



Gas jet deposition of diamond structures by thermal activation on an expanded surface



Alexey Rebrov

Kutateladze Institute of Thermal Physics of the Siberian Division of the Russian Academy of Sciences, Novosibirsk 630090, Russian Federation

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ABSTRACT

We present new approaches to gas phase deposition of diamond and diamond-like structures, films, and crystals. They are based on the activation of initial gases on hot expanded surfaces due to multiple collisions of molecules. The possibility of realizing the diamond synthesis at a very wide range of regime parameters is a distinctive feature of the method. The success of the research is due to the computer modeling of gas dynamic processes which allows us to predict and analyze the results of experiments. We dealt with a mixture of hydrogen and methane. Some results were obtained by using a separate or joint delivery of components to activating surfaces. The vitality of this new method is proved. The development and optimization of the method led to the creation of an energy saving system.

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1. Introduction

By now the gas phase methods of deposition of diamond and diamond-like materials, known under the habitual term “chemical vapor deposition” have left the stage of research and found many applications in various fields of instrument-making and industry. The thorough reviews of the past and recent years [1–5] describe the methods of deposition which are based on various ways of activation of initial gas mixtures. The thermal activation by hot wires, activation in plasma flows or high frequency discharge, by laser radiation or in flames, or by detonation—all these methods led to the commercial production of synthetic diamonds. This is a reason for the decrease of interest in researches of new methods of gas phase diamond synthesis. Meanwhile, there is a lack of unambiguous understanding of what determines the processes of deposition, particularly, what fragments are responsible mostly for design of diamond structures. Methyl molecule CH_3 and atomic hydrogen are not alone active fragments by formation of diamond structures. In previous researches of diamond synthesis by activation at high plasma temperatures there are data on participation C, CH, CH_2 , C_2H_2 , C_2H in deposition. In other words, the search and elaboration of new methods to our mind are not fully exhausted. These circumstances justify the appearance of the new method which is described below.

The main objectives of this article are as follows: 1) to present a new direction of the diamond deposition technology by the thermal activation of the hydrocarbon containing gases on expanded surfaces; 2) to review the experience we have acquired in the Kutateladze Institute of Thermal Physics in elaboration and development of the new method.

E-mail address: rebrov@itp.nsc.ru.

Studies to be presented relate to thermal activation. In a sense, their progenitor is a hot wire or hot filament CVD. In such processes the high concentration of atomic hydrogen in the flow which interacts with a deposition surface is one of the main conditions of the design of a diamond structure. To realize it, the hot filament activation is not a productive avenue, because of the slow diffusion transportation of active fragments to a substrate. Much more effective is the use of an expanded surface of a heat-resistant gas channel (a cylinder in our case). Besides, it allows us to control the gas delivery more thoroughly by operating in a rather wide range of regimes (flow rates, stagnation and background pressures, temperatures, etc.).

The successful experiments on deposition of polymer films from high velocity tetrafluoroethylene produced from polytetrafluoroethylene in the high temperature chamber (see [6]) gave an important impetus to creating the new method. The first experiments with activation of the mixture hydrogen and methane in the channel from the tungsten foil which contains a tungsten spiral, have shown the possibility of deposition of diamond structures deposition from a high velocity flow [7].

2. General peculiarities of the method

The most essential characteristic of the new approach to diamond synthesis is the using of multiple collisions of gases to be activated with hot surfaces of the channel directing the flow to a substrate. Fig. 1 depicts a rather simplified scheme of the new technology in an experimental realization. The mixture hydrogen and methane enters through the molybdenum cylinder to the activating heat-resistant cylinder (for example, made of a tungsten foil; red in the figure). Such elements of the device are described in [8]. On the foil surface of temperature 2000 K and higher, the partial decomposition of H_2 and CH_4

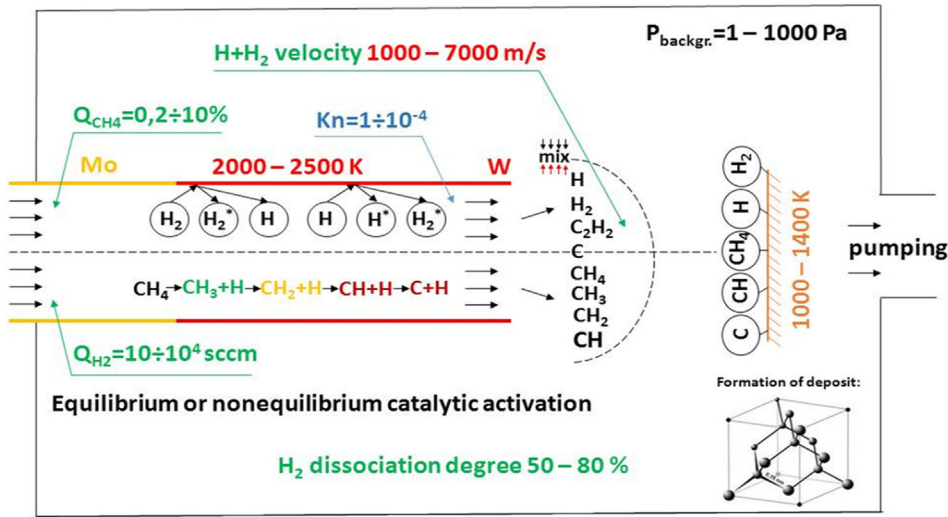


Fig. 1. Principal scheme of the new method with specification of operation conditions.

takes place. The mixture of the rest entering gases and their fragments expands in the direction of the substrate (the surface for deposition) whose temperature is 1000–1500 K lower than that of the foil.

The flow between the exit from the cylinder and substrate depends on the regime of use in experiments and can vary being
 a free molecular flow,
 a free supersonic jet with a shock wave at the substrate surface,

a free supersonic flow mixing with the background gas, or,
 a subsonic jet flooded in the background gas.

The ranges of operation conditions are pointed out in Fig. 1. They justify the uniqueness of the capabilities of the method. In fact, the range of background pressures is 10^{-1} – 10^5 Pa; the hydrogen flow rate, 10^{-1} – 10^4 sccm; the Knudsen numbers, 1 – 10^{-4} ; the temperature of the activating surface, 2000–2500 K; and the dissociation degree of hydrogen, 50–80%. No prior method, mentioned in the introduction, admits so wide ranges of parameters.

3. Gas dynamic modeling is a valuable instrument to predict and analyze flows

Numerical modeling is inevitable for the above- mentioned variety of gas dynamic structures. The most effective method for a rather low gas pressure is the direct simulation Monte Carlo (DSMC). The axis-symmetrical algorithm of DSMC [9,10] was adapted to calculating flows in cylindrical channels and past them. The verification of the program in the axisymmetric formulation was carried out for a flow without chemical reactions [11].

The calculation region (Fig. 2) is divided into 1.25×10^5 ring cells; 5.5×10^6 particles were used.

In calculations the model of soft spheres with variable cross-section for translational relaxation was used [12]. For the simplification of computation the internal energy of hydrogen molecules was not taken into account. It has no essential influence on gas dynamic structure in the cylinder and before substrate. According to the chosen calculation scheme, molecular hydrogen with room stagnation temperature enters

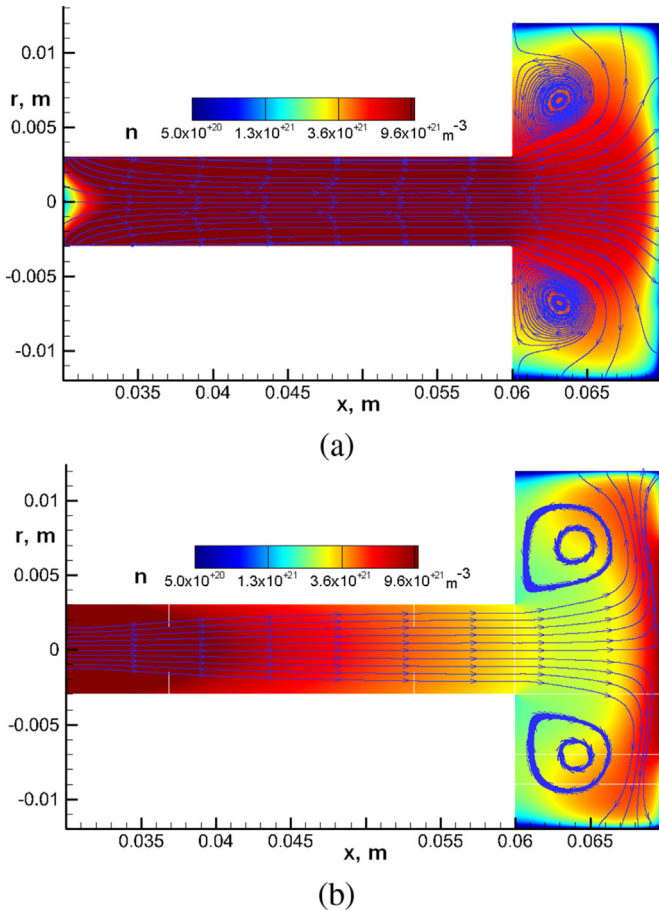


Fig. 2. Distribution of density and streamlines a) of atomic hydrogen; b) of molecular hydrogen.

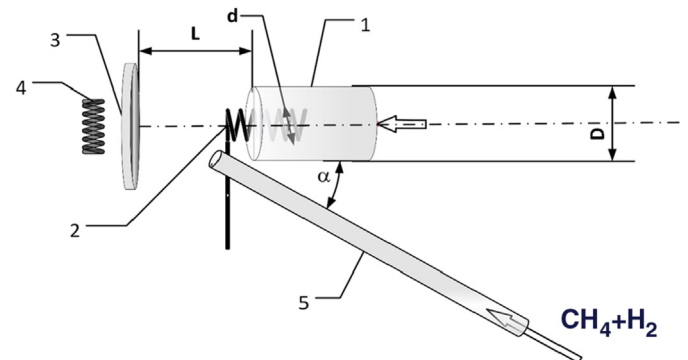


Fig. 3. Schematic of the experiment with separate supplies of gases.

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