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ARTICLE

Effect of nitrogen admixture to underwater argon arc discharge plasma for nuclear decommissioning

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Underwater arc discharge is being studied as one of the remote dismantling processes for fuel debris retrieval at the Fukushima Daiichi Nuclear Power Plant. In this study, the effect of admixture of N_2 molecules to argon arc is investigated to make the plasma with higher enthalpy. A steady-state DC arc thermal plasma is generated using a DC voltage between the anoxic copper anode and the 2%-thoriated tungsten cathode. The optical emission spectrum of the plasma plume was measured through a quartz window. The spectral analysis of Ar I lines was performed. The arc current and voltage were recorded. The total flow rate of the discharge gas was 25 L/min, where the nitrogen partial pressure ratio was adjusted in the range of 0 – 32%. It was found that the resistance of the arc plasma column increased as the nitrogen mixture ratio in the discharge gas increased. The electron temperature T_e became increased up to approximately ~ 2 eV with increasing the volumetric ratio of N_2 gas. On the other hand, it was found that the electron density N_e generally increases several times when nitrogen is mixed. The pH value of the irradiated water was found remarkably to decrease in the course of plasma irradiation in the open atmosphere.

Keywords: decommissioning by thermal plasma; argon-nitrogen arc plasma; optical emission spectroscopy

1. Introduction

Establishing a method for decommissioning the Fukushima Daiichi Nuclear Power Station is an urgent issue. One of the most important issues is the removal of fuel debris. Fuel debris has strong radioactivity and must be retrieved by remote control. At this time, it is necessary to cut and crush the clumped fuel debris to an appropriate size so that it can be easily removed, and a method of thermally performing this process by arc plasma has been proposed [1].

Cutting, crushing, and retrieval of the fuel debris should be carried out under submerged condition under water as much as possible, since radiation shielding and fuel cooling can be expected by the water [2]. Although there are several reports to describe cutting ability of debris-simulating materials with arc plasmas generated in water, there are very limited number of basic studies on their characteristics [3], while there are many characteristics of the arc plasmas generated under water or ejected into the water to be examined. Attempting to apply such underwater arc plasmas to nuclear decommissioning work, particularly to Fukushima-Daiichi Power Plant, inevitably entails the risk of severe accidents such as re-criticality of the nuclear fuel or diffusion of radioactive materials.

One of the essential features of the arc decommissioning is the elimination of optical fibers when the laser cutting method is introduced. Under the Fukushima Daiichi's radiation environment, the optical fibers will lose their transparency to guide the laser light, due to the gamma-ray-induced color centers. Therefore, the electrical discharge and arc plasma may be another candidate for the Fukushima's decommissioning process for cutting and crushing of structural materials as well as debris, instead of laser dismantling. From the above points of view, in order to realize safe decommissioning method by underwater arc discharge, the basic characteristics of the argon thermal arc plasma has been clarified from the fundamental viewpoint by the present authors' group. Their basic discharge characteristics of the argon thermal arc plasma have been investigated, such as electron temperature, density, flow velocity, heat transfer, and their dependence on the discharge conditions. However, the subject of the research so far has been limited to pure argon arc discharge plasma, which is easy to generate [4,5]. Due to the difficulty of handling, plasmas of gas

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mixture of argon and molecular gases have not been dealt with, since they frequently suffer from discharge instability or excessive damage to the electrodes for the arc discharge. However, the admixture of the molecular species to the mother-rare gas such as argon is basically expected to increase the enthalpy of the arc-plasma flow, and considered to be effective to thermal cutting or dismantling processes by the plasma method. In this application, the plasma should have not only “high-temperature” but also “high-enthalpy”, including high-density atomic radicals that can deposit larger energy to the solid materials to be decommissioned. If the plasma contains high-density atomic radicals even at the same temperature, surface recombination energy of the atoms into molecules at the target surface can effectively treat the decommissioned materials including nuclear debris.

Therefore, in this research, a gas mixture of argon and nitrogen instead of pure argon is adopted as the arc discharge process gas. In the previous study, a non-transferred arc plasma generated in the atmosphere, not in water, was examined, and it was confirmed that the addition of nitrogen to Ar significantly increased the power density of the atmospheric-pressure thermal-plasma jet, although the degree to which the temperature and enthalpy of the arc plasma flow increases in the water has not yet been well investigated [6]. This is exactly one of the objectives of this study. Secondly, another chemical advantage can be expected to the admixture of nitrogen to the under-water plasma. When the arc thermal plasma containing nitrogen gas is ejected into water, high-temperature nitrogen oxides could be locally generated due to chemical reactions of nitrogen radicals in the plasma and surrounding water molecule [7]. Consequently, an atmosphere rich in nitric or nitrous acid is created locally. Then, it is considered that the pH of water is lowered locally. That is, the plasma-irradiated portion may be decontaminated with a nitric acid or nitrous acid, which can realize the plasma decontamination of the surface of the dismantled objects. Consequently, measurement of changes in pH of the plasma-irradiated water is also considered as one of the important issues in this study.

2. Experiments

2.1. Experimental Devices

Figure 1 shows a schematic diagram of the present experimental setup, where (a) illustrates the system of arc discharge plasma generator and peripheral equipment applied in this study, and (b) is the structure of the electrode assembly and cooling water accommodated in the plasma chamber. An arc electrode consisting of an anode made of oxygen-free copper and a cathode made of 2% thoriated tungsten is placed. On the anode, a nozzle hole (~ 6 mm) is opened, and the arc plasma generated by arc discharge is blown out of this nozzle into the chamber on the feed-gas flow as a plasma-plume jet. That is, this plasma-plume jet is generated as a non-transferred arc discharge plasma continuously flowing to the downstream direction in the water. The inside of this plasma chamber can be roughly evacuated and replaced with any kind of gases, and can also be filled with tap water [4,5].

A resistor was connected in series between the two electrodes, and a DC power supply with drooping characteristics for general arc welder was used to supply the DC voltage. The cathode can be moved up and down with respect to the fixed anode by an external motor drive. After lowering the cathode and short-circuiting the electrodes, the motor drive direction is reversed to raise the cathode at a designated position and initiate the arc discharge. After that, the discharge voltage and the argon gas flow rate are set to desired values, and the underwater arc discharge experiment is performed. The arc current is recorded by a voltmeter connected to the output terminal of the power supply, while the arc current is picked up with a DC clamp meter surrounding the arc-current supply cable. The total discharge volumetric flow rate is fixed at 25 L/min[std] for any nitrogen concentration. Since the chamber is open to the atmosphere, the internal pressure in the chamber is always roughly equal to atmospheric pressure. After fixing the arc discharge current and voltage of the pure argon arc plasma to 80 A and 15 V, respectively, so that the conditions for the distance between the electrodes can be set constant as much as possible, the arc current is adjusted by the mixing ratio of nitrogen and the

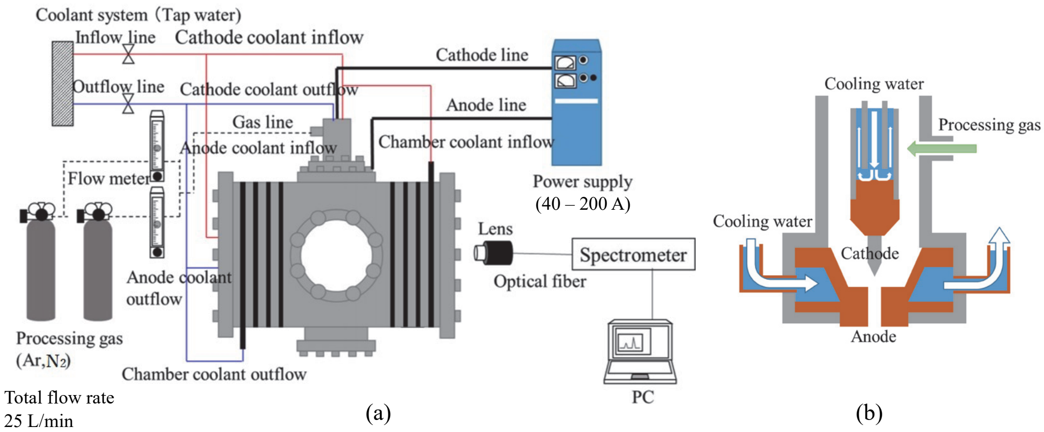

Figure 1. Schematic diagram of the present experimental setup: (a) the underwater arc discharge plasma equipment and peripherals, (b) structure of the electrode assembly and cooling water.

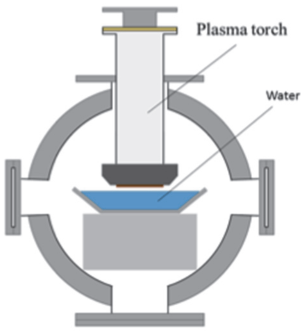

Figure 2. Schematic diagram of arc-torch-water arrangement condition for pH variation measurement.

applied arc voltage [4,5].

Next, an optical emission spectroscopic (OES) measurement of the underwater arc plasma plume is carried out [8]. The light emission of the plasma plume was measured by a spectrometer through a quartz glass window attached to the side of the chamber, and the dependence of the plasma parameters on the discharge conditions was analyzed using the OES measurement method. During the plasma parameter measurement experiment, the chamber is completely filled with tap water, and the arc plasma plume is ejected from the anode nozzle into the water in the chamber [4,5].

Finally, another experiment was carried out by reducing the amount of water to be irradiated in order to include the effect of entrainment of air in the pH measurement experiment, and to quantitatively detect the amount of pH change in water. In this experiment, as shown in **Figure 2**, 1 litter of water was stored in a pull metal vat (water depth 3 cm), and the water surface was placed so as to be fixed at the distance to contact the anode nozzle face of the plasma torch.

3. OES measurement

The target of this research is the atmospheric-pressure arc plasma generated in the underwater condition, which is classified as a thermal plasma. It is justifiably considered that the assumption of local thermodynamic equilibrium (LTE) holds well there. Therefore, the emission spectrum of the argon-nitrogen mixed arc plasma is measured using OES method [8], and determined the electron density and the excitation temperature determined from the excited-state populations, which is considered to be almost the same as the electron temperature.

3.1. The Boltzmann plot method

The line-spectrum intensity $I(\lambda)$ due to the transition from the upper-level p to the lower-level q at the wavelength λ is expressed by the following equation:

$$I(\lambda) = \frac{1}{4\pi}\frac{hc_0}{\lambda}\frac{g_p}{g_i}A_{pq}N_p\exp\left(-\frac{E_p}{k_\mathrm{B}T}\right), \tag{1}$$

where h is the Planck constant, c_0 is the speed of light, g_p and g_i are the statistical weights of the level p and that of the ion, respectively, A_{pq} is the transition probability from the level p to q, N_p is the number density of the level p, E_p is the excitation energy of the level p, k_B is the Boltzmann constant, and T is the temperature of the system in the equilibrium state. From Eq. (1), the following practical description is obtained:

$$\ln\left[\frac{I(\lambda)\lambda}{A_{pq}g_p}\right] = -\frac{E_p}{k_\mathrm{B}T} + \ln K, \tag{2}$$

where K is a constant summarized as a coefficient.

When the line intensity emitted from the specific excited sates belonging to the same atomic species is plotted semi-logarithmically according to the term in the left-hand side of Eq. (2) on the vertical axis while E_p on the horizontal axis, the reciprocal of the slope of the approximation line connecting them is proportional to the temperature T, which determines the value of the temperature of the arc plasma. This method is called the Boltzmann plot method [4,5]. The temperature obtained from this relationship is often referred to as the excitation temperature T_ex [8]. In non-equilibrium plasmas, T_ex generally does not agree with the electron temperature T_e. However, it can be considered that T_ex becomes almost the same as T_e in a steady-state high-current arc plasma with a discharge current of several tens of amperes or more where the LTE condition is established. It should be noted that in the Boltzmann plot method, the accuracy of the determined temperature can be further improved by selecting spectra in which E_p is widely distributed and whose wavelengths are close to each other [4].

3.2. The Stark broadening method

Light-emitting atoms in the plasma are affected by the microscopic electric field caused by their surrounding charged particles as the Stark effect, causing broadening of the spectral lines, which is referred to as the Stark broadening. Especially for hydrogen atoms, the Stark broadening of the emission line is so remarkable that can be measured with a general-purpose spectrometer. Since this microscopic electric field depends on the number density of the charged particles, the ion density and electron density in the plasma can be measured from the Stark broadening of the spectral line of the Balmer series of hydrogen atoms.

Let $\Delta\lambda_\mathrm{S}$ [nm] be the Stark broadening in the 656.28 nm emission spectrum (H_α line) of the Balmer series of hydrogen atoms. Then, the electron density N_e [cm^{-3}] of the plasma is given by the following equation [4]:

$$N_\mathrm{e} = 10^{17}\times\left(\frac{\Delta\lambda_\mathrm{S}}{1.098}\right)^{1.471}. \tag{3}$$

Spectral lines observed in the plasmas often have a Voigt function-type shape, in which profiles are superimposed by two or more factors. The broadening of the H_α line can also be deconvoluted into Gaussian and Lorentzian broadening. In atmospheric pressure arc plasmas, the profile of the Stark broadening can be almost entirely regarded as Lorentzian [4].

3.3. Saha's equation

When the plasma is in the state of thermodynamic equilibrium, the ionization degree of the gaseous plasma can be obtained as a function of its temperature, density, and ionization energy of the gas, which is formulated as Saha's equation. The Saha's equation gives the electron density as a function of the electron temperature, which is described as follows [9]:

$$\frac{X^2}{1-X} = \frac{1}{Nh^3}(2\pi m_e k_B T)^{\frac{3}{2}} \exp\left(-\frac{\chi}{k_B T}\right), \quad (4)$$

where N is the summation of the number density of the neutral molecules and that of the ions, m_e is the electron mass, χ is the ionization potential of the ground state of the neutral molecule, and X is the ionization degree defined as

$$X = \frac{N_e}{N}. \quad (5)$$

As later explained, the line intensity of the H_α line becomes very weak when the nitrogen gas is added into the argon arc discharge. At this occasion, the electron density is determined by Eqs. (4) – (5), instead of Eq. (3). When the temperature T of the plasma is obtained from the Boltzmann plot, the total number density N in Eqs. (4) – (5) is determined by the ideal gas law. Then Eq. (5) can be solved for the ionization degree X as a quadratic equation.

4. Results and discussion

4.1. Current-voltage characteristics of the arc plasma

Figure 3 shows the measurement results of the current-voltage characteristics of the underwater arc plasma. These are the results of measurements at nitrogen gas volumetric flow rates of 0, 8, 16, 24 and 32%. In Fig. 3, the horizontal axis represents the current value, while the vertical axis represents the resistance value (voltage/ current) of the underwater arc plasma. It was found that regardless of the nitrogen content, the monotonic decreasing dependence of the arc current on the discharge voltage was confirmed [4]. However, the absolute value of the resistance of the arc plasma was found to become larger in the lower current region and smaller in the higher current region. In addition, it can be seen that the arc plasma resistance increases as the nitrogen concentration increases, regardless of the arc-current value. In addition, at low discharge voltage, the difference in the resistance value of the arc plasma becomes remarkable depending on the mixing ratio of nitrogen. On the other hand, in the high arc-discharge current region, the resistance values are almost the same irrespective of the mixing ratio of nitrogen. The reason for this phenomenon is considered to be that the excitation or ionization energy of nitrogen molecules is greater than that of argon.

4.2. Excitation temperature of the nitrogen-argon plasma determined from the excited-state population of Ar I of the arc plasma

Figure 4 (a) and **(b)** shows an observed spectrum of the underwater arc argon plasma without nitrogen

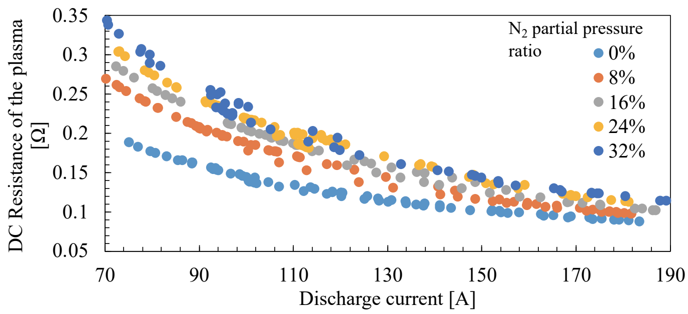

Figure 3. Dependence of DC resistance value of the arc plasma plotted against arc discharge current for several values of nitrogen partial pressure in the discharge gas.

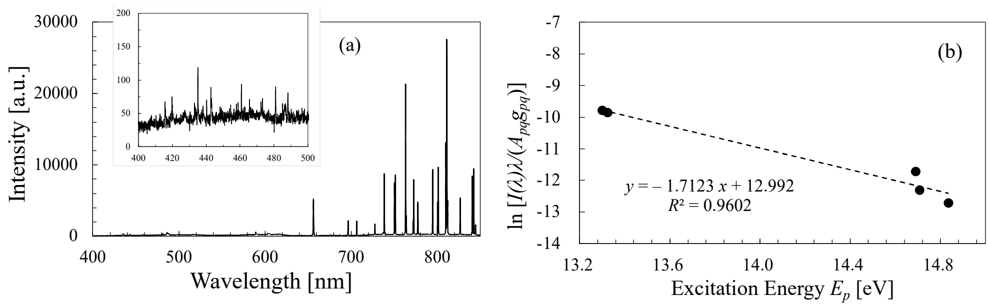

Figure 4. (a) Observed spectra of the underwater argon plasma without nitrogen admixture with the discharge current of 130 A and argon flow rate 25 L/min with an enlarged portion over the wavelength region 400 – 600 nm by inset, and (b) corresponding Boltzmann plot of Ar I populations.

admixture with the discharge current of 130 A, and the resultant Boltzmann plot drawn with the observed populations of argon atoms, respectively. Many spectral lines are observed in the wavelength regions of 400 – 450 nm and 700 – 800 nm in Figure 4(a), which are identified to be the lines of Ar I. The observed H_α (656 nm) and H_β (486 nm) lines are attributed to the Balmer series, emitted by the excited hydrogen atoms, which are generated by decomposition of discharge water in the chamber by the thermal arc plasma. In the Boltzmann plot Figure 4(b), five Ar I lines were chosen, whose spectroscopic characteristics are summarized in Table I. The intensity of these lines integrated over each peak directly gives $I(\lambda)$ in Eq. (2), which yields Figure 4(b) with the spectroscopic data specified in Table I. In the previous study of the underwater argon plasma, it was already established that these excited levels were most appropriate to determine the excitation temperature as the appropriate value of the electron temperature as in the state of thermodynamic equilibrium from the viewpoint of OES measurement [4]. The maximum difference in the wavelength in Table I is approximately 20 nm, which can be covered in a single-shot observation of our spectrometric system. On the other hand, the energy difference of the upper levels should be larger to improve the reliability of the temperature as the slope in the Boltzmann plot, and consequently, these five lines have been chosen in the previous study [4, 5].

In conducting the present experiments, it was found that the higher the nitrogen flow rate, the more unstable the underwater arc becomes. Consequently, regarding data with a high nitrogen admixture ratio, the measurement could not be performed in the low-power region, and the upper limit of the volumetric mixture ratio of nitrogen feed was found to be 32%. Furthermore, when nitrogen is mixed, the cooling water of the electrode starts boiling owing to the thermal load by the high-power arc discharge. This phenomenon is attributed to the high enthalpy of the underwater arc or the excessive power applied to the electric arc discharge circuit. In this case as well, it was considered to be dangerous to continue the discharge experiment for a long time, and consequently, the measurement was quitted.

Figure 5 (a) and **(b)** shows the electron temperature and the electron density, respectively, plotted against the arc discharge current under several conditions of nitrogen admixture ratio. Concerning the electron temperature, it is considered to be the same with the excitation temperature in the atmospheric-pressure arc plasma since the plasma is considered to be in the state of LTE. Generally speaking, when nitrogen is mixed into the underwater arc with argon gas, the excitation temperature determined with the Boltzmann plot increases throughout the present experiments. One of the reasons is that nitrogen molecules have higher ionization energy, i.e., higher enthalpy per unit mass, compared to argon atoms. Furthermore, nitrogen molecules are dissociated into the nitrogen atoms in the thermal plasma, and when they come into contact with water, they recombine to form molecules, which also enhances the heat transfer coefficient due to the deposition of the recombination energy to the surrounding water. As Figure 5(a) shows, the small amount of N_2 admixture has a large impact on the temperature of the arc plasma, while the large amount of admixture may change the thermal structure of the arc plasma due to the difference in the thermal conductivity.

Table 1. Emission spectra and spectroscopic constants applied to create the Boltzmann plots [4].

Designation of the transition	Wavelength of the line λ_{pq} [nm]	Excitation energy of the upper level E_p [eV]	Transition Probability A_{pq} [10^7 s^{-1}]	Statistical weight of the upper level g_p
$4d[1/2]_1 - 4p[1/2]_1$	687.13	14.71	0.278	3
$4d[1/2]_0 - 4p[1/2]_1$	693.77	14.69	0.308	1
$4p'[1/2]_1 - 4s[3/2]_2^o$	696.54	13.53	0.0639	3
$6s[3/2]_2^o - 4p[5/2]_3$	703.03	14.83	0.267	5
$4p'[3/2]_2 - 4s[3/2]_2^o$	706.72	13.30	0.380	5

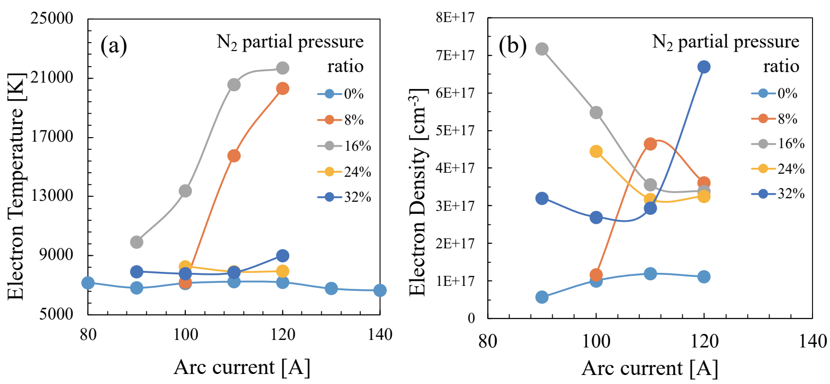

Figure 5. Electron parameters of the underwater N_2-Ar arc plasma. (a) Dependence of the electron temperature of the underwater nitrogen-argon mixed arc discharge plasma measured for different N_2 mixture ratio, and (b) that of the electron density.

4.3. Electron density of the nitrogen-argon arc plasma

Meanwhile, as for the electron density, when the nitrogen partial pressure was increased, it became difficult to confirm the H_α line to find the Stark broadening. Hence, the electron density was evaluated by Saha's formula, Eqs. (4) – (5). The reason for this phenomenon is considered to be that a nitrogen molecular band spectrum, the First Positive System (1PS), derived from nitrogen plasma is observed around the peak of the H_α line, which hinders clear observation.

It was found that the electron density generally became several times larger when the nitrogen gas was mixed than that of the pure Ar underwater arc plasma, but no regularity was observed for the arc current dependence. In the evaluation of the electron density in the present study, the Saha's equation was applied. Meanwhile, this equation assumes the establishment of thermodynamic equilibrium. In the case of the underwater arc plasma, it is indeed in the state of thermodynamic equilibrium when the arc plasma is generated between the electrodes. However, recombination reaction of the plasma progresses rapidly in the underwater condition, where optical emission spectroscopy is performed, which may cause the non-equilibrium population kinetics between the excited states of the argon atom. Therefore, there remains the possibility that the plasma is not in perfect LTE.

4.4. pH evaluation of the plasma irradiated water

Figure 6 (a) and **(b)** shows the relationship between the plasma-irradiation duration and the H^+ cation concentration in the water to which the plasma is irradiated at each nitrogen partial pressure, where the vertical axis of (a) is in pH units and that of (b) is in molar concentration of H^+ cation in the water. The increase in the hydrogen ion molar concentration per unit time obtained from Figure 6 (b) is 3.4×10^{-7} mol·L^{-1}·s^{-1} and 3.2×10^{-7} mol·L^{-1}·s^{-1} at nitrogen partial pressure ratios of 8% and 16%, respectively, showing no significant difference. This is partly because the reaction of water molecules with nitrogen plasma to produce nitrous or nitric acid was sufficiently carried out at 8% of the volumetric nitrogen concentration, and was saturated to some extent. In addition, changes in pH were observed even during irradiation with a pure argon arc discharge plasma that does not contain nitrogen in the discharge gas. The reason for this is that when the arc plasma is injected into the water under the geometrical conditions shown in Figure 2, the air is brought in from the free interface, and nitrogen in the surrounding air are entrained in and the reaction progresses to form nitrous or nitric acid.

5. Conclusion

There was no significant difference in the current-voltage characteristics depending on whether or not nitrogen was mixed in the argon arc plasma. That is, even in the underwater arc plasma of nitrogen-argon gas mixture, a shift from negative resistance characteristics to positive resistance characteristics and a decrease in plasma resistance were observed with increasing the discharge current. However, it was found that the resistance of the arc plasma increases as the nitrogen inflow increases, regardless of the current value, because of the characteristics of nitrogen gas.

Namely, the admixture of the nitrogen gas to the argon arc results in the increase in the discharge power if the discharge current is kept constant. Consequently, the larger electrical energy is deposited to the arc plasma. Due to this mechanism, an increase in the excitation temperature was observed in the underwater arc argon plasma when nitrogen was added to the discharge gas. Since the plasma was in an approximate thermodynamic equilibrium state, the temperatures of electrons, ions, and neutral particles can be considered to similarly rise due to nitrogen admixture.

In the meantime, since it was difficult to detect the Stark broadening when the nitrogen was mixed, the electron density was calculated from the excited-state density distribution using the Saha's equation assuming thermodynamic equilibrium. As a result, it was found that, in general, when nitrogen is mixed, the electron density increases several times compared to the pure argon underwater arc discharge plasma. However, no regular rule was found for the dependence of the increment of the electron density on the arc discharge current.

When water was irradiated with argon plasma mixed with nitrogen, it was found that the pH of the irradiated water decreased monotonously. It was found that the hydrogen ion concentration in the water increased at a constant rate by the irradiation with nitrogen-containing arc plasma.

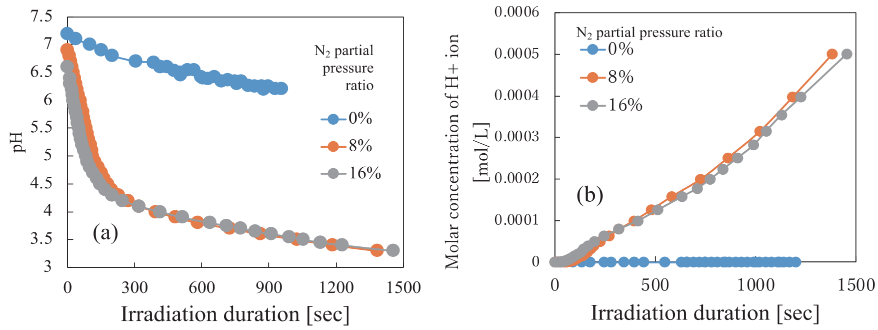

Figure 6. Variation in the hydrogen ion concentration of the underwater N_2-Ar arc plasma plotted against the plasma irradiation duration. (a) in the unit of pH, and (b) in the unit of molar concentration of H^+ ion.

This kind of nitrous or nitric acid atmosphere could be applied for the surface decontamination of the dismantled materials in the nuclear reactor decommissioning.

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