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NUMERICAL AND EXPERIMENTAL IMAGES OF MULTIPHASE PLASMA JET DURING PLASMA PROCESSING OF DISPERSED MATERIALS

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Abstract. *This study presents results of analytical and experimental investigation on behaviour of dispersed particles, granules, also melted and concentrated ceramic material domains in the entrance region of plasma jet outflowing from plasma chemical reactor exhaust nozzle. High thermal resistant granules, fibre and fine particles have been produced from zeolites employing a specific plasma spray technique generating non-equilibrium plasma jet at atmospheric pressure. 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 70 – 80 kW, temperature of gas leaving the reactor – 2500 – 3500 K, flow velocity in the outlet – 900 – 1500 m/s.*

Performed experimental and analytical studies showed that process of plasma melting and conversion of melt into fibre depend on following main factors: i) plasma generator 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; iiiii) initial domain formation, splat layering and process of spray pyrolysis.

1 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 fibre 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 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 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, fibre, 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 aluminium oxide and zeolites.

2 EQUIPMENT AND PROCEDURE

“Fluent” [10] and “Jets&Poudres” [11,12] software was improved and applied to model for specific plasma jet. Fluent is used to solve the governing equations are as mass, momentum and energy conservation equations. Turbulence is accounted for by using the k-ε model and the perfect gas law is solved to close the system of equations.

For the boundary conditions, it is assumed that the plasma jet exits in a chamber where the pressure is atmospheric.

The software has been used to solve Navier-Stokes and energy equation based on the dynamic k-ε model for the fluid jet in the space between two walls of sudden expanded tube. The system was assumed to be axisymetrical and the fluid flow turbulent.

According to the conception of thermal boundary layer, there was postulated that all changes in a thermal process are intramural and close to the surface ($\partial t/\partial y \neq 0$ inside and $\partial t/\partial y = 0$ outside boundary layer). We have also concluded that $\partial^2 t/\partial x^2 = 0$, since the thermal boundary layer is very thin near the cavity bottom wall. So the energy equation in the Cartesian coordinate system assumed the conformation as

$$\rho u \frac{\partial T}{\partial x} + \rho v \frac{\partial T}{\partial y} = u \frac{dp}{dx} + \frac{1}{\mathrm{Re}\,\mathrm{Pr}} \frac{\partial}{\partial y}(\mu \frac{\partial T}{\partial y}) + \frac{\mu}{\mathrm{Re}}(\frac{\partial u}{\partial y})^2, \tag{1}$$

the momentum equation as

$$\rho u \frac{\partial u}{\partial x} + \rho v \frac{\partial u}{\partial y} = -\frac{dp}{dx} + \frac{1}{\mathrm{Re}} \frac{\partial}{\partial y}(\mu \frac{\partial u}{\partial y}). \tag{2}$$

As closing equations the equation of continuity

$$\frac{\partial(\rho u)}{\partial x} + \rho v \frac{\partial \rho v}{\partial y} = 0, \tag{3}$$

and equation of state as

$$p = \frac{\gamma - 1}{\gamma} \rho T \tag{4}$$

were used.

The other typical 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 low temperature plasma spraying equipment used in this research was developed at Lithuanian Energy Institute (Fig. 1). 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 linear single-chamber DC plasma torch with button type cathode and step-formed anode. The similar plasma spray torch 100 kW of power is analyzed in details elsewhere in [13,14]. Such plasma torch has very stable operating parameters (outlet plasma jet temperature, velocity and their fluctuation). Mean-mass outlet jet temperature and velocity was determined from 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^{-1} (main gas flow rate throw plasma torch – 17 g·s^{-1}, additional – 0 – 5 g·s^{-1}, propane butane gas – 0,12 g·s^{-1}).

Mean-mass outlet plasma temperature is 3000 – 3700 K, outlet average velocity – 900 – 1500 ms^{-1}, the plasma forming gas is air.

A specific plasma chemical reactor (Fig. 2) for melting of zeolite and fibre production was constructed. It is directly connected to the plasma torch anode and consists of one straight sections 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 diameter varies from 0.01 to 0.015m. Such design enables to get high outlet flow velocity (supersonic jet).

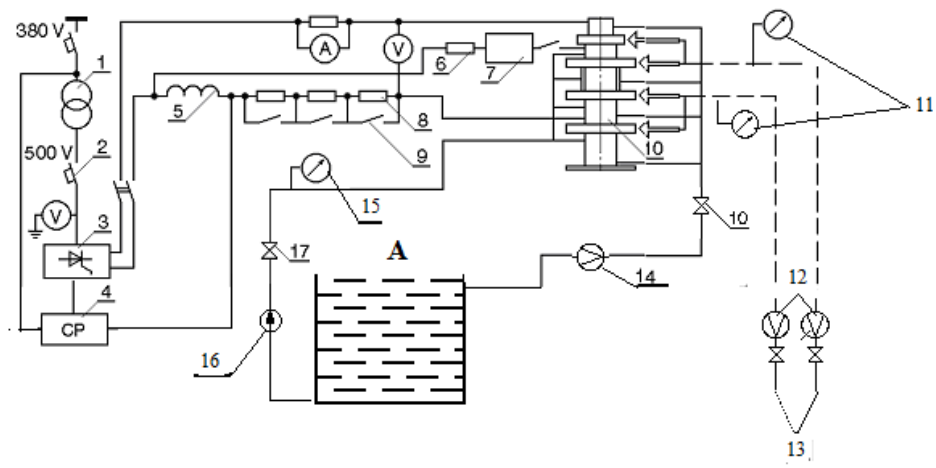

Fig.1. Schematic presentation of experimental set-up. Explanation is given in the text.

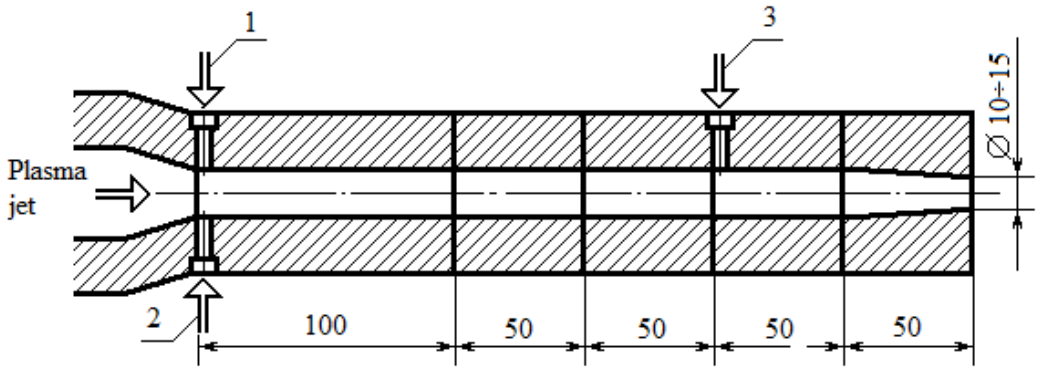

Fig.2. Scheme of experimental PCR channel. 1 – injection of powder precursor, 2 – injection of propane-butane, 3 – injection of additional gas

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 $kg{\cdot}m^{-3}$. 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.

Series No.	113	120	123
Diameter, mm	10	13	15
Flow velocity, $m \cdot s^{-1}$	1515	1092	898
Power, kW	67	69	74
Plasma forming gas flow rate, $g \cdot s^{-1}$	14,8	14	14
Carrier gas flow rate, $g \cdot s^{-1}$	1	1	1
Additional air flow rate, $g \cdot s^{-1}$	4,5	3,5	3,8
Flow rate of propane-butane, $g \cdot s^{-1}$	0,5	0,5	0,5
Mean-mass gas temperature in the reactor exhaust cross section, K	1892	2522	2734

Table 1. Process parameters

Plasma forming gas was supplied via the specific ring into the reacting arc zone, the propane-butane gas was injected via second ring at the end of anode or in the entrance region of PCR. Therefore the length of plasma torch has increased. Precursor powder was injected into air plasma flow inside the reactor (Fig.2, pos.1).

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 µs.

The diameter of particles was evaluated from the cross-sectional scanning electron microscopy (SEM) observation and the surface zone phase composition was analyzed by X-ray diffractometer.

3 RESULTS AND DISCUSSION

Numerical research of two-phase high temperature jet was carried out using “Fluent” and “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, hard spherical dispersed particles are 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 mixing with plasma jet, solid particles have lower temperature than that of jet, and certain time is needed to heat them up. Particles have to be rather fine in order to quickly heat 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 velocities Fig. 3, the temperature of dispersed particles approximately at the distance x/d=10 exceeds average temperature of gas jet and is 1200 – 1600 K.

As can be seen from fields of velocities Fig. 4, velocity of dispersed particles near the covering surface exceeds average gas jet velocity and depending on the sizes of

particle reaches 150 – 320 m·s^{-1}. The ability to use calculation results into practice, the simulation data were compared with experiments [15, 16].

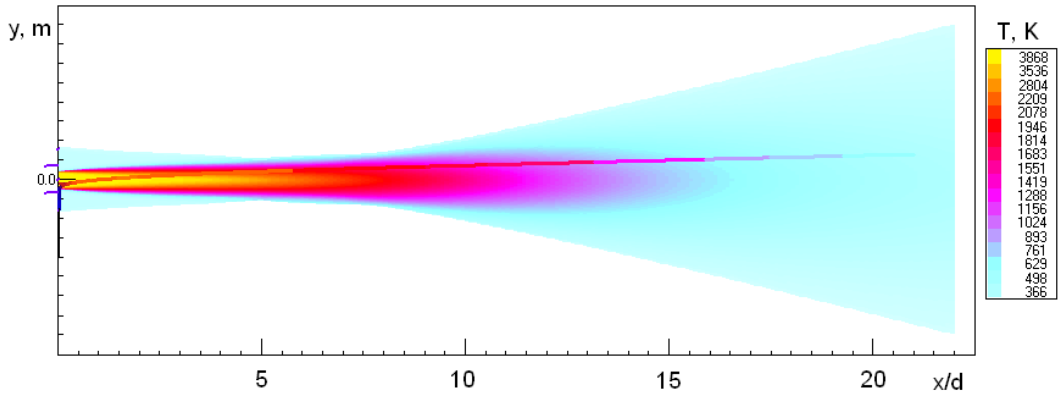

Fig. 3. Fields of temperatures in plasma jet and the trajectory of single 50µm particle

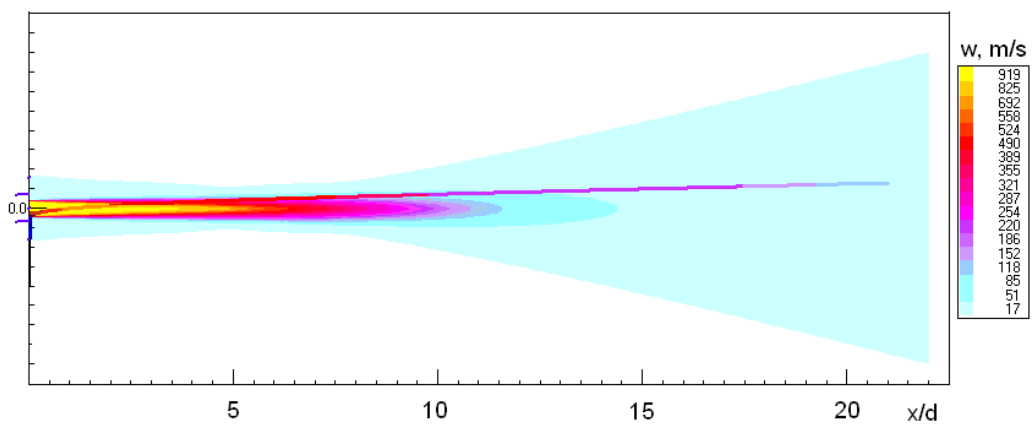

Fig. 4. Fields of velocities in plasma jet and the trajectory of single 50µm particle.

Powder particles and melted domains have been used as micro-probes for the visualization of the motion in plasma as well as for the observation of interaction between plasma and melted or partly melted domains.

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

High speed cameras images showed that there exists a turbulent flow at the exhaust region of plasma jet (Fig. 6, 7). The subsonic and supersonic flow has been observed at the same operating regime of plasma generator generator without or with supply of raw material respectively. It means that in the case of powder supply the diameter of the reactor channel decreases and supersonic flow occurs.

The high speed cameras images (Fig. 6) showed, that 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. pos. 1) is stretched into many small and tiny filaments (Fig. 5 pos. 5). Big and heavy granules (Fig. 5 pos. 2) have slow motion and are separate from any fiber filament. Granules move with the velocity of 40, 50 m·s^{-1}, because they were stopped by melted mass.

Small, fast granules and fiber filaments (Fig. 5 pos. 3, 4) which can be seen on high speed video, move with the velocity of 220 – 100 m·s^{-1}.

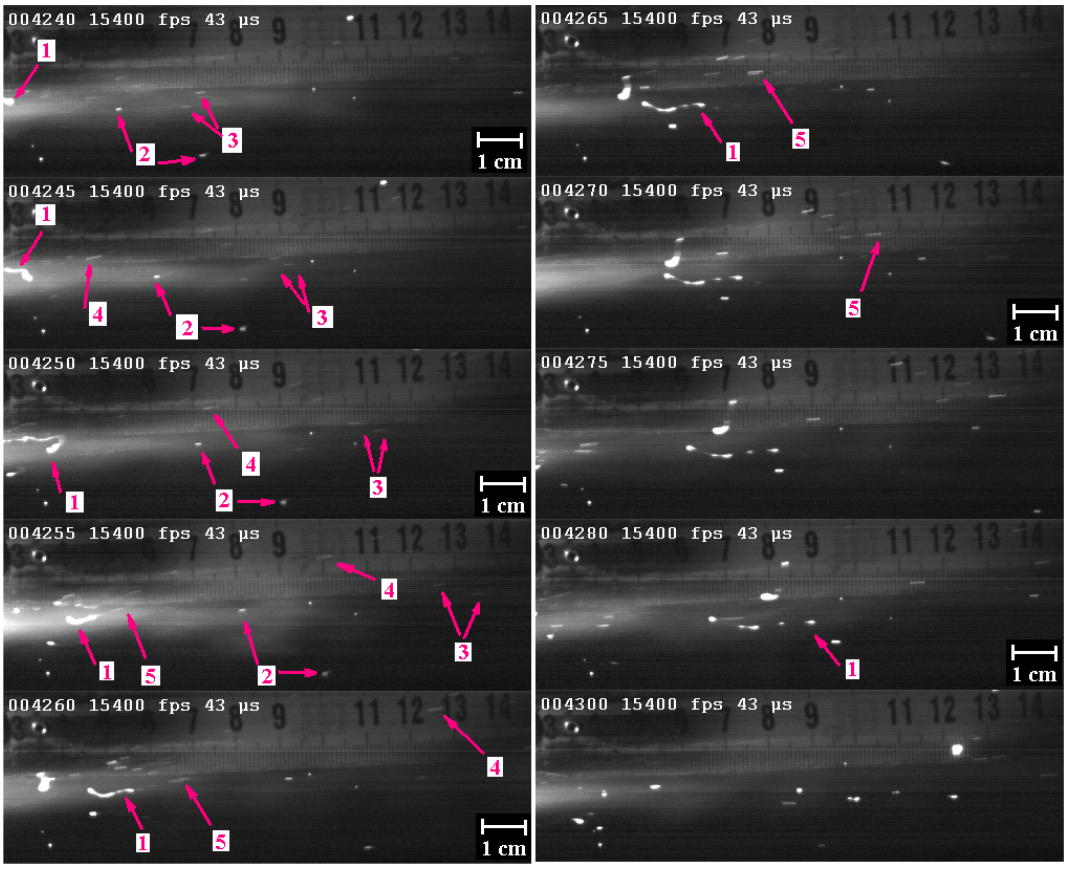

Fig. 5. Images of spraying process in subsonic plasma jet.

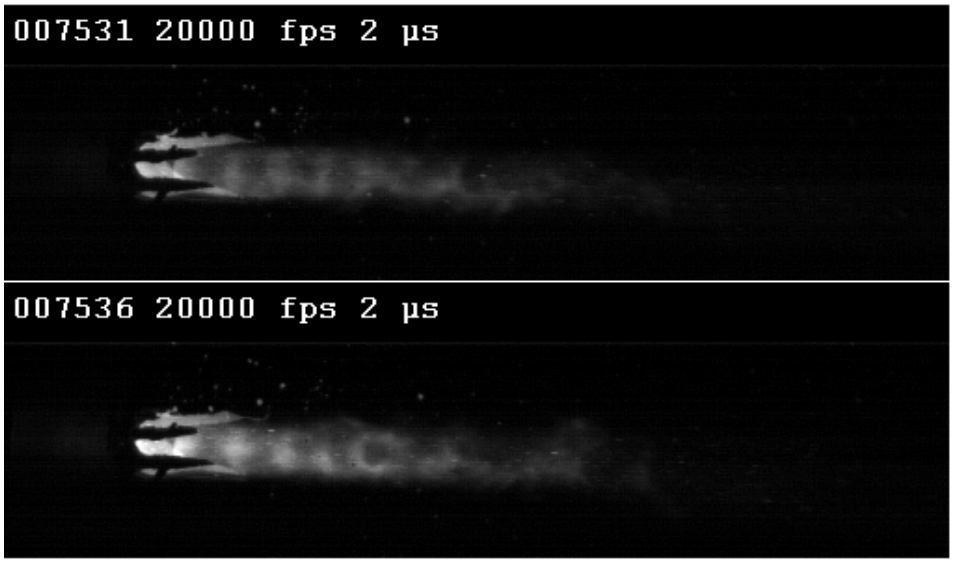

Fig. 6. Images of spraying process in supersonic plasma jet.

Observation and velocity measurements of particles, granules and melted domains with high speed camera in a supersonic two-phased plasma flow of Mach number up to

1.5 shows that 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 supersonic plasma jet are observed at the distance from the exhaust nozzle of the plasma torch to *x/d*=6.

Also the supersonic flow was observed after the channel was coated by melted substance. The possible explanation of the observed fact is the formation of a certain Laval nozzle during thermal melting of complex substances (Fig. 7).

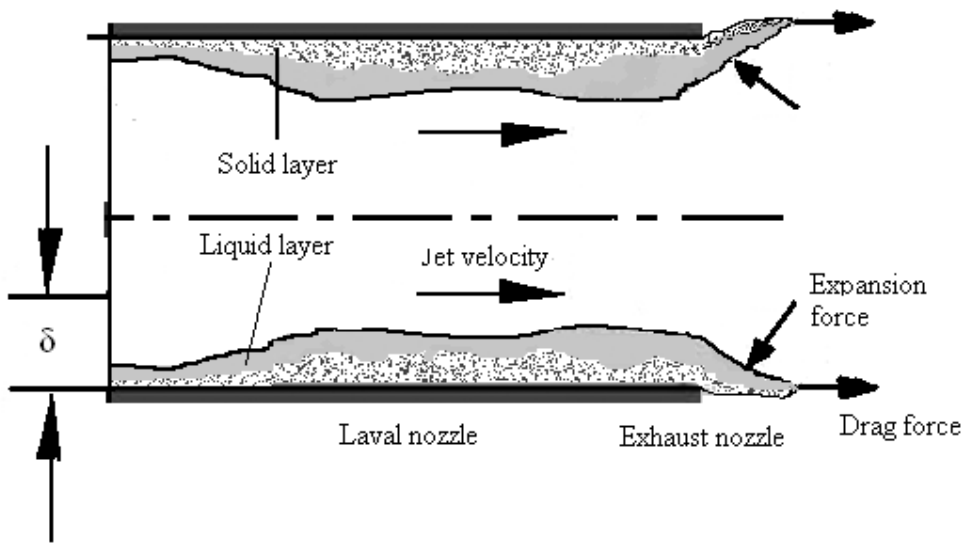

Fig. 7. The model of motion of liquid substance in then reactor and the mechanism of fiber formation. Arrows at the end of nozzle shows the direction of prevailing forces and the motion direction of melted material

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 fibre filaments occur. In other cases melted substance is slowly moved (0.5 $m \cdot s^{-1}$) 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. The mechanism of this motion (Fig. 7) is still discussed. Changes of the liquid phase thickness, viscosity and surface tension always determine the fibrillation process and yield of mineral fibre.

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 fibre 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. The morphology of fiber, obtained by plasma melting of zeolite raw material is shown in Fig. 8.

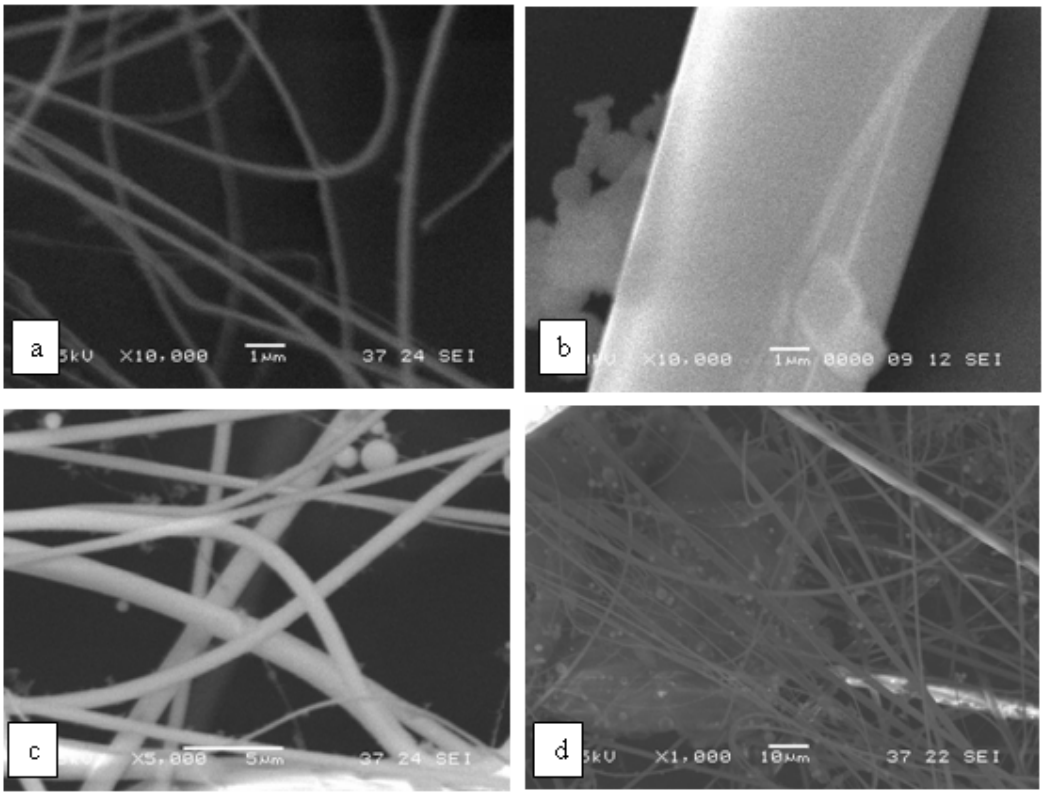

Fig. 8. Morphology of zeolite fibers, produced by different spraying regimes. The outlet section diameter is 0.01 m (a, b), 0,013 m (c) and 0.015 m (d).

The average fiber thickness was from 0.5 to 5 μm. On evidence of structure analysis data the formation of non-fiber inclusions having shapes of beads with various diameters is not inherent to the fiber material obtained during plasma melting of zeolite (Fig. 8). Overage fibre length is about 70 mm.

4 CONCLUSIONS

High speed camera‘s method was applied to study the influence of nozzle and parameters of the arc on behaviour of multiphase flow as well as on expansion of heterogeneous plasma flow after the nozzle. The structure of melted domain flow and heterogeneous plasma both before and after confinement nozzle was revealed. For central arc zone the motion of melted domains was found to be influenced on 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; iiiii) initial domain formation, splat layering and process of spray pyrolysis respectively.

The supersonic flow was observed after the channel was coated by melted substance because of the decrease of reactor diameter or formation of a Laval nozzle during thermal melting of complex substances.

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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