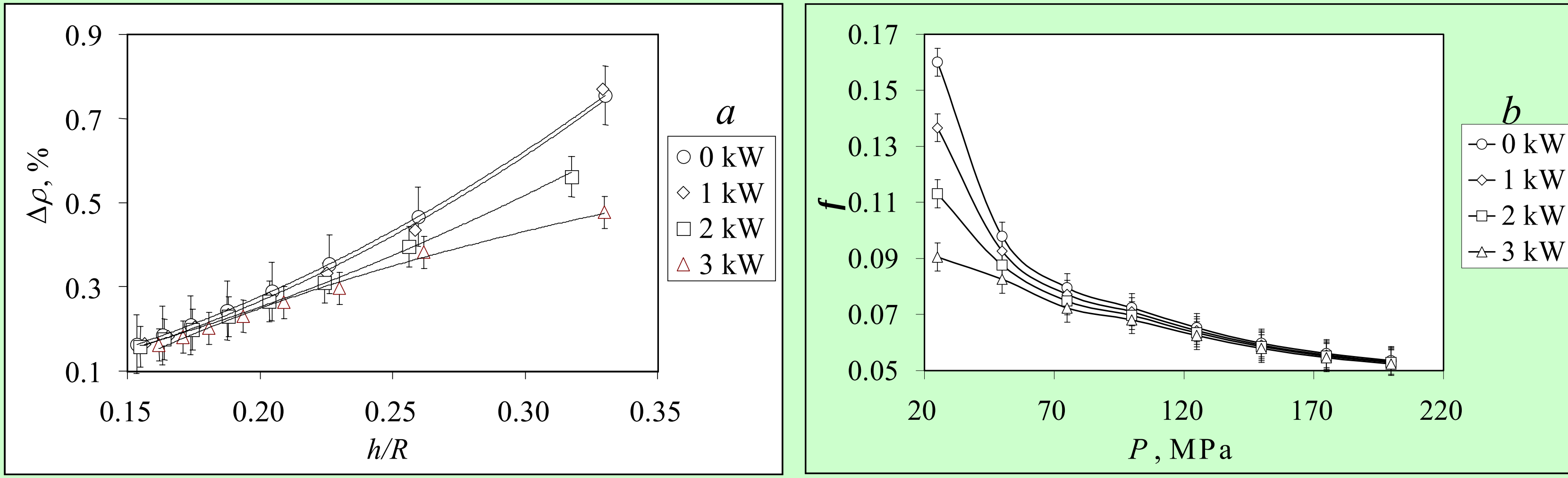
III) $P > P_C$, $I > I_C$ - dense compacting of nanopowder with a change of its elastoplastic properties;

IV) $P > P_C$, $I < I_C$ - compacting of nanopowder without plastic deformation of nanoparticles to preserve the nanostructure of the green compact.

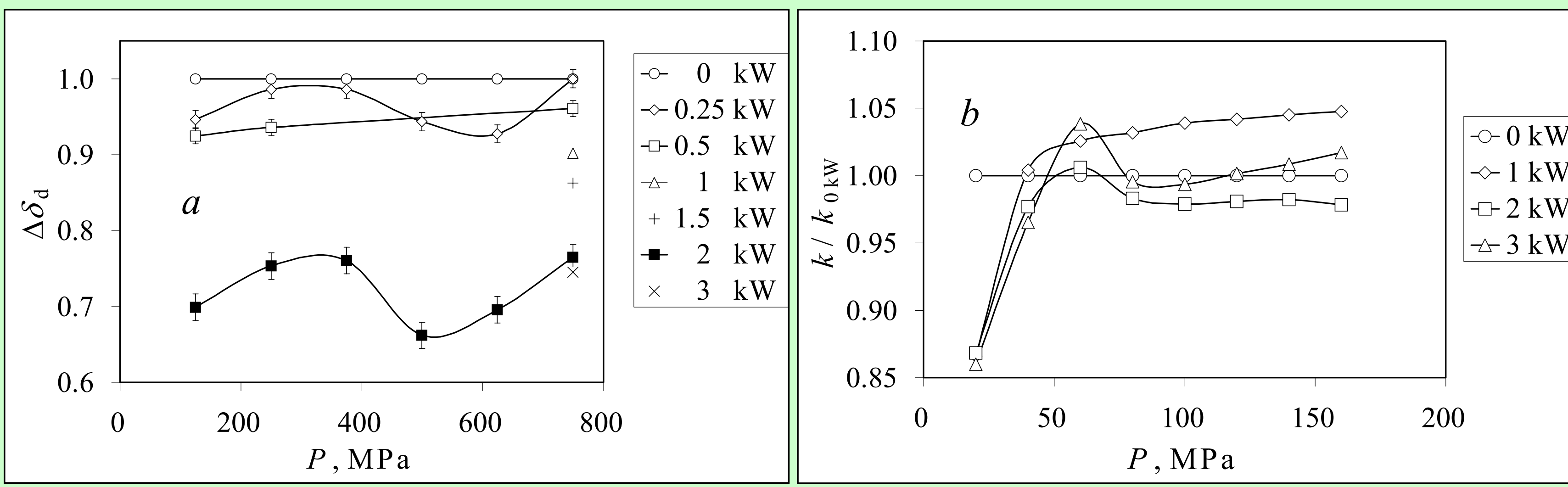


Calculated dependence I_c vs P from experimental data of ultrasonic compaction of dry zirconia nanopowder YSZ

[Khasanov et.al., MRS Symp. Proc., V.520, 1998, p. 77- 82].



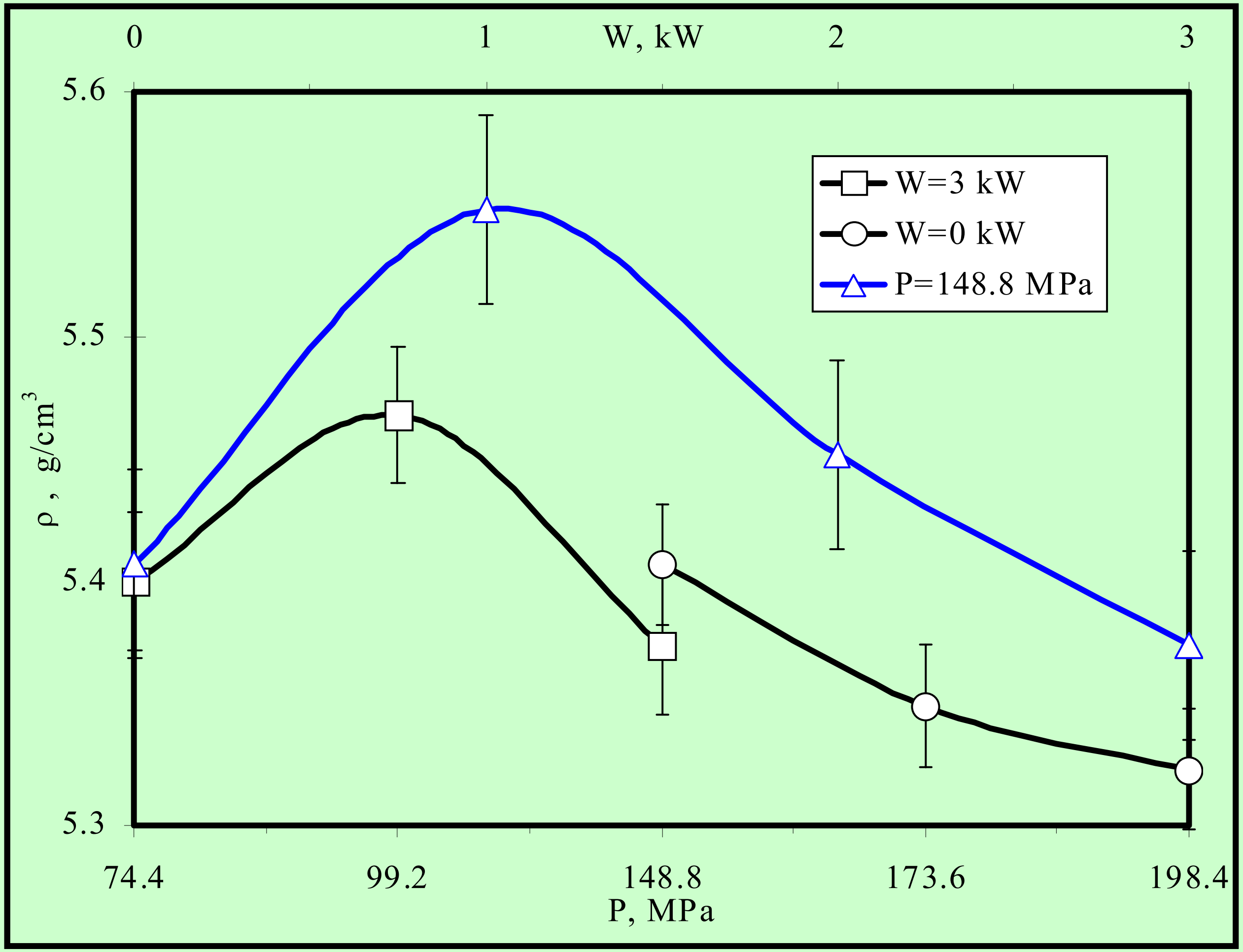
Influence of PUA on the density differential $\Delta\rho$ throughout the green compact height (a) and on the coefficient of die-wall friction f (b) for YSZ nanopowder



Variations of springback in a diameter $\Delta\delta_d$ of YSZ nanopowder and interparticle friction coefficient of the sonicated green compacts relative to non-sonicated ones (b)

[Khasanov et al., Key Eng. Mat., Vols. 270-273 (2004), p.73-76].

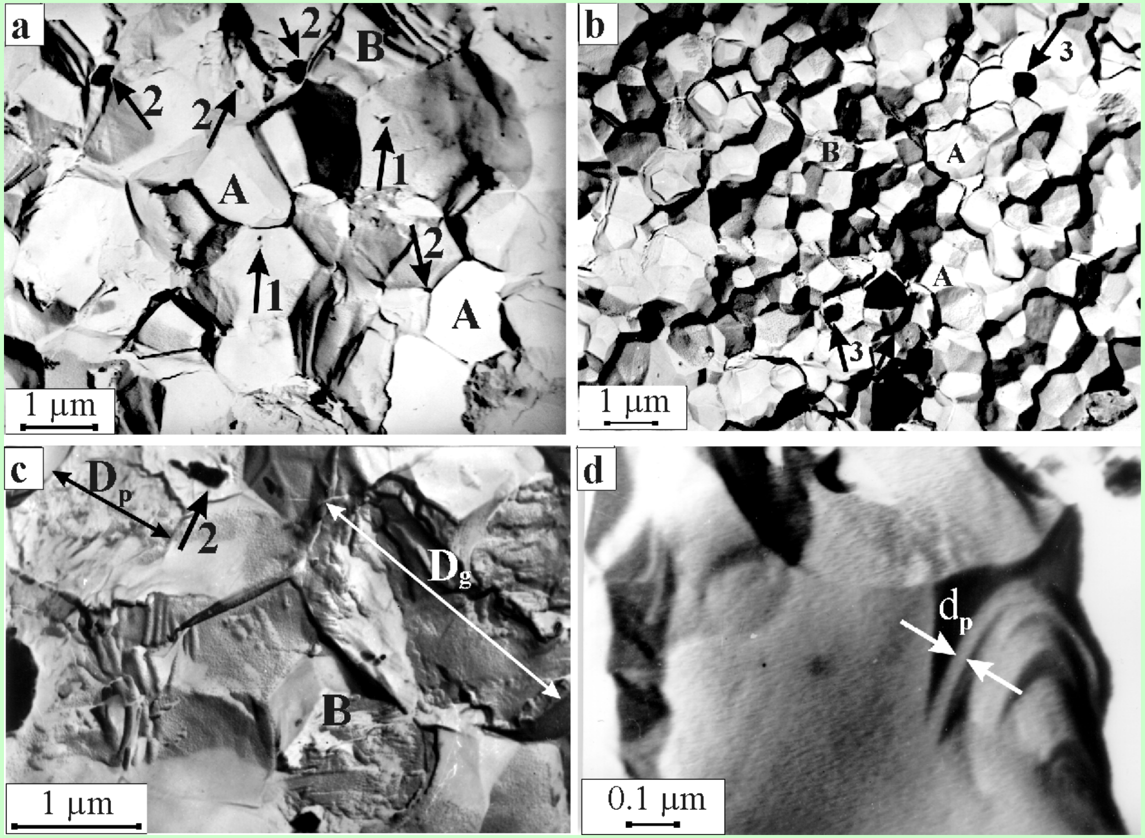
Compaction under ultrasonic action influences the *density* of *sintered ceramics*



The $Ba_{0.6}Sr_{0.3}Ca_{0.1}TiO_3$. **ceramics density** versus P under PU-action and without it at **nanopowder compaction**, and versus W at P = 148.8 MPa.

Thin discs of the green compacts (D = 50 mm, h = 1.2 mm) **were broken at compaction without US-action at P < 148.8 MPa**, **whereas under US-action they could be compacted without failure at P = 74.4 MPa and more.**

Compaction under ultrasonic action
influences the *grain growth* during sintering and leads to *formation of nanostructure*



The TEM - images of the zirconia ceramics fracture surface *[Khasanov, et al., In: Fracture Mechanics of Ceramics, V.13, Kluwer Academic/Plenum Publishers, 2002, p. 503 – 512]*

a, c - ceramics sintered after **common** compaction;

b, d - ceramics sintered after **ultrasonic** compaction.

Denotations: A - the facet surface with smooth fractures; B - the facets with a pattern; D_g – the grain; D_p – the pack; d_p – the plates-flakes. 1 - the extracted ceramic parts located in a body of spall facets; 2 - on their boundaries or in a joint of boundaries; 3 – the ceramic parts occupying the whole spall facets.

Compaction under ultrasonic action
influences the *grain growth* during sintering and leads to *formation of nanostructure*

The SEM micrographs of the PZT ceramics spall surfaces

sintering after **ultrasonic** compaction — sintering after **common** compaction

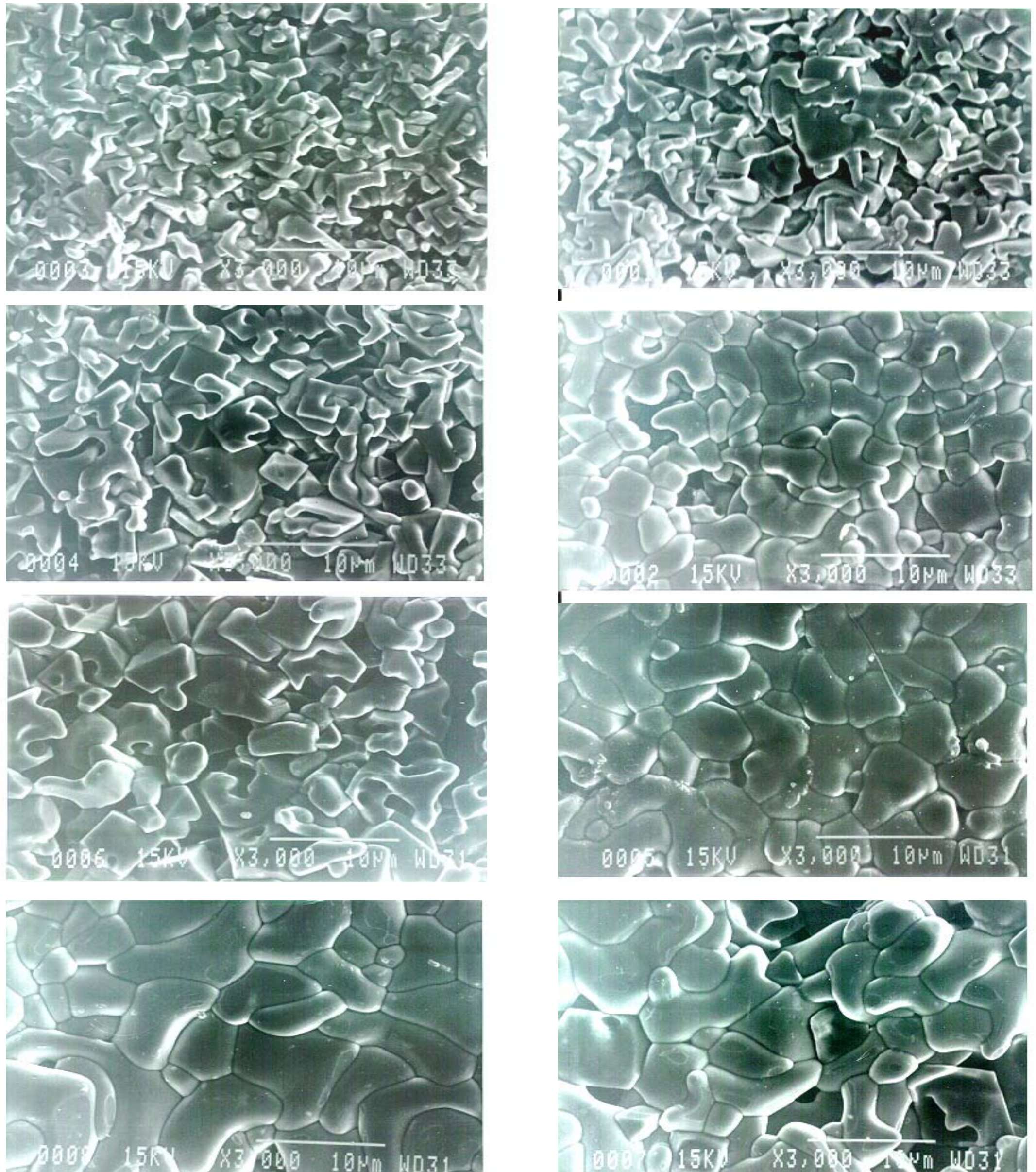
The temperature of sintering:

1100°C

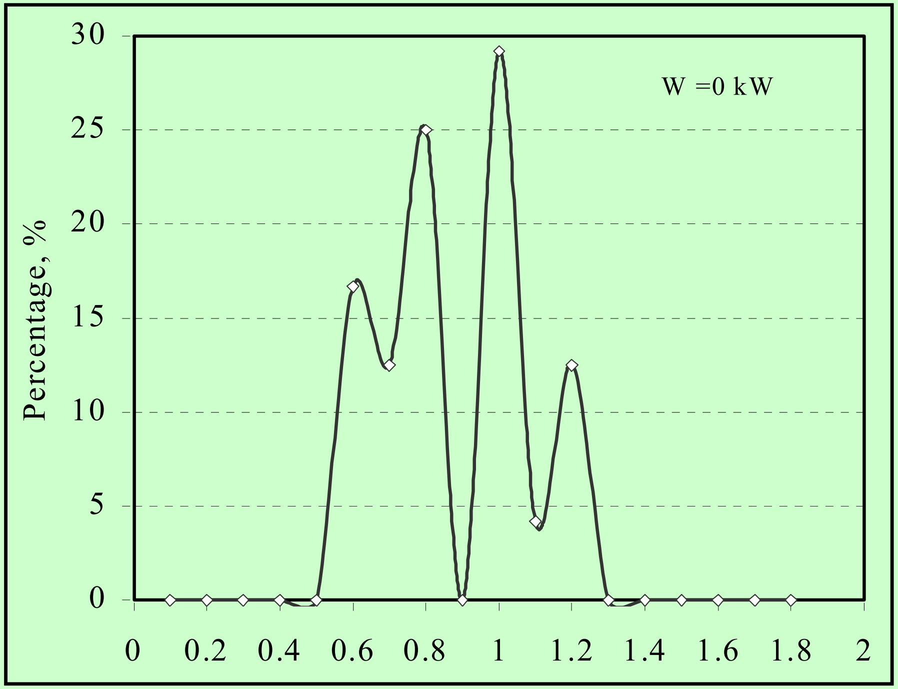
1150°C

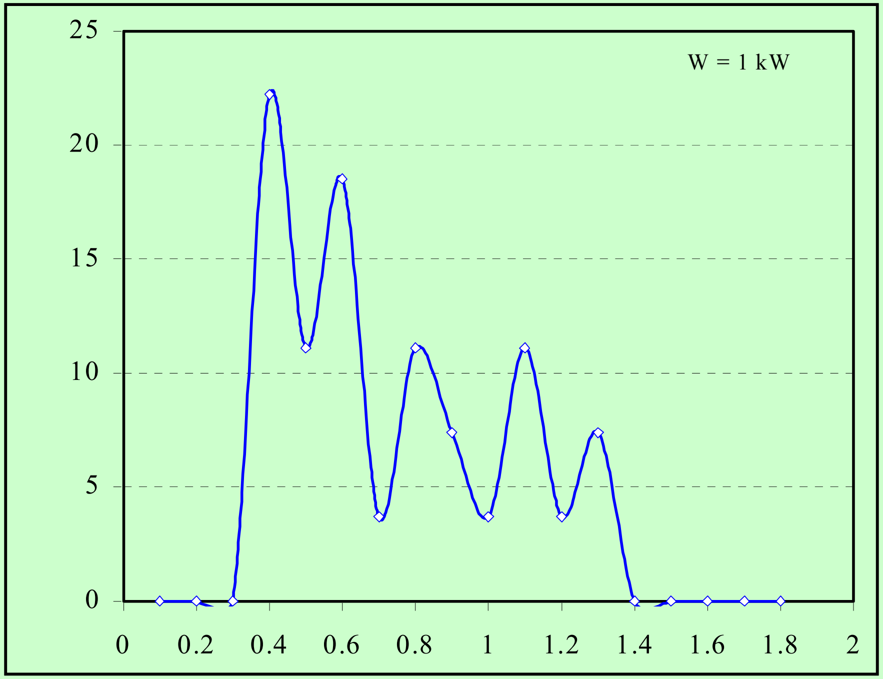
1200°C

1250°C



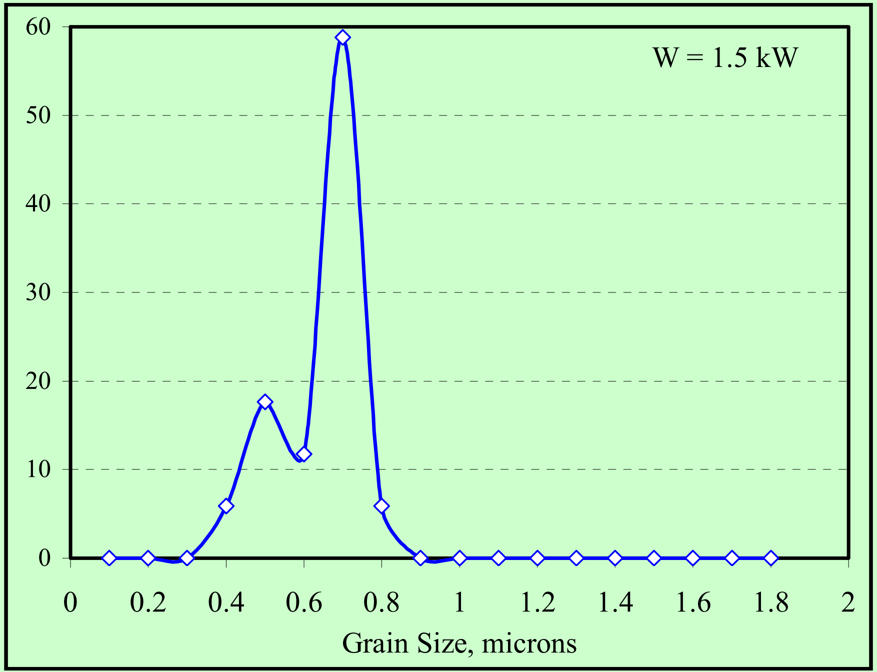
Compaction under ultrasonic action influences the *grain sizes of sintered ceramics*





The granularity of $(Ba,Sr,Ca)TiO_3$ ceramics sintered at 1450°C. Green compacts were compacted without PU-action (W = 0 kW), and under PU-action (W=1 kW; W=1.5 kW).

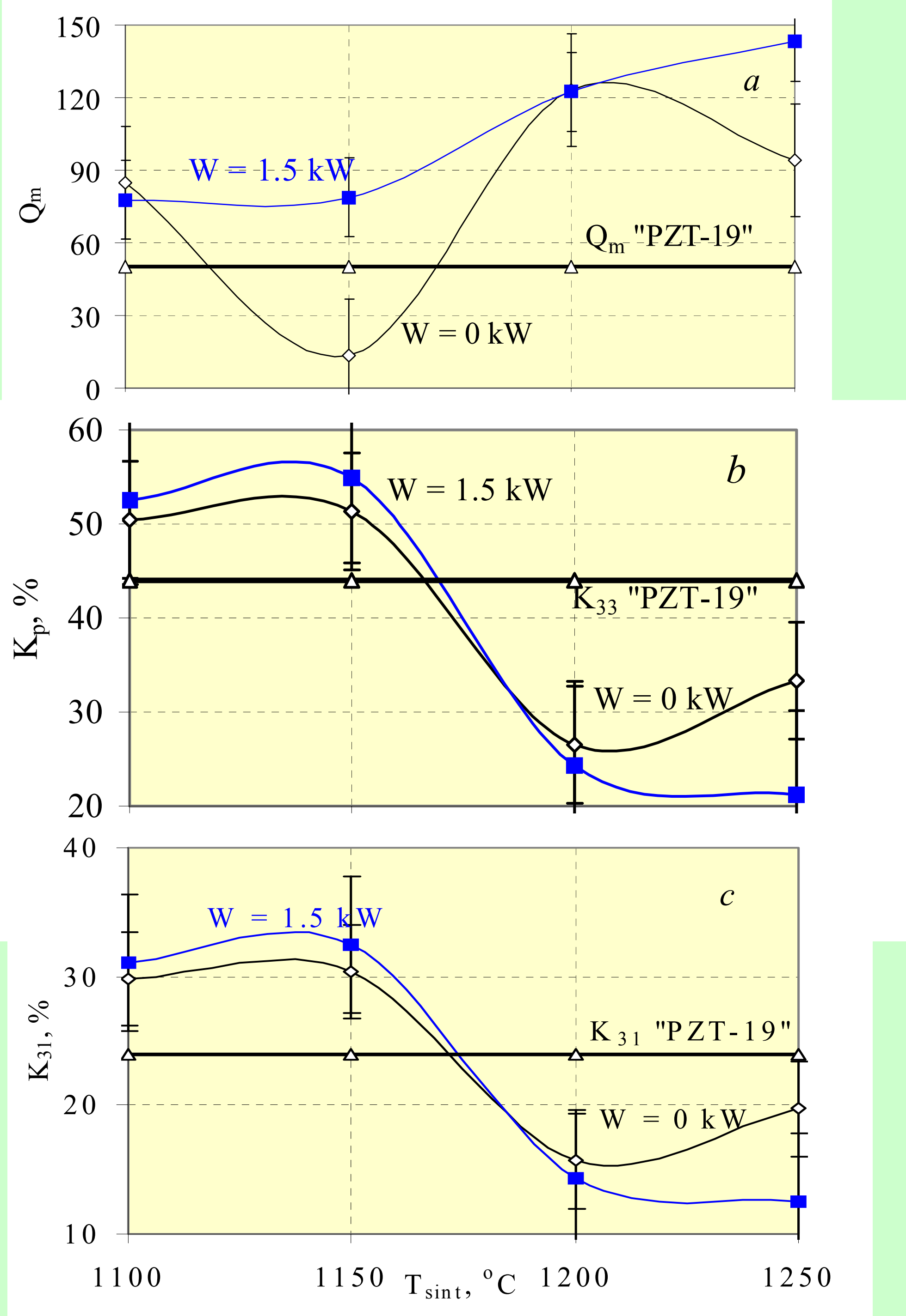
Ultrasonic compaction promotes sintered ceramics with grains less than 1 μm and with more uniform size distribution.



Compaction under ultrasonic action influences the *piezoelectric properties* of PZT ceramics

The electromechanical quality factor Q_m (*a*), piezoelectric coupling coefficients K_p (*b*), K_{31} (*c*) vs sintering temperature for PZT ceramics compacted under ultrasonic action (**W = 1.5 kW**) and without it (W = 0 kW).

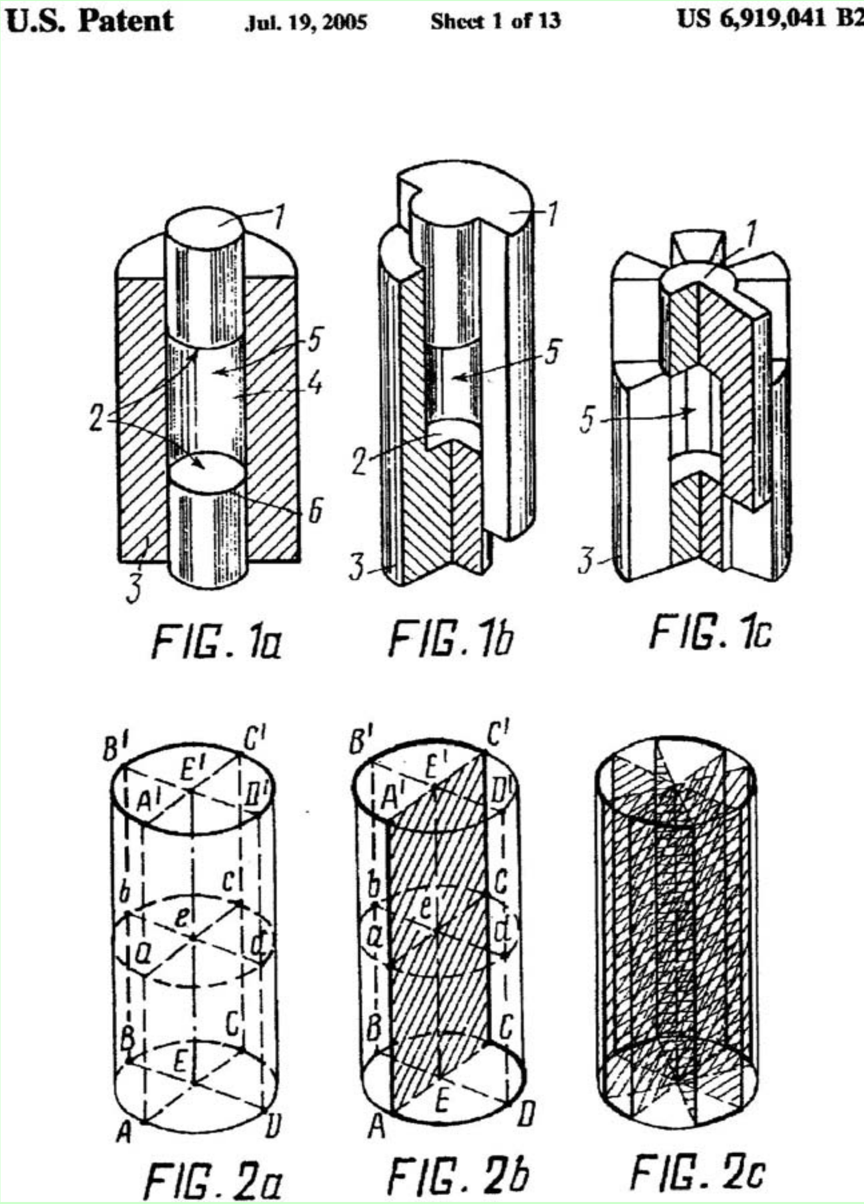
The "PZT-19" – values for the same industrial piezoceramics *[Physical Encyclopaedia, V.4, Moscow, BRE Publisher, 1994, 704 p.]*



The method of compaction under powerful ultrasonic action can be combined with another developed method of dry powder compaction - *“the collector method”*

The “collector method” uses the special design of the die, where the active and passive shaping surfaces are combined in the one shaping member of the mold according to the principle of minimizing the particle-die friction forces and the certain rules of mutual moving of such shaping members.

[RF Patent No.2225280 (03/12/2004);
Eurasian Patent No.005325 (02/24/2005);
US Patent No.6919041 B2 (19/07/2005);
Europatent Application No. 02805039.1 (17/03/2004), publication No.1459823, European Patent Bulletin No.2004/39, 22.09.2004].

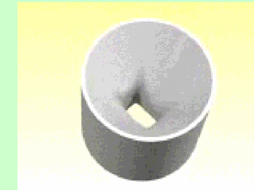


Various ceramic products developed using the "ultrasonic" and "collector" methods of dry nanopowder compaction

Impellers for the gasoline pump from Y-TZP nanoceramics

Seals for the hydraulic pumps of automobile engines from Y-TZP nanoceramics

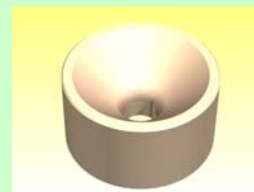
Nanoceramic cases of microwave duplexers from $(Ba,W)TiO_3$



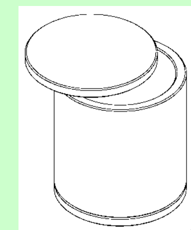
Rectangular die from Y-TZP nanoceramics for cable production

Die for first drawing from Y-TZP nanoceramics

Die for cable drawing from Y-TZP nanoceramics



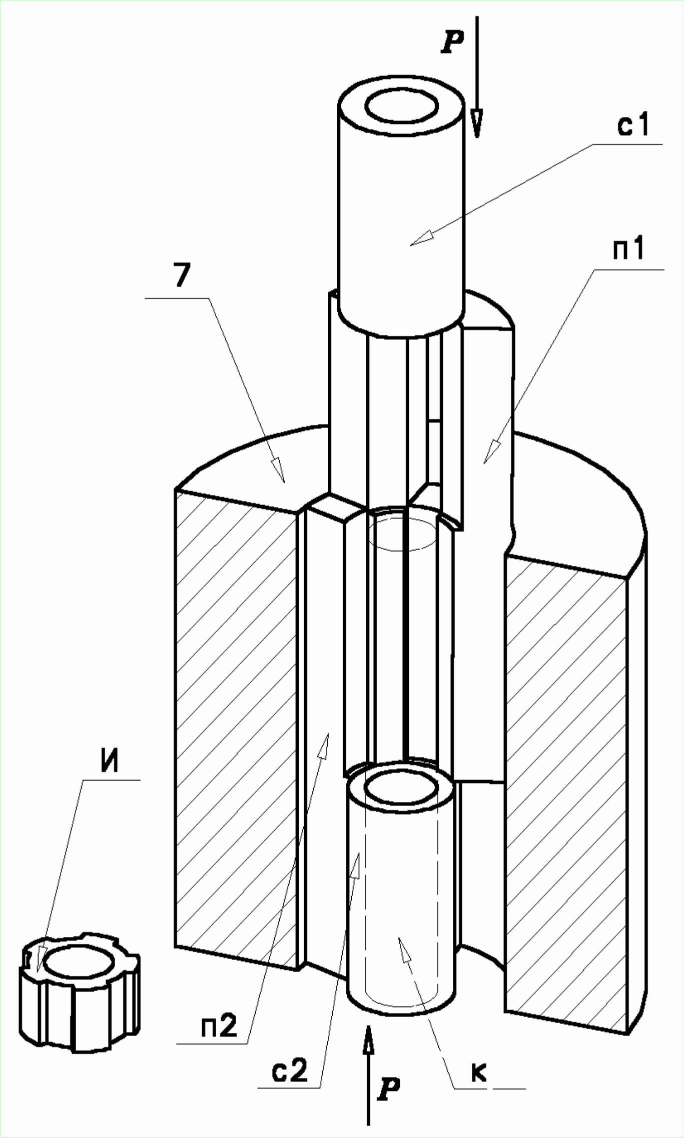
Extrusion head from Y-TZP nanoceramics



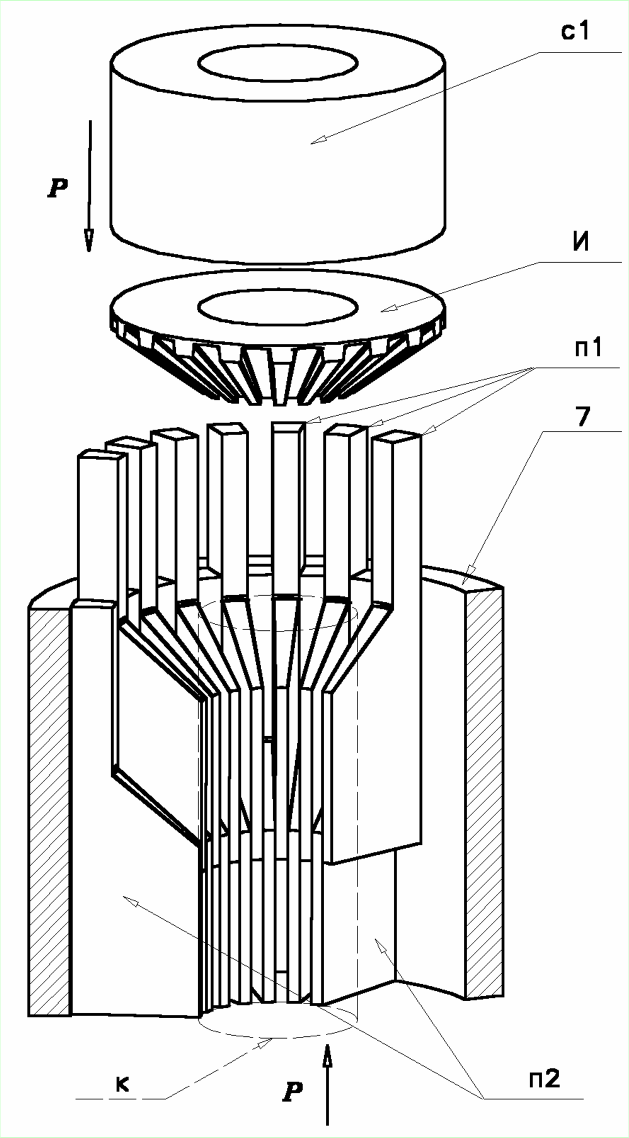
HTSC-shield from ceramic $YBa_2Cu_3O_{7-x}$ (50 mm in diameter and 50 mm in height)

The targets from ZrB_2 for magnetron sputtering

The mold to compact the impeller



The mold to compact the conic gear



Achieved technical characteristics of the developed pilot products

- Grain sizes in sintered articles from nanopowders ZrO_2-Y_2O_3, $(Ba_{0,6}Sr_{0,3}Ca_{0,1})TiO_3$, $(Ba,W)TiO_3$ are 100-300 nm, density up to 99% with uniform distribution in the article volume
- The required stringent tolerances of nanoceramic cases of microwave duplexers from $(Ba,W)TiO_3$ 0.5 - 1 microns were achieved.
- The structural zirconia nanoceramics has tetragonal phase (Y-TZP), bending strength up to 600 MPa in conjunction with fracture toughness of 14 $MPa*m^{1/2}$, hardness of 12 GPa. The wear resistance of hydraulic pump seals is greater by 20 fold in comparison with standard material NAMI GSTAF-40.
- Piezoelectric ceramics PZT had Q-factor of 80 at resonance frequency of 234 kHz, mechanical coupling coefficient of 0.54 which are higher than ones for similar industrial PZT.
- The Q-factor of cavity resonator from HTSC ceramics $YBa_2Cu_3O_{7-x}$ was 2700 at frequency of 10 GHz and T=4.2 K. The shielding coefficient of the external field of 10 kA/m with frequency of 10 kHz at T=78 K was $k \geq 100$. The sensitivity of ceramic HTSC SQUIDs was 1-2 $\mu V/\Phi_0$ at 80 K.

Basic peculiarities of the developed methods

The use of PU-action at compaction of dry nanopowders allows

- ultrasonic activation and disaggregation of nanopowder;
- packing of nanostructured green compacts with homogeneous density even of complicated shape;
- excluding the application of binders as cause of impurities and additional residual porosity in ceramics
- inhibition of the grain growth during ceramics sintering and leads to formation of nanostructure.

The technology advantages

- The rise of economical efficiency of production process due to reduction of technological operations and their energy intensity as well as application of relative simple industrial equipment: conventional hydraulic presses, furnaces, ultrasonic generators.
- Improvement of ceramics quality

Conclusions

The dry uniaxial pressing of nanostructured powders under powerful ultrasound action and collector compaction are effective methods for net-shaping of structural and functional nanoceramics because of they allows

- uniform density distribution in articles having complex shape owing to even packaging particles when their vibration displacement is comparable with a particle or agglomerate size;
- decrease of interparticle and die-wall friction forces;
- fracture of agglomerates during compaction;
- nanostructure formation at sintering green compacts;
- elimination of plasticizer and binder application as potential sources of impurities.