# Agglomeration Process of Carbon Dusts in Laboratory Plasmas*)

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Micro-meter size carbon dusts have been formed in a 70 mm diameter, 370 mm long plasma container after running a 16 hour steady state Ar discharge. The formed carbon dusts have been observed by a scanning electron microscope (SEM) to identify their surface structures. The observed images have indicated that dusts with both flake and cauliflower structures were present on the surfaces of sample collectors. Carbon dusts with the flake structures exhibited smoother surfaces than dusts with cauliflower structures. Dust samples were also collected from arc spot parts of the carbon electrode surfaces by placing carbon compound adhesive tapes. Micro-meter size dusts were found collected on the surface of the tapes, indicating formation of dusts during arc discharges.



Keywords: carbon dust, hydrogen, agglomeration, plasma wall interaction, sputtering

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## 1. Introduction

Dust formation process in edge plasma attracts extensive attentions from the viewpoint of estimating tritium inventory in a nuclear fusion device [1]. This is because that small dusts having large surface to volume ratio are expected to absorb considerable amounts of tritium atoms and molecules. Carbon made first walls of a fusion device emit carbon clusters and hydrocarbons, which diffuse into edge plasma and agglomerate to form dusts in low temperature region of the edge plasma.

The origin of carbon dusts in fusion experiment devices should be the surface of plasma facing carbon walls. Carbon should be released in the forms of atoms, clusters and particles from the walls most probably by sputtering [2–4]. They can be disintegrated to form carbon hydride molecules in the edge plasma, and may grow up to carbon dusts as they touch down onto the surface of a sub-micro-meter size carbon particle. Clarification of fundamental mechanisms from carbon atom emission leading to agglomeration of a carbon dust is indispensable to predict the size and spatial distributions of carbon dust particles in fusion experiment devices. The understanding of the fundamental process should also yield important information on the amount of dust contained in a future fusion device.

Carbon dusts are also known to exist in cosmic space as the mean free paths for molecular attachment reactions are shorter than interstellar distances [5]. Thus, study on dust formation in low temperature plasma has been started partly due to the interest to clarify the formation mechanism of large carbon hydride molecules identified in spectroscopic investigations for space sciences. Sizes and shapes of carbon dusts formed in laboratory plasma conditions have been characterized by a scanning (SEM), transmission (TEM) electron microscopes, and focused ion beam (FIB) systems [6]. The SEM images have indicated that the productions of carbon dusts with an agglomeration of sub-micro-meter size particles. Measured dust size distribution exhibits a broad peak.

A small experimental setup has been designed and built to form carbon dusts to clarify the governing factors for dust formation processes in edge plasma of a fusion experiment deice. The designed system confines plasma inside of a graphite container, and micro-meter size carbon dusts have been observed by a scanning electron microscope (SEM) after plasma discharge operation. The comparison between the shape of formed dusts and the predictions obtained by theoretical model calculations should yield deeper understanding on how carbon dusts are formed in an actual fusion experiment device.

## 2. Experimental System

Shown in Fig. 1 is the designed experimental system that confines a glow plasma inside of a graphite container. The container is installed in a 35 mm inner diameter, 370 mm long vacuum chamber made of stainless steel and glass. A carbon cathode is installed inside of the graphite container that serves as the anode of the discharge. Two kinds of sample collectors, $SiO_2$ glass pieces, and carbon compound tapes, were placed on surfaces of the anode and the cathode.

Formed carbon dusts can be pumped out of the sys-

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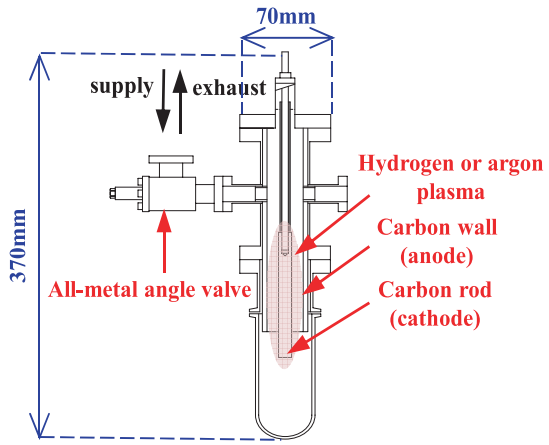



Fig. 1 Schematic diagram of experimental system.

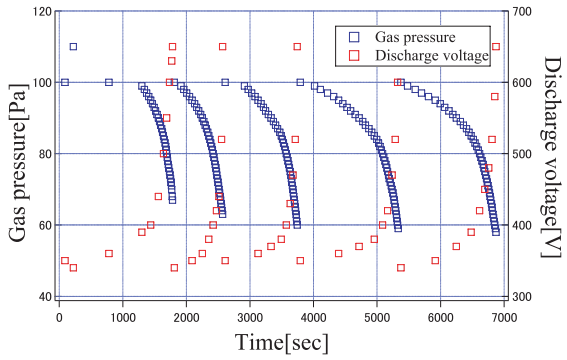
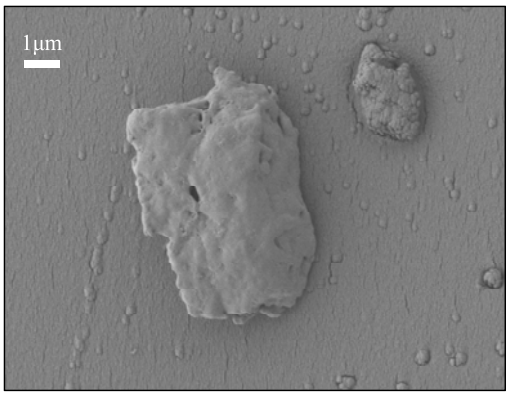
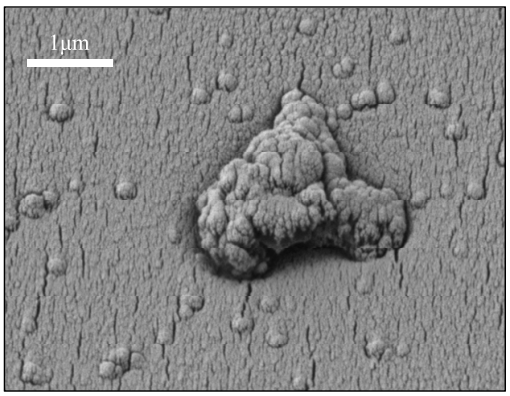



Fig. 2 Time dependence of gas pressure and discharge voltage for 100 mA hydrogen discharge.




(a). Flake structure




(b). Cauliflower structure

Fig. 3 The SEM images of carbon dusts formed on the slide due to argon plasma discharge.

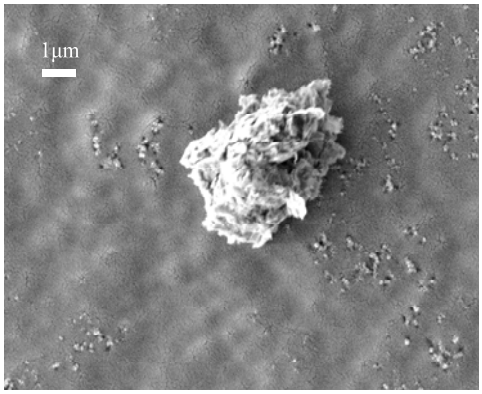
tem provided some gas flow exists in the plasma. In order to avoid any gas flow due to external supply and exhaust, the discharge was maintained with the sealed condition; the connecting valve to the gas supply and exhaust system was closed during the plasma operation. This procedure had made the continuous operation of hydrogen plasma extremely difficult. The pumping effect associated with glow discharge had rapidly reduced hydrogen pressure as shown in Fig. 2 and made steady state operation longer than 1 hour impossible. We are now modifying the discharge chamber structure to keep enough hydrogen pressure for longer period of time, and will report the result hereafter using Ar instead of hydrogen.

## 3. Experimental Results

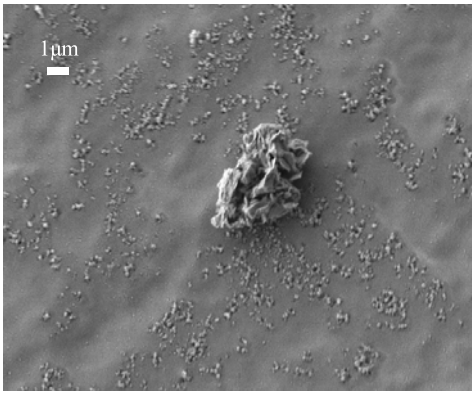
A stable Ar glow discharge had been maintained with the voltage about −650 V for 16 hr to form carbon dusts. The discharge chamber had been sealed by a metal seal angle valve after charging argon up to a gas pressure around 150 Pa. Argon gas had not been further supplied into the vacuum chamber during the discharge. As the consequence, Ar gas pressure of 150 Pa had reduced to 62 Pa after the continuous discharge operation. A glass target was placed below the carbon rod cathode, and was irradiated by argon plasma. The target was taken out of the system after the discharge operation to observe dusts formed on the surface by coating the sample surface with 5 nm-thick platinum to mitigate surface charge up during SEM observation. Figures 3 (a) and (b) show SEM images of carbon dusts formed on the surface of the glass target. Micrometer size carbon dusts have been observed, and the observed carbon dusts indicated both flake and cauliflower structures.

There had been some occasions that the arc discharge occurred in the initial phase of running a glow discharge. The carbon electrodes have been severely eroded by the arc discharge, and the electrode surface required thorough polishing to re-ignite a stable glow discharge. In order to confirm the formation of carbon dusts on the surface eroded by an arc discharge, carbon compound adhesive tapes were pressed onto the surface of the carbon container and that of the carbon cathode rod after the arc discharge. The surface of the carbon adhesive tapes had been investigated with a SEM. Micro-meter size carbon dusts of both flake and cauliflower structures have been observed.

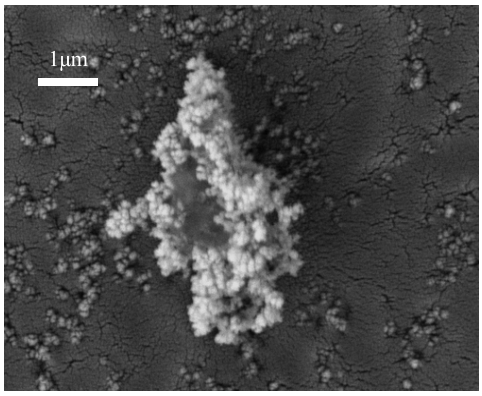
The carbon adhesive tapes were placed below the graphite container and carbon rod as sample collectors and Ar plasmas were run for 4 hr and 8 hr. Constant current discharge at 15 mA current had reduced the original Ar gas



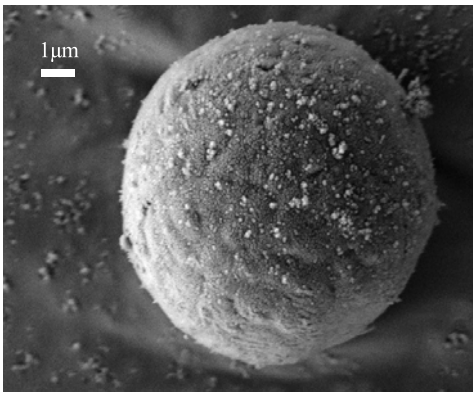

(a). Flake structure grain, 4 hour discharge.




(b). Flake structure grain, 8 hour discharge.




(c). Folded structure grain, 8 hour discharge.




(d). Spherical structure grain, 8 hour discharge.

Fig. 4 The SEM images of carbon dusts formed on carbon sample collectors.

pressure of 100 Pa to 91 Pa for 4 hr, to 58 Pa for 8 hr, respectively. Figure 4 (a) shows a typical SEM image of a carbon dust formed on the sample collector surfaces after 4 hr of Ar plasma discharge. Individual particle was found predominantly with flake structure with micrometer size. As the duration of discharge has been prolonged up to 8 hr, we have seen several dusts with different structures other than flakes. Though several dusts have sizes and structures like the ones found in 4 hour discharge operation, as shown in Fig. 4 (b), several dusts with structures with folding, and spherical shape were found, as shown in Figs. 4 (c) and 4 (d), respectively. Namely, elongation of discharge time from 4 hour to 8 hour seemed to advance the stage of agglomeration of dusts.

## 4. Discussion

Both flake and cauliflower structures carbon dusts have been observed on the glass dust collection target placed in an argon plasma discharge. The SEM images have indicated the surface conditions of the two type carbon dusts are difference. The surface of flake structure dusts appeared to be much smoother compared with that of the cauliflower structure dusts as shown in Figs. 3 (a) and (b). The surface conditions of carbon dusts have been different even though both carbon dusts have similar structures like flakes. The difference in the structure and that in the surface roughness give valuable hints on the existence of different formation paths up to the development of observed micro-meter size carbon dusts. There are some kinds of carbon dusts in fusion plasma [7]. A part of the carbon deposited layer is formed on the surface of the inner diverter tiles, and then the layer is peeled off by the thermal load and plasma irradiation. Cauliflower shape is formed at the growth by the interaction with the plasma but has the formation process has not been completely understood yet [8].

We have also found that there were a lot of smaller grains around the large size carbon particle. Because the surface of the glass collector plate was considered much smoother than graphite collectors, these small grains observed on the glass collectors were thought to be the precursors of larger dusts. However, these small grains may possibly be attributed to the platinum coating that had been made to diminish surface charge up which forbids SEM observation of the target surface. To answer this question, carbon deposition onto an electrical conducting material is underway.

The source of the observed carbon dusts on $SiO_2$ collectors should be the carbon wall and carbon rod in the vacuum chamber under Ar plasma bombardment. Released carbon should be in the forms of atoms, clusters and particles from the walls and the rod. The molecular dynamics calculations have predicted that the carbon dusts developed in gas phase have two dimensional structures in which sp2 orbital is dominant like graphene, and carbon dusts devel-

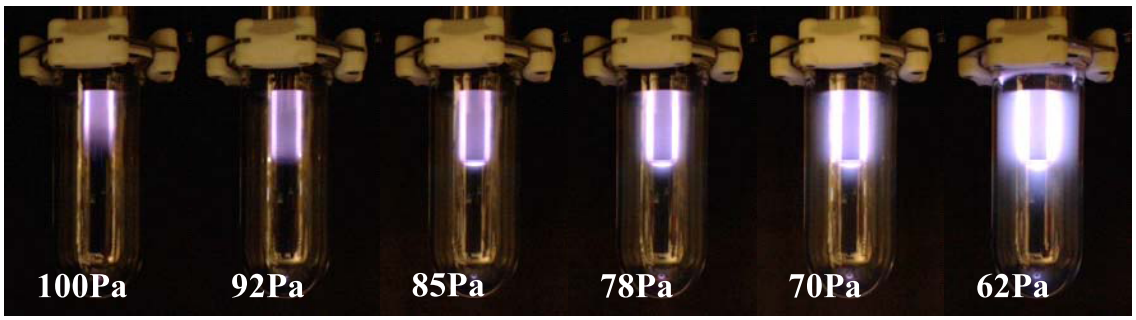



Fig. 5 Structure of hydrogen plasma around carbon cathode rod in the vacuum chamber monitored by a CCD camera.

oped on material surface have three dimensional structures for which sp3 orbital is dominant [9]. Judging from the smoothness on the surface, some of the observed carbon dusts are considered to grow on the surface of the walls of carbon target and/or the vacuum chamber.

Formation process of carbon dusts was not understood whether they grow in vapor phase or grow at the surface of materials [10]. Carbon collectors employed in these experiments are carbon compound adhesive tapes that can emit gas even more than the graphite anode and cathode. Kinds of gasses emitted from carbon sample collectors were examined by a Quadrupole Mass Analyzer (QMA: Balzers Prisma-200). Hydrocarbon gases were detected from a carbon compound adhesive tape used as the collector. Namely, the dust collectors produce hydrocarbons that can be the origin of dust formation. Small grain like structures seemed buried in the surface of collector as shown in Fig. 4. One may notice that the number density of small grains on the surface can be larger for 8 hour's case than 4 hour's case. The effect due to gas emission from the carbon sample should be investigated further, so as to exclude the process irrelevant from the formation of dust in fusion experiment devices.

We have confirmed that the designed experimental setup is at least enough to form carbon dusts with an Ar plasma discharge. The formation process of carbon dusts in low temperature inert gas plasma is important to investigate, because inert gas may be puffed into the diverter region in a fusion device to reduce kinetic energies of plasma particles for avoiding erosion of diverter plates due to high energy plasma particle bombardments. The formation of carbon dusts during hydrogen plasma discharge is certainly more important, because hydrogen isotopes are the fuel for nuclear fusion and are expected to form carbon dusts through several different channels of interaction with carbon first walls. Thus, we have been preparing investigation on the formation process of carbon dusts under the hydrogen plasma discharge, but have not been successful due to discharge pumping problem.

As we have presented in Fig. 2 hydrogen plasma operation with 100 mA discharge current rapidly reduced pressure inside of the test chamber due to discharge pumping. We have tested to see if any discharge condition that keeps the hydrogen gas pressure stable for more than several hours exists. This is because long time discharge duration is required to form carbon dusts, and no dust formation has been observed in discharge period shorter than two hours under hydrogen discharge condition. In case of Ar, the pressure becomes stable after several times of gas recharge into the chamber. Namely, by repeating the procedure shown in Fig. 2, Ar plasma can be maintained at a constant discharge power. One possible solution to overcome rapid discharge pumping for hydrogen is to install a gas reservoir of enough capacity, and this is what we are installing to the present system.

The question of spatial distribution of formed carbon dust is a very important. We have investigated the number of dusts formed on the surface of carbon electrodes by collecting samples placing carbon compound adhesive tapes after the discharge operation. The method is inadequate to draw a quantitative result, but the tendency that more dusts are formed on the bottom surface of the anode than cathode has been recognized. When the distribution of dust number density on the carbon surface was investigated, more dusts were found toward the bottom of vertically sustained cathode carbon rod.

The spatial distribution of dust particles should be closely related to the geometrical shape of plasma itself. Figure 5 shows the change in shape of the positive column of hydrogen plasma around the carbon rod monitored by a CCD camera installed in front of the vacuum chamber. The discharge current had been kept constant at 100 mA, after filling up the discharge chamber with 100 Pa of hydrogen. As the hydrogen pressure decreases with time of hydrogen discharge, the plasma glow became more prominent. The region of positive column had been extended with decreasing the gas pressure of hydrogen. This indicates longer mean-free-path of electrons at lower pressure, and possible change of reaction processes in the region of larger positive column and that in the region of smaller dark spaces.

Plasma parameter of the positive column has been investigated by putting in a Langmuir probe at a position about 1 cm from the cathode carbon rod. The measured electron temperature and density were 4 eV and $10^8$ cm$^{-3}$,

respectively, for 50 mA discharge current at 100 Pa Ar pressure. The measured $I - V$ characteristics of the probe depended heavily upon the presssure, as the plasma spatial distribution showed rapid change as shown in Fig. 5. Thus, plasma parameters have to be studied after the discharge conditions are stabilized by installing the gas reservoir.

# 5. Summary

A small experimental setup has been designed and built to study the formation processes of carbon dusts. Micro-meter size carbon dusts have been observed on the surfaces of glass and carbon sample collectors placed at locations of carbon wall anode, and carbon rod cathode after Ar plasma discharge for 4, 8, and 16 hr. The observed carbon dusts have been characterized by a scanning electron microscope (SEM). The carbon dusts have two different structures of flake and cauliflower in the initial phase The surfaces of carbon dusts of the flake structure formed at the carbon wall anode are much smoother than those of cauliflower structures. These observations seemed to indicate that the formation processes to develop micro-meter size carbon dusts are different for a flake structure and a cauliflower structure Carbon dusts of other forms are also found on the surface of carbon sample collectors.

The molecular dynamics calculations have predicted the carbon dusts nucleate on solid surface have the three dimensional structure, and the present observation may be the evidence of dust formation on the glass target surface. The present system will be equipped with a laser observation system so that early stage hint of dust formation at the surface, or the sheath region can be detected. The fundamental process governing the shape of the produced dust will be further investigated using different kinds of discharge gas.

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