Suppression of Carbon Dust Formation by Nitrogen Injection into Hydrogen Plasmas in Detached Plasma Conditions

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Chemical erosion of carbon materials and dust formation in low-temperature and neutral particle–dominated plasmas were investigated using high-pressure inductively coupled plasmas. Experiments were performed with $Ar/H_2/N_2$ mixture plasma irradiation to graphite targets. The addition of just a few percent of nitrogen gas to hydrogen led to significant suppression of carbon dust formation on the graphite target. From optical emission spectroscopy, CN band spectra were observed strongly in $Ar/H_2/N_2$ plasmas with a decrease of CH and C_2 band emission intensity. These results showed that CN bond formation, which caused chemical erosion of carbon by producing volatile CN, HCN, and C_2N_2 particles, might have been a key suppression mechanism of the carbon particle aggregation.

Keywords: carbon dust formation, plasma-surface interaction, chemical sputtering, graphite material

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Carbon materials are used for the plasma-facing components (PFC) in fusion devices because of their superior thermomechanical properties. However, dust particles are formed by plasma–surface interactions in fusion experimental devices [1–5]. Because carbon dust retains large amounts of hydrogen isotopes, the dust particles in fusion reactors cause safety problems mainly concerning the tritium inventory. The suppression of dust formation is an important issue in future fusion reactors. In this study, we report on experiments conducted to investigate the influence of nitrogen injection into argon/hydrogen plasmas on carbon dust formation by using high-power inductively coupled plasmas (ICP) [6, 7]. Nitrogen injection has been considered and tested as one of the methods for tritium and co-deposits removal in carbon PFC [8, 9]. Carbon dust formation in addition to tritium removal efficiency should be studied in nitrogen containing plasmas.

Experiments have been performed in $Ar/H_2/N_2$ mixture plasma irradiation to an isotropic graphite target (IG-430U, Toyo Tanso Co. Ltd.) at a surface temperature of $\sim$1020 K. The argon gas flow rate is 101 Pa m^3/s, and the flow rates of hydrogen and nitrogen gas into the argon plasma are 3.4-5.1 Pa m^3/s and 0-0.51 Pa m^3/s, respectively. The working gas pressure is $\sim$4 kPa. The electron temperature is $\sim$1 eV. The atomic hydrogen and ion flux onto the graphite target are $\sim 10^{23}$ $m^{-2}s^{-1}$ and 2-8×10^{19} $m^{-2}s^{-1}$, respectively. These plasma features are very helpful while studying the chemical erosion of carbon and the dust formation mechanism in the detached divertor plasma conditions. The irradiation time is set to 180 minutes. The surface temperature is measured through a quartz window with a radiation thermometer. A Scanning Electron Microscope (SEM) was used to observe the generated dust particles on the targets.

The dominant erosion process of the graphite target is due to chemical sputtering by low-energy hydrogen atoms under Ar/H_2 plasma irradiation. Many carbon dust particles are observed on the graphite target eroded by chemical erosion [7]. Figure 1 shows the number density and size of the carbon dust particles, target weight loss, and optical emission intensity of CH (431.4 nm), C_2 (516.5 nm), CN (388.3 nm), and NH (336 nm) band spectra normalized to Ar I (750.4 nm) emission as a function of N_2 injection ratio into hydrogen, where the N_2 injection ratio is defined as the nitrogen gas flow rate normalized by the sum of nitrogen and hydrogen flow rates. The number density of carbon dust particles is obtained by counting dust particles with 20 SEM photographs having an area of $\sim$0.04 mm^2 each, which are sampled from the whole target surface area. The detection limit of this technique is $\sim$0.8 μm. It is found that carbon dust formation is strongly suppressed in the argon/hydrogen plasmas with a small amount of nitrogen gas injection. At increasing nitrogen gas injection ratio, the weight loss of the graphite target increases slightly and the number of carbon dust particles decreases drastically. The size of the dust particles reaches a peak at N_2 injection ratio of 0.1-0.3% and then decreases rapidly with increasing N_2 injection ratio. In addition, the significant appearance in the optical emission from CN and NH radicals is ob-

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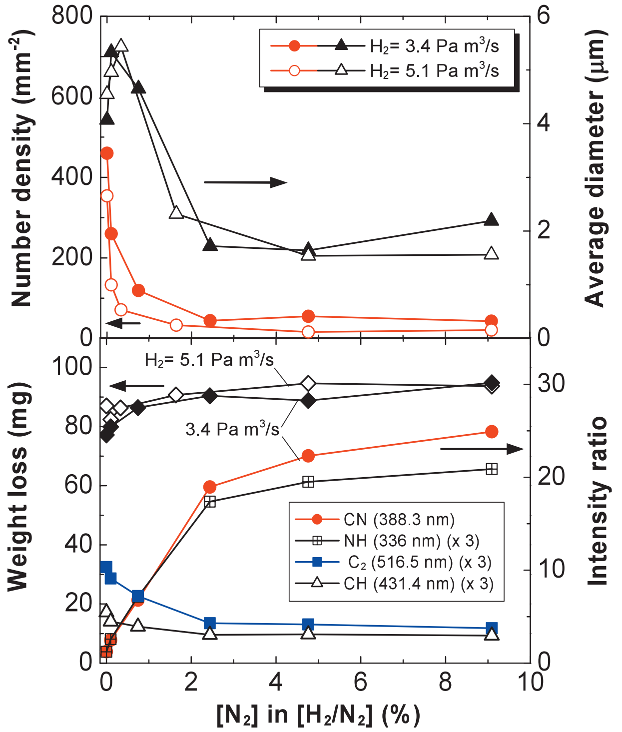

Fig. 1 Number density and average diameter of carbon dust particles observed on the target as a function of nitrogen injection ratio. The lower figure shows the variation of target weight loss and CH, C_2, CN and NH emission intensity normalized to Ar I emission.

served near the target surface. The CH intensity decreases by ~40% when the N_2 injection ratio is increased from 0% to ~9%, and the C_2 intensity decreases by ~60%. The increase in weight loss is caused by chemical sputtering of carbon by nitrogen atoms, which generates volatile CN, HCN, and C_2N_2 [10–12], and results in the increase in CN emission intensity. The production of CN and NH radicals by the reactions of hydrocarbon radicals with nitrogen atoms also contributes to the increase in CN and NH intensities. Carbon dust formation is determined by the competition between the erosion and agglomeration mechanisms of carbon. The enhancement of erosion by chemical sputtering by atomic nitrogen is considered as one of the causes for the suppression of carbon dust formation. Figure 2 shows SEM photographs of carbon dust observed on graphite surfaces irradiated by Ar/H_2/N_2 plasmas at different values of N_2 content. In the Ar/H_2 plasma, most of the carbon dust particles have a spherical shape at a surface temperature of ~1020 K [7]. With increasing nitrogen gas injection ratio, the carbon dust shape changes into polyhedral particles at an N_2 content of 0.3-0.7% and clusters made of smaller particles at an N_2 content > ~2%. These results show that the injection of a small amount of nitrogen (< 1%) leads to the agglomeration growth of fine carbon crystals, although the number density decreases. With increasing N_2 injection ratio above ~1%, however, the agglomeration process of carbon particles itself is strongly suppressed.

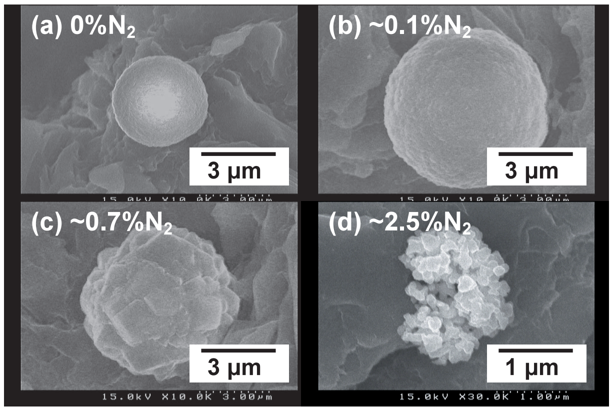

Fig. 2 SEM pictures of carbon dust particles formed on the graphite target irradiated by Ar/H_2/N_2 plasmas at different nitrogen injection ratio.

Ar/N_2 plasma irradiation experiments were performed to investigate carbon dust formation on the graphite surface eroded only by atomic nitrogen. There were no or only few dust particles on the graphite surface eroded by Ar/N_2 plasma irradiation. Moreover, C_2 band spectra were not observed during Ar/N_2 mixture plasma irradiation. From these observations, it seems that the increase of the concentration of C species bonded to N results in the interruption of carbon aggregation for an N_2 injection ratio of > ~1%.

From the calculation of the equilibrium composition of the C-H-N reactive molecular system, it is found that the number density of hydrocarbon and carbon species (C_2H, C_3, C_4, etc.) decreases compared to the C-H system at temperatures of 1000-4000 K, owing to the production of volatile HCN in this temperature range.

In conclusion, experiments in Ar/H_2/N_2 plasma irradiation to graphite material show that nitrogen injection into Ar/H_2 plasmas leads to the significant suppression of carbon dust formation in the number of carbon dust particles and increase in the optical emission intensity of CN and NH radicals with N_2 injection ratio of 0-1%. Moreover, SEM pictures of carbon dust show that the crystal growth of carbon dust is enhanced in this N_2 injection ratio range. In contrast, the variations of size and number density are not expected with a further increase of N_2 injection ratio. The suppression of carbon dust formation seems to be caused by the production of nitrogen-containing species from the chemical reactions in the gas phase and the chemical sputtering by nitrogen atoms.

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