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#### **KEYWORDS**:

Main subjects: plasma spraying, plasma and dispersed particles flow visualization Fluid: high speed plasma flow, high temperature multiphase flow Visualization method(s): high speed camera imaging Other keywords: mineral fiber production, DC plasma torch

#### **ABSTRACT**:

The research presents results of experimental and numerical investigation on behaviour of dispersed particles, granules, also melted ceramic material domains outflowing from the atmospheric pressure plasma torch. High thermal resistant granules and fibre have been produced from aluminium oxide, copper oxide, zeolites, dolomite and its mixture with quartz sand employing a specific atmospheric pressure plasma spray technique. Produced fibre can be used as high temperature insulation, as filter for ultrafine particles or as a concrete additive for strenghtening of building materials. Experimental installation was developed for operating by feeding air, nitrogen or hydrocarbon containing gases mixed with dispersed particles. The power of plasma torch was in the range of 53 - 85 kW, the mean temperature of gas leaving the reactor -1800 - 3000 K, plasma flow velocity in the outlet -500 - 1300 m/s. A high-speed RedLake MotionPro video camera was used for instantaneous imaging of plasma spray process. The velocity of plasma jet is supersonic: the shock diamonds of the flow visually can be observed. Observations by camera suggest that multiphase jet in exhaust plasma chemical reactor nozzle consists of melted domains, grains of different sizes and fiber filaments. Experimental tests showed that zeolites powder, injected into high temperature jet, is melted very quickly. The high-speed imaging let to distinguish moving structures, determine the spraying stream geometry, and calculate the magnitude and speed of sprayed materials.

The interaction of plasma jet and hard ceramic particles were numerically investigated by means of "Jets&Poudres" software, applied to simulate plasma spraying processes.

Performed experimental and analytical studies showed that process of plasma melting and conversion of melt into microfiber depend on following main factors: - plasma generator characteristics and operating regimes; - plasma flow formation, spraying characteristics; - plasma forming gas and powder injection mode and place; - size and fraction of sprayed particles, the injection rate parameters and specific details of ceramic fiber formation.

**INTRODUCTION.** Manufacture processes of refractory materials and high temperature thermal insulation materials are linked to labor-consuming, preliminary mechanical processing of source raw material and to great energy consumption for their thermal treatment. On-stream technological process is required for these purposes. In connection with the rising demand of high quality thermal insulation materials working at high temperatures the new methods of their manufacture are sought. One of such methods at present time is plasma technology having good prospects. This technology enables joining together the processes of melting of raw material and manufacture of material required (high temperature insulation fiber, nano-dispersed particles or spherical ceramic granules) and so forming a single process, using kinetic energy of high temperature flow generated by the plasma generator [1, 2].

Plasma-assisted melting and conversion of melt into different products have become key process in the scale of many applications. Authors of present paper referred elsewhere [2] that plasma spraying may affect the formation of fine particles or granules. The process of spray pyrolysis may occur during plasma spraying as a dominant process of composition changes during plasma spraying. The employment of plasma spray pyrolysis may be directed at the fabrication of variety of ceramic coatings, production of fine mineral fiber and granules, synthesis of micro- and nanostructured particles and plasma polymer products [3, 4].

Currently, methods of plasma processing of inorganic refractory materials are investigated thorough, mechanical, chemical and tribological properties of final product are well studied [5–6]. However, in a worldwide scientific-technical literature there is a lack of data regarding the issue of mechanisms of structure formation in plasma spray processes, it is not determined how the parameters of process influence the quality, elemental composition, surface structure, thickness, adhesion. It is not clear enough how it is possible while changing the flow regime of particles and

### V. GRIGAITIENĖ, R. KĖŽELIS, V. VALINČIUS, M. MILIEŠKA

gas mixture to obtain the desirable shape and structure of product, porosity, physical and chemical properties. There is lack of data on the investigation of arc plasma spray processes at atmospheric pressure.

Research on gas dynamic and thermal processes during plasma spray pyrolysis is a complicated task. Research is impeded by the fact that at the same time in plasma jet occur many different phenomena: chemical reactions, dissociation, ionization, fusion of dispersed particles, erosion of their surfaces, etc. Intensity of processes depends not only on temperature, but also on the concentration, diffusion of particles, operational regime of plasma generator, heat conduction, roughness, etc. of wall. Coefficients of heat-mass transfer and gas dynamic characteristics in plasma jet are a function of many variables, and there is no possibility to mathematically simulate a process. Therefore research of processes is carried out using both experimental and numerical methods.

Kinetic and potential energy of plasma flows and jets is extensively employed in the synthesis of hard melt ceramic structures, therefore the interaction of plasma jet, dispersed particles and melted material is very important [7]. The process of turbulent mixing in high temperature fluid jets and transport of dispersed particles has been noticed by number of authors [7–9, etc.] in recent years. Reporting various methods of improving mixing of particles in turbulent jet they also indicated that kinetic energy of turbulent jets and flows is suitable to a number of applications, including plasma synthesis of coatings, fiber, micro- and nanostructured particles. Therefore this study presents results of investigation on behaviour of melted and concentrated ceramic material solutions in the entrance region of plasma jet outflowing from plasma chemical reactor (PCR) exhaust nozzle. High thermal resistant granules, fibre and fine particles have been produced from zeolites.

**EXPERIMENTS.** The experimental low temperature plasma spraying equipment (Fig. 1.) used in this research was designed and constructed in Lithuanian Energy Institute. It consists of following main systems: electricity supplies (1-3), plasma torch with a stream reactor for powder injection (10), gas supply and monitoring system (11-14), cooling system (15, 16) and operation control and data monitoring system (4-9). Continual data monitoring of operating plasma torch allows the test bench functioning.

Experiments were performed using a linear direct current (DC) plasma torch with button type cathode and stepformed anode. The similar plasma spray torch 100 kW of power is analyzed in details elsewhere in [10,11]. The operating parameters of such plasma torch are very stable. Mean-mass outlet jet temperature and velocity was determined from the heat balance. The capacity of plasma torch, mass flow rate of gases, cooling water flow rate and its temperature were measured and gas enthalpy calculated. Parameters of the plasma torch ranged within the following limits: power (P) – 70 – 80 kW, arc current (I) – 150-200 A, voltage (U) – 300 – 430 V, total gas flow rate (G) – 17 – 23 g·s<sup>-1</sup> (main gas flow rate throw plasma torch – 17 g·s<sup>-1</sup>, additional – 0 – 5 g·s<sup>-1</sup>, propane butane gas – 0,12 g·s<sup>-1</sup>). Mean-mass outlet plasma temperature is 3000 – 3700 K, outlet average velocity – 900 – 1500 ms<sup>-1</sup>, the plasma forming gas is air.

A specific plasma chemical reactor (Fig. 2.) for melting of zeolites and fiber production was constructed. It is directly connected to the plasma torch anode and consists of one straight section having length of 0.1 m and four sections 0.05 m of length. All sections are made of stainless steel and are cooled by water. The internal diameter of the reactor is equal to 0.015 m and the total length is 0.3 meter. The outlet section inner diameter varies from 0.01 to 0.015m. Such design enables to get a very high outlet flow velocity (supersonic jet).

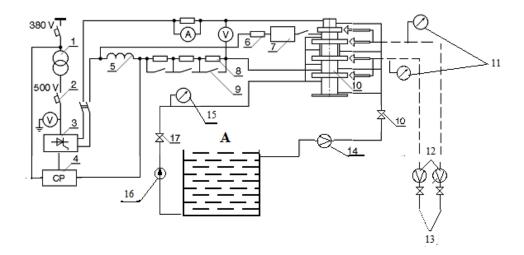


Fig.1. Schematic presentation of experimental set-up. Explanation is given in the text.

Three different regimes of plasma source were applied. The main process parameters are given in Table 1.

As dispersed material for the plasma treatment was powder of waste oil-cracking catalyst (zeolite) with following chemical composition [mass %]:  $Al_2O_3 - 40.9$ ,  $SiO_2 - 55.2$ ,  $Fe_2O_3 - 0.9$ ,  $TiO_2 - 1.4$ , CaO - 0.5, MgO - 0.49,  $Na_2O - 0.2$ . The particle size was approximately 50 µm, density - 830 kg·m<sup>-3</sup>. A non-equilibrium plasma flow confined by walls of a funnel-shaped channel of the reactor is considered to be converted to stable after injecting powder material with carrier-gas in the form of a tangential flow along the axis before the nozzle.

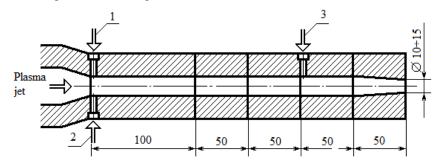


Fig.2. Scheme of experimental plasma chemical reactor channel. 1 – injection of powder precursor, 2 – injection of propane-butane, 3 – injection of additional gas

Series No.	1	2	3
Outflow diameter, ·10 <sup>-3</sup> m	10	13	15
Flow velocity, m·s <sup>-1</sup>	1515	1092	898
Power, kW	67	69	74
Plasma forming gas flow rate, g·s <sup>-1</sup>	14.8	14	14
Carrier gas flow rate, g·s <sup>-1</sup>	1	1	1
Additional air flow rate, $g \cdot s^{-1}$	4.5	3.5	3.8
Flow rate of propane-butane, g·s <sup>-1</sup>	0.5	0.5	0.5
Mean-mass gas temperature in the reactor	1892	2522	2734
exhaust cross section, K			

Plasma forming gas was supplied via the intermediate ring into the reacting arc zone. The propane-butane gas was injected via the second ring at the end of anode or in the entrance region of the reactor. Therefore the length of plasma torch has increased. Precursor powder was injected into the air plasma flow inside the reactor (Fig.2., position1). The investigation of formation of multi-phase flow parameters has been performed also experimentally. A high-speed

RedLake MotionPro video camera was used for instantaneous imaging of plasma spray process. A fast, 12 bits CMOS camera (MotionPro from Redlake) equipped with a zoom lens and a neutral density filter is used to visualize the plasma jet with dispersed particles emission. The camera exposure time is  $2 - 43 \ \mu s$ .

The special software "Jets&Poudres" [12, 13] was applied for a plasma spraying modeling. The software has been used to solve Navier-Stokes and energy equation based on the dynamic  $k-\varepsilon$  model for the fluid jet. For the boundary conditions, it is assumed that the plasma jet exits in space where the pressure is atmospheric. The outflow diameter was 0,01 m and the boundary conditions were as follows: 1) constant temperature of fluid flowing on the entrance; 2) zero normal gradients at the inflow; 3) rectangular distribution of velocities temperatures; and 4) constant of the inflowing jet turbulence.

The diameter and cleanliness of the produced fiber was evaluated using a scanning electron microscopy (SEM) observation method.

**RESULTS AND DISCUSSION.** Numerical research of two-phase high temperature jet was carried out using "Jets&Poudres" software, created on the basis of General Mixing (Genmix) software, however, improved and applied to model the specific plasma jet. When the parameters of plasma jet are achieved as desirable, the initial powder is injected into the flow. Deformation of plasma jet flow fields is not considered while modeling and calculating, inflow profiles of temperatures and velocities are rectangular. Plasma jet flows steady in one direction, without recirculation of diffusion effects.

After the mixing with plasma jet, solid particles have lower temperature than the plasma jet, and a certain time is needed to heat them up. Heating of particles in plasma jet occur releasing heat by convection, whereas inside particles heat is transferred by conduction. As it can be seen from fields of temperatures in Fig.3., the temperature of dispersed particles approximately at the distance x/d=10 exceeds the average temperature of gas jet and is 1200 - 1600 K.

As can be seen from the fields of velocities Fig. 4., the velocity of dispersed particles near the covering surface exceeds the average gas jet velocity and depending on the sizes of particle reaches  $150 - 320 \text{ m} \cdot \text{s}^{-1}$ . The ability to use calculation results into practice, the simulation data were compared with the experiments [14, 15].

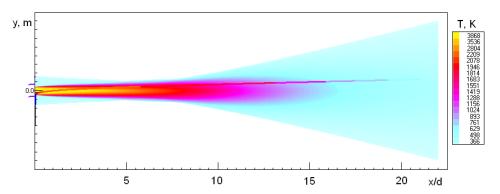


Fig. 3. Fields of temperatures in plasma jet and the trajectory of a single 50µm zeolite particle

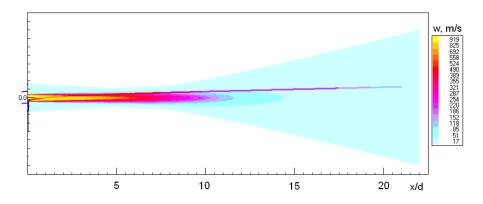


Fig. 4. Fields of velocities in plasma jet and the trajectory of a single 50µm zeolite particle.

The visual observations by a high speed camera suggest that multiphase jet in the exhaust of PCR nozzle consists of melted domains, solid grains of different sizes and fiber filaments. Experimental tests showed that zeolites powder, injected into a high temperature zone, is melted very quickly.

The high speed cameras images showed that there exist a turbulent flow at the exhaust region of plasma jet (Fig. 5, 6) and the multiphase flow, sprayed from plasma torch, consists of melted grains of different shapes and sizes. The fiber formation begins at the spraying distance x/d=2. The melted mass (Fig. 5., position 1) is stretched into many small and tiny filaments (Fig. 5 position 5). Big and heavy granules (Fig. 5 position 2) have a slow motion and are separate from any fiber filament. Granules move with the velocity of 40 - 50 m·s<sup>-1</sup>, because they were stopped by the melted mass. Small, fast granules and fiber filaments (Fig. 5 positions 3, 4) which can be seen on high speed video, move with the velocity of  $220 - 100 \text{ m·s}^{-1}$ .

Observation and velocity measurements of particles, granules and melted domains with a high speed camera in a supersonic two-phase plasma flow of Mach number up to 1.5 shows that the time constant of particles can be evaluated by comparing the experimental results with theoretical values.

The supersonic multiphase plasma jet can be get using the exit nozzle of diameter 10 - 13 mm. The high speed camera images (Fig. 6) showed, that the "shock diamonds" of the supersonic plasma jet are observed at the distance from the exhaust nozzle of the plasma torch to x/d=6.

As is visible from camera image the liquid film on the cooled wall of the reactor arises due to heat energy release on the boundary layer. The friction energy is maintained by the viscous dissipation of energy during the viscous motion. So, forces of the viscosity progress the evolution of viscous boundary layer. Due to the significant kinetic energy melted material is carried away from the liquid volume and the formation of fiber filaments occurs. In other cases, the melted substance is slowly moved ( $0.5 \text{ m} \cdot \text{s}^{-1}$ ) over plasma jet reactor walls toward the exhaust nozzle then separate from the solid surface and leaves the main melt in a rectilinear motion forming a small droplets less than 1 mm in diameter. The released droplet turned toward the jet edges and forms granules of different diameter. Changes of the liquid phase thickness, viscosity and surface tension always determine the fibrillation process and yield of mineral fiber.

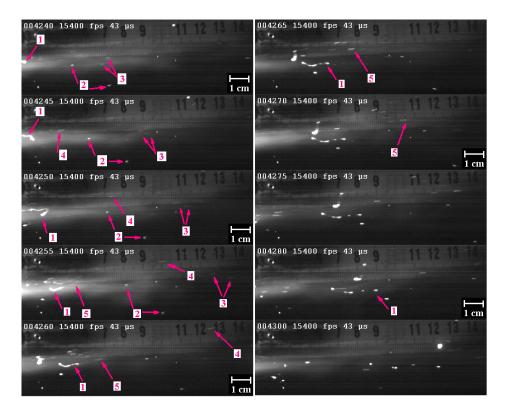


Fig. 5. Images of spraying process in a subsonic plasma jet.



Fig. 6. Spraying process in a supersonic plasma jet.

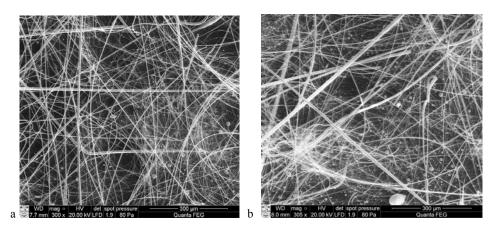


Fig. 7. Morphology of zeolite fibers, produced by different spraying regimes. The outlet section diameter is 0.01 m (a) and 0.015 m (b).

### V. GRIGAITIENĖ, R. KĖŽELIS, V. VALINČIUS, M. MILIEŠKA

With the aim of intensification of heat exchange between the high temperature flow and the injected particles in order to obtain the fiber products a small amount of propane-butane gas was added (Fig.2, pos.2). Gas combustion products ( $H_2O$  and  $CO_2$ ) increase radiation heat transfer between the high temperature flow and particles considerably. As a result melting process was intensified. Additional gas injection (Fig.2, pos.3) allows changes melt viscosity for optimal fiber production.

The main characteristics of fiber material including diameter and length of fibers, the amount and the shape of non-fiber inclusions were studied using scanning microcopy (SEM). The morphology of fiber, obtained by plasma melting of zeolite raw material is shown in Fig. 7. When the diameter of the outlet section was 0.01 m, the fiber was produced in a supersonic flow. The filaments are thinner and less additional granules are obtained, but the outflow was stuffed up by a melted mass. In another case (Fig. 7., b), the diameter of the outflow was increased to 0.015 m, the fiber was produced in the subsonic plasma flow regime. The average fiber thickness was from 0.5 to 5  $\mu$ m and the average fiber length is about 0.07 m.

**CONCLUSIONS.** The flow visualization using a high speed camera was applied to study the influence of nozzle and parameters of the arc on behavior of the multiphase flow as well as on expansion of heterogeneous plasma flow after the nozzle. The structure of the melted domain flow and plasma both before and after confinement nozzle was revealed. The subsonic and supersonic flows were generated and observed, and the main operating regimes of zeolites fiber production were established. In the central spray zone the motion of melted domains was found to be influenced on the following main factors: i) plasma source characteristics and operating regime; ii) plasma flow formation, characteristics and its interaction with walls of the reactor; iii) plasma forming gas and powder injection approach and place; iiii) powder composition, size and fraction, its injection rate parameters.

The obtained results could be applied for optimizing operating modes of designed by the authors an electric arc reactor for industrial plasma processing of dispersed materials with high efficiency and low specific power. It was found that flow velocity and temperature relief determines the increasing of particles, granules and melted domains velocity and temperature in the plasma jet in the distance up to x/d=12. The maximal velocity of particles slightly exceeds the mean plasma jet velocity.

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#### References

- 1. Kėželis R., Balkevičius V., Kaminskas A. Regularities of glass fiber manufacturing by means of plasma technology // Power engineering, 2000, N 4, pp.110-116.
- 2. Valinčius V., et al., Preparation of insulating refractory materials by plasma spray technology. J. of High Temp. Mat. Proc., 2006, 10, pp. 365 375.
- Karthikeyan J. et al., Plasma spray synthesis of nanomaterial powders and deposits, Mat. Sci. Eng., 1997, A238 pp. 275-286.
- 4. Dresvin S. V., et al. Physics and Technology of Low-Temperature Plasmas. Iowa State Univ. Press, 1977, p. 352.
- Kogelschatz U., Atmospheric-pressure plasma technology. Plasma Phys. Control. Fusion . 2004, p. 46 B63– B75.
- 6. Gupta C. K., Sathiyamoorthy D. fluid bed technology in materials processing. Technology & Engineering, 1997, p. 498.
- Valinčius V. et al. Interaction of dispersed ceramic particles and melted domains with turbulent plasma jet. Proc. of the sixth int. symp. on turbulence, heat and mass transfer 6, Rome, Italy, September 14-18, 2009. New York, Wallingford(UK): Begell House, 2009, Inc., ISBN 978-1-56700-262-1, pp. 291-294.
- 8. Sirignano W. A. Fluid dynamics and transport of droplets and sprays. Cambridge University Press, 1999, p. 309.
- 9. Raffel M., et al. Particle image velocimetry. Springer, 2007, p. 448.
- 10. Valatkevičius P., Valinčius V., Kėželis R. The effect of gas inlet location and intensity on plasma torch characteristics. Proc. of 15th Int. Symp. on Plasma Chemistry, Orleans, 9-13 July, 2001, pp. 1585-1590.
- 11. Valinčius V. et al. Electric and thermal characteristics of the linear, sectional DC plasma generator. Plasma sources science and technology. ISSN 0963 0252, 2004, vol. 13, , pp. 199 206.
- 12. Delluc G., et al. A numerical tool for plasma spraying. Part I: modelling of plasma jet and particle behaviour Abstracts and full paper CD of the 16th ISPC Taormina, Italy, June 22-27, 2007, p. 6.
- Delluc G., et al. A numerical tool for plasma spraying. Part II: Model of statistic distribution of alumina multi particle powder CD the 16th ISPC Taormina, Italy, June 22-27, 2007, p. 6.

- 14. Valatkevičius P., Krušinskaitė V., Valinčiūtė V., Valinčius V. Preparation of catalytic coatings for heterogeneous catalysts employing atmospheric pressure non-equilibrium plasma, Surface & coatings technology. 2003, ISSN 0257-8972 / Vol. 173-174, p. 1106-1110.
- 15. K. Brinkiene, R. Kezelis. Effect of alumina addition on the microstructure of plasma sprayed characteristics yttria stabilized zirconia. J. Euro. Cer. 2005, S, 25 2181-2184.