Journal of International Scientific Publications:
Materials, Methods & Technologies, Volume 6, Part 2

FORMATION OF AN ION-CLUSTER BEAM FOR MATERIAL PROCESSING

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Abstract

In this paper the possibility of accelerating gaseous cluster-ion beams for nanomaterials processing are discussed. In order to realize surface processing technology, a high-energy gas cluster ion beam irradiation system was developed. The results of the experimental study of formation of an intense cluster-ion beam of argon and nitrogen are presented. The fundamental phenomenon influences on the main parameters of cluster-ion beam (cluster-ion size, energy, at al.) are considered.

Key words: *supersonic jet, gas cluster, molecular beam, ionization, cluster ion, surface processing*

1. INTRODUCTION

Today beams of atomic or molecular ions are in fact become an integral part of many modern technologies (Giannuzzi and Stevens, 2004, Orloff, 2008). Conventional now ion cleaning, etching, implantation, sputtering and deposition of thin films of various materials - all of which are based essentially on the individual collisions (or a cascade of binary collisions) of incident ions with near-surface atoms. A completely different situation occurs during a collision the energetic large clusters with the surface. In this case, nearly simultaneous interaction of many particles of cluster around the same number of atoms in a solid occurs. This leads to deposition a high-energy density into a very small volume of the target material, and strong nonlinear effects: lateral sputtering, dry etching, and shallow implantation. Recently it was shown that the accelerated gas cluster ion beams have a number of unique advantages that allow them to be considered a promising basis for new technologies, including nanotechnologies (Popok and Campbell, 2006, Yamada, 2007).

Cluster we call the aggregates of a few units up to tens of thousands of atoms or molecules, combined into a single complex. The properties of the clusters are in the middle between the individual molecules and bulk matter. Under laboratory conditions, the clusters are formed during a supersonic expansion of gases through a nozzle into vacuum. It's known that in supersonic jet due to free expansion the gas temperature drops down to cryogenic values and clusters can form (Pauly, 2000). Forces, holding molecules in the cluster, could effect on the threshold energy of activation and ionization of molecules in a clusters, and result to broadening of energy levels (Korobeishchikov et all, 2005, Zarvin et all, 2008).

One of the distinguishing features of cluster ions is the possibility to widely regulate an energy corresponding per one unit of the cluster. By varying the size of the cluster and its energy it is possible, for example, to generate an intensive flow of material with a specific energy of several eV/molecule. As is well known it is very difficult to do using the traditional monomer ion beam due to the effects of repulsion. Thus, we can successfully implement the various modes of irradiation: from the deposition of the soft mode before scattering of the irradiated target.

It was be found (Yamada, et all., 2003), that for the purpose of modifying the surface requires intensive flows of clusters of up to thousands units with energy up to 30-40 keV and radiation doses

up to 10^{15}-10^{17} ions/cm^2. Apparently, the only source that can provide high-intensity clusters flow is molecular beam generated from the supersonic gas jet. Having picked geometry of a nozzle and by varying the parameters of gas (stagnation pressure and temperature) clusters of different sizes can be obtained.

However, kinetic energy, obtained the cluster during the free supersonic expansion is determined by the initial enthalpy of stagnation gas and generally does not exceed 0.1 eV per particle (atom or molecule) that it is not enough for surface treatment. To speed up the cluster it is necessary to ionize a cluster, and then accelerate this up to the desired energy in the electric field. It is known that the ionization energy threshold (~ 10-20 eV) significantly greater than the binding energy of the particles in the gas cluster (from 0.1 to 5 eV). Obviously, the ionization may lead to collapse (fragmentation) of the cluster, what may lessen the intensity of cluster-ion beam. Thus, the formation of high-intensity accelerated cluster ion beam is a complex physical and technical task that depends on many parameters which cannot be solved analytically.

In this paper we present the first results on the formation of intensive cluster ions beams from the supersonic jets of Ar.

2. EXPERIMENTAL

The work was performed on an experimental setup LEMPUS of Novosibirsk State University. A detailed description of the stand is given in (Zarvin et all, 2000). To perform this work has been upgraded the setup. General view of the experimental stand is shown in Fig. 1. Now the stand is equipped with high-vacuum system is completely oil-free: turbomolecular pumps Shimadzu TMP-2203LMC, TMP-1003LM and oil-free mechanical pumps Anest Ivata ISP-500, ISP-250 (Japan). The source of the jet is located inside the expansion chamber at the three-component coordinate mechanism. Capacity pumping system is on the expansion chamber - 2000 l/sec, on the ionizer chamber - 1000 l/sec.

Schematic diagram of the formation of a cluster ion beam is shown in Fig. 2. The experiments were carried out with a supersonic conical nozzle having a throat diameter of $d_* = 0.19$ mm and angle $\alpha = 12^\circ$. The molecular beam was formed by a skimmer with a diameter of the inlet ds = 1 mm. Stagnation pressure P_0 was varied from 1 to 10 bar, stagnation temperature T_0 was fixed 295°C. Ionization of the clusters was carried out by electron impact. The electron source was a tablet of lanthanum hexaboride, heated up to a temperature of 1800°C. The electron current was 50-200 mA.

To determine the optimal parameters for the formation of an intense cluster beam measurements of the total intensity of the neutral molecular flow were made by varying the stagnation pressure P_0 and the distance of the nozzle - skimmer x_{ns}. The measurements were performed using a closed ionization pressure sensor located on the axis of the molecular beam. Since the clusters are completely destroyed by collisions with the walls inside the sensor, the equilibrium density of the gas in the sensor is proportional to the intensity cluster molecular beam (Alexandrov and Kusner, 1989, Handbook of Nanophysics 2, 2011).

To register the distribution of cluster ions to the energy classical method of retarding potential was used. After differentiation of the recorded curve, we obtained the distribution of cluster ions by size.



Fig. 1. General view of the experimental setup LEMPUS-1.

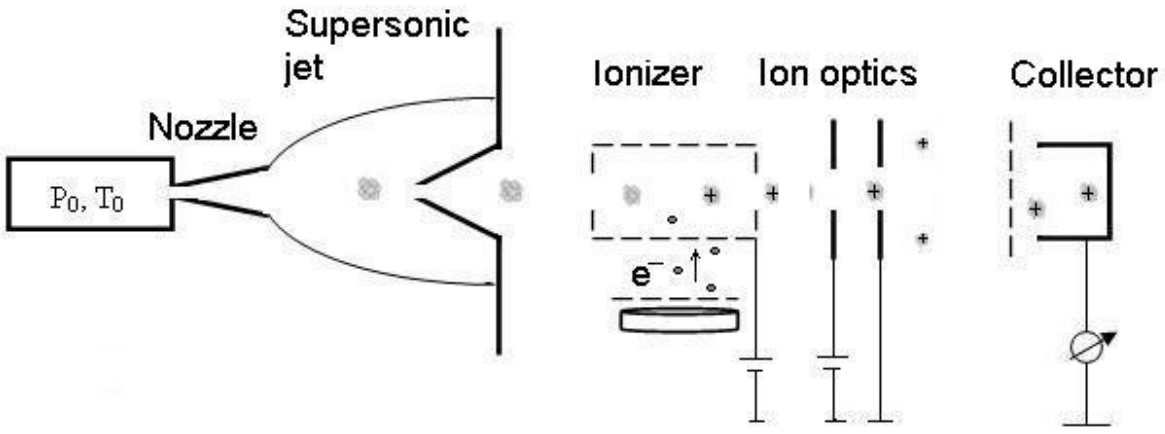

Fig. 2. Scheme of the formation of the cluster ion beam.

3. RESULTS AND DISCUSSION

In Fig. 3 shows the values of pressure on axis of molecular flow P_a and background pressure in the ionizer section P_b, measured simultaneously at a fixed pressure $P_0 = 5$ bar at different nozzle-skimmer distances. Here and further the distance nozzle - skimmer is measured in caliber (diameter of the nozzle throat), x/d_*. There is an arrow shows the position of the boundary of the jet - direct shock wave (Mach disk), calculated from the known empirical formula:

$$x_M = 0.67 * \sqrt{P_0 / P_k}\ , \qquad (1)$$

where P_k is background pressure in the expansion chamber. Obviously, the background pressure in the ionizer chamber may influence the correctness of the sensor readings of the intensity of the molecular beam. Therefore, the true pressure in the molecular beam P_{mb} defined as:

$$P_{mb} = P_a - P_b \qquad (2)$$

When moving from the Mach disk to the nozzle increases leakage of gas through the skimmer into ionizer chamber according to the isentropic density distribution of gas in the jet ($\sim 1/x^2$, on the figure the dotted line). As a result, the background pressure P_b increases proportionally. The pressure of the molecular beam P_{mb} also dramatically increased in proportion to $\sim 1/x^2$ after leaving the Mach disk ($x/d_* < 330$). However, the increase in background pressure leads to molecular beam scattering, which leads initially (at $x/d_* \sim 240$) to a deviation from linear growth, and then (at $x/d_* < 200$) and a pressure drop P_{mb}. Thus, for these conditions the molecular beam is formed with a maximum intensity at distances nozzle - skimmer 150-200 calibers.

Using this algorithm of measurements, we calculated the intensity of the clustering molecular beam for several fixed stagnation pressure (Fig. 4). Despite the fact that the size of the jet (the distance to the Mach disk) changes markedly with increasing P_0, the optimum distance of the nozzle - skimmer of about the same - 150-200 calibers. The maximum intensity ($\sim 2*10^{18}$ molecules/sm^2*sec) is attained at $P_0 = 6$ bar.

It should be noted a significant non-linearity depending on the maximum intensity of the clustering molecular beam from the pressure P_0: at $P_0 = 1$ bar, the maximum intensity of more than 15 times smaller than at $P_0 = 3$ bar. One of the reasons is the influence of background gas in the expansion chamber. At low pressures transient regime of expansion formed with a blurring of shock waves limiting outside the core of the jet and, consequently, with a noticeable penetration of background gas, resulting in a decrease in the intensity of the molecular flow. In this case, at $P_0 = 1$ bar, there is no central Mach disk (calculated position - $x/d_* = 280$).

Another important reason is the various stages of condensation of gas flow in a supersonic flow. As you know, in a free jet heavy particles (in this case – clusters) are concentrated at the jet axis as a result of gas-dynamic separation [6]. Accordingly, the intensity of the beam is determined clustering molecular condensate fraction (fraction of gas in a bound state) and the size of the clusters. For our

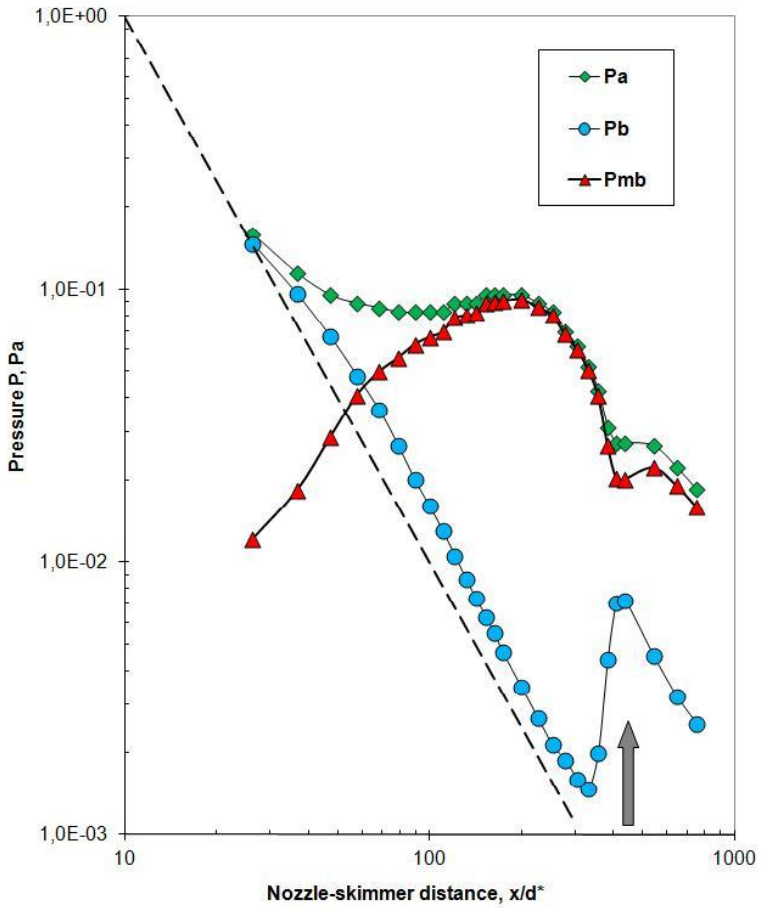

Fig. 3. The pressure on axis of molecular flow, P_a, background pressure in the ionizer section P_b, and pressure in the molecular beam P_{mb} depend of nozzle-skimmer distances at P_0 = 5 bar.

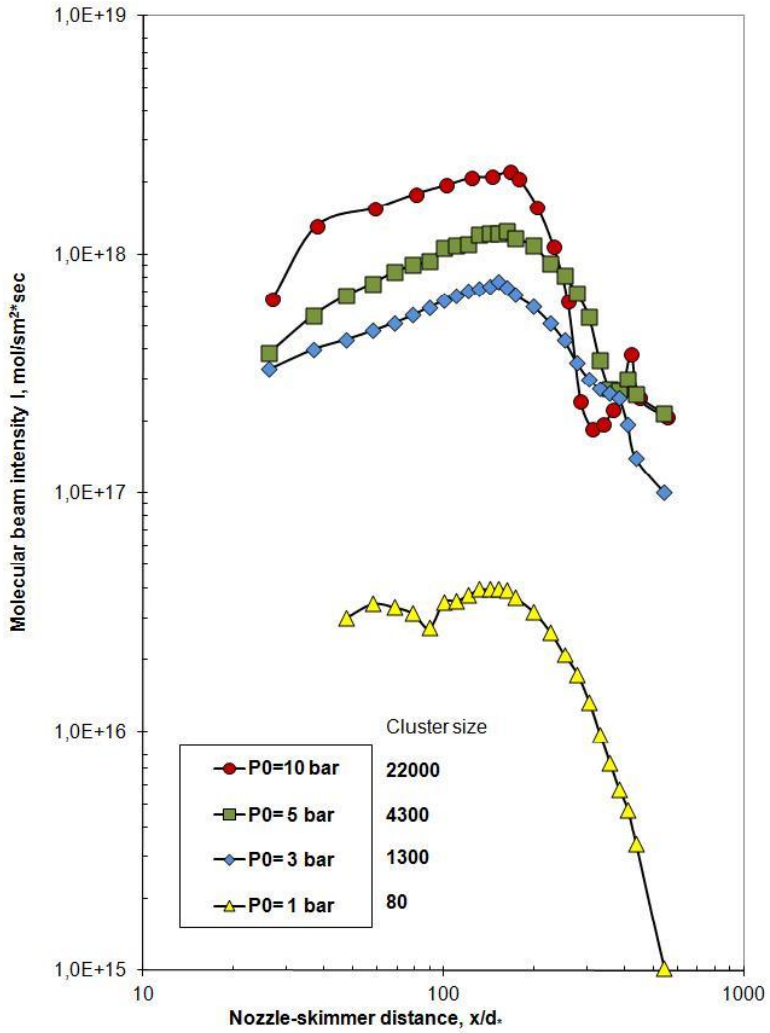

Fig. 4. The intensity of cluster molecular beam (molecules/sm^2*sec) depends of pressures P_0 and nozzle-skimmer distances.

conditions, we calculated the dimensionless similarity parameters for flows with condensation, Hagena`s parameter:

$$\Gamma^* = k * (d_{eq})^{0.85} * P_0 / T_0 , \quad (3)$$

where d_{eq} is the equivalent diameter of the nozzle (Hagena, 1992).

Calculations show that at P_0 = 5 bar flow is in the phase of massive condensation, which is characterized by a sharp increase in the proportion of the total condensate, and relatively weak growth

of the average cluster size. As a result, the intensity of the clustering molecular beams growing much faster than linear growth stagnation pressure. When P_0> 2 bar flow moving into the development phase of condensation, in which the condensate fraction reaches a constant value (25-30%), and the average cluster size increases significantly. As a result, at this stage, the intensity of the molecular flow increases linearly with pressure P_0. The figure also shows the average cluster size N for the corresponding pressures calculated from empirical formulas (Hagena, 1992):

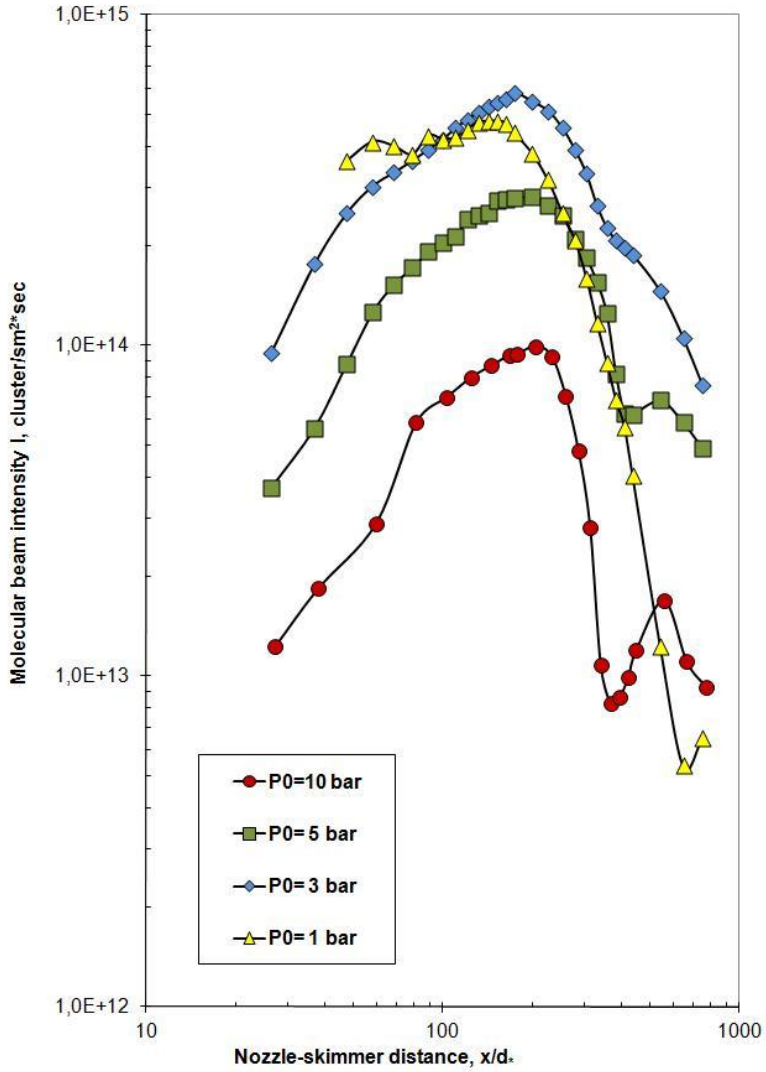

Fig. 5. The intensity of cluster molecular beam depends of pressures P_0 and nozzle-skimmer distances.

$$N = 38.4 * (\Gamma^* / 1000)^{1,64} \text{ for } 300 < \Gamma^* < 1800,$$

$$N = 33 * (\Gamma^* / 1000)^{2,35} \text{ for } \Gamma^* > 1800 \quad (4)$$

At maximum pressure the average cluster size reaches a value of 22 000 molecules/cluster, whereas at P_0 = 1 bar, the average size of 80 molecules/cluster.

Using the mean cluster sizes intensities of the molecular beam were calculated in terms cluster/sm^2*sec, the results are shown in Fig. 5. It is seen that, except for the very low pressure, with increasing P_0 the flow of clusters on the axis of the jet decreases. This is explained by the previously mentioned factors - the proportion of output saturation and condensation on the continued growth of the average cluster size. Thus, the condensation at this stage continues, mainly due to consolidation of small clusters. So under such condition it is possible obtained a cluster beam very high intensity: up to 4-10^{14} clusters/sm^2*sec.

In Fig. 6 shows the distribution of cluster ions Ar in size, recorded by the retarding potential method at several P_0. As expected, the average size of cluster ions increases with increasing stagnation pressure: at P_0=2 bar average size of cluster ions is near 500 units, at P_0=5 bar - near 800 units, at P_0=6 bar - near 1000 units. Comparison with the calculations of the initial cluster size shows that the ionization of the neutral cluster is a marked decrease in its size.

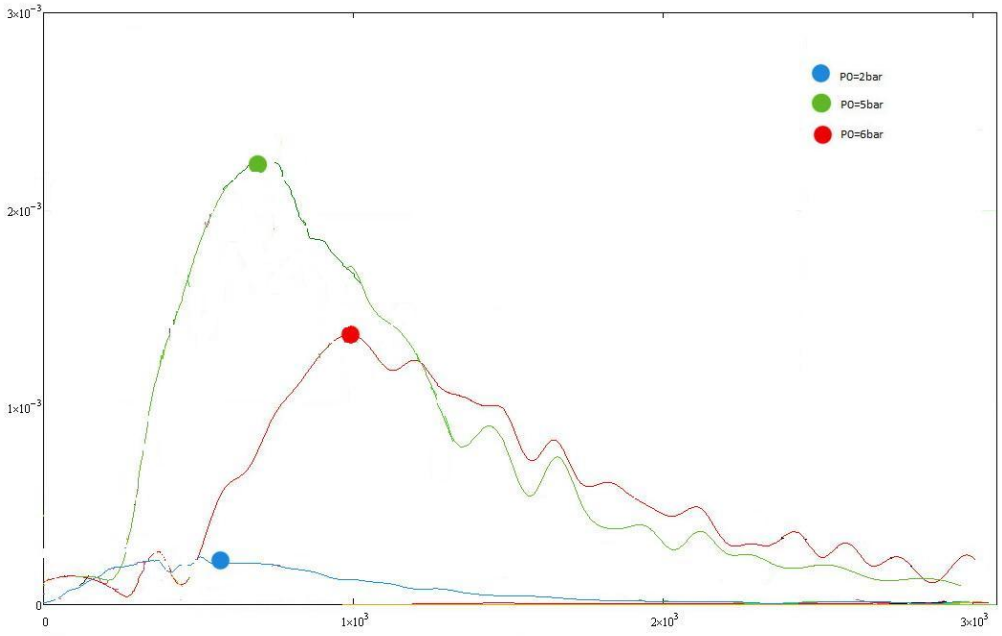

Fig. 6. Ar cluster size distribution at P_0=2 bar, 5 bar, 6 bar measured with the retarding potential method. I_e=105 mA, U_e=100V, U_{ext}=50V

4. CONCLUSIONS

As a result of experimental investigations the optimal conditions for the formation of a clustering molecular beam from supersonic jets of argon have been determined. A maximum intensity of 4*10^{14} clusters/cm^2*sec for the clusters with an average size of 1000 molecules, and 8*10^{13} for clusters with an average size of more than 20,000 molecules have been reached. The distributions of cluster ions in size have been recorded. It was found that the average size of the ionization of cluster ions is several

times smaller than the source of the neutral cluster. At present, works are underway to optimize the regimes of ionization in order to increase the current of clustering ion beam.

ACKNOWLEDGEMENTS:

The present work has been supported by the Program "Novosibirsk State University – National Research University" and by the Ministry of Education and Science of the Russia, project No.3.749.2011.

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