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主 論 文 の 要 旨

論文題目　Synthesis of Nanofluids and Nanocarbon using Controllable Solution Plasma (ソリューションプラズマ制御によるナノ流体とナノカーボンの合成)

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論 文 内 容 の 要 旨

I researched characteristic of solution plasma condition and controlling method for synthesis of nanofluids and nanocarbon. Typically, high active species such as ions, radicals and electrons were generated in solution plasma. In addition, collision was occurred as over current was flown by electrical breakdown of material. Therefore, solution plasma was developed due to their various applications which were synthesis and decomposition of materials using chemical and physical reaction. Those reactions were occurred at the same time. As a result, I found system temperature was related with small sized nanoparticles from part 1 to part 3. In addition, selective decomposition of organic compound was related with Coulomb force of solution plasma from part 4 to part 6.

In part 2, solution plasma condition was researched for synthesis of small sized nanoparticles. Thus, Characteristics of physical and chemical reaction were researched at short type solution plasma, Glow like solution plasma and arc type solution plasma. Typically, molecular of solution was excited by generation of plasma. However, not only elements of solution but also materials of electrodes were excited as glow like solution plasma transited to short and arc type solution plasma. Detail characteristics of physical and chemical reaction were researched at, an innovative type of plasma discharge in liquid, named solution plasma. During this specific type of plasma, various phenomena generally occurred such as glow like solution plasma, short type solution plasma and arc type solution plasma. Possible mechanisms of the transition of solution plasma were suggested. During plasma generation, hydrogen ions were accelerated by electric field. The erosion of electrodes and the rise of solution temperature can be explained by the kinetic energy of hydrogen ion. As a result, the kinetic energies of hydrogen ion were calculated to be 5.67 eV, 5.43 eV and 7.47 eV, respectively, for STSP, GLSP and ATSP. The temperature of the media (water) increased rapidly as the black body radiation was observed. In addition, tungsten atoms were excited when the kinetic energy of hydrogen ion increased. Severe erosion of electrodes was observed in STSP and ATSP. It can be explained by the electrical energy of solution plasma, where the electric field was perpendicular to each electrodes surface, anode and cathode was eroded in plane. In addition, the needle shape could be explained by not only vertical erosion of hydrogen ion but also corrosion on cylinder surface of electrodes in ATSP. In STSP and ATSP, the calculated surfacetemperatures were 4900 K and 5400 K, respectively. The tungsten electrode could be melted or evaporated prior to plasma discharge and contributed the significant generation of Tungsten active species. In this paper, we have analyzed the physical phenomena of solution plasma in an innovative dynamic approach. It is proposed that the approach can be beneficial to better knowledge of plasma in liquid process.

In part 3, nano-materials were synthesized for application using developed solution plasma. Small-size and well-dispersed gold nanoparticles useful as nanofluids have been synthesized in an electrical discharge in a liquid environment, termed solution plasma processing. The electrons and the hydrogen radicals are responsible for the reduction of the gold ion to the neutral form in the plasma gas phase and liquid phase, respectively. The gold NPs exhibit the smallest diameter of 4.9 nm when the solution temperature was kept at 20°C. In the paper, the nucleation and growth theory explains the diameter of the NPs after the reduction reaction closely related with the system temperature, the NP surface energy, the dispersion energy barrier, and the nucleation rate. The negative charge on the NPs surface during and after the synthesis in the SPP causes repulsive forces among the NPs avoiding their agglomeration in solution. The increase of the average energy in the SPP determines a decrease of the zeta potential and an increase of the NPs diameter. A large enhancement of the thermal conductivity of 9.4 % was measured for the synthesized nanofluids containing the smallest size NPs.

In prat4, the characteristics of synthesized nanofluids for coolant and electrode for fuel cell were investigated. The phonon thermodynamics theory for liquids was applied to explain the thermal characteristics of gold nanofluids synthesized by a simple, one-step, and chemical-free method using an electrical discharge in a liquid environment termed solution plasma process. The specific heat capacity of nanofluids was measured with a differential scanning calorimeter using the ratio between the differential heat flow rate and the heating rate. The decrease of the specific heat capacity with 10 percent of gold nanofluids relative to water was explained by the decrease of Frenkel relaxation time with 22 percent, considering a solid-like state model of liquids.

In part 5, low pass filter was constructed for advanced controlling of glow like solution plasma. In addition, control mechanism was researched. Elements of solution plasma such as radicals, ions and electrons were also behavior and controlled by electrical device. Thus, electrical circuit was introduced for controlling of solution plasma condition. Translation of plasma energy was reduced using low pass filter circuit in order to generate glow like solution plasma. Transfer of thermal energy was reduced from solution plasma to solution by interrupt of ion oscillation. Possible mechanisms for controlling of solution plasma condition using low pass filter circuit are suggested. First, total impedance of circuit was constructed in order to reduce effect of changed SP impedance. Second, parallel capacitor was consisted for separating with electron and ion oscillation from SP oscillation. Third, ion oscillation was delayed, because RC filter circuit causes time delay. Consequently, electron temperature was enhanced from 10,000K to 24,000 K. Electron density also increased from 2.14×10^{22} /m^{3}to 2.89×10^{23} /m^3 by using low pass filter circuit.

In part 6, In addition, advanced electrical characteristics were controlled by composition and electric capacity of low pass filter RLC circuit. Each components of circuit would show the following characteristics; Plasma oscillation was divided with ion and electron oscillation by a parallel capacitor of 2 nF. Maximum current was controlled by a series resistor when plasma was generated. In addition, pulse width of current was related with value of a series inductor. Therefore, the kinetic energy and ion oscillation of hydrogen ions could be controlled by Coulomb force. As the kinetic energy of hydrogen ions was controlled by electrical components, characteristic of solution plasma showed glow like condition which has low ion density of 7.45×10^{16} /m^3 and high electron temperature of 66000 ± 2900 K. Consequently, Glow like Solution Plasma (GLSP) and Arc type Solution Plasma (ATSP) could be controlled successfully by controlling of the instantaneous kinetic energy. The increase of the hydrogen excitation with 9 percent of GLSP relative to ATSP was explained by decrease of the Coulomb force with 20 percent.

In part 7, finally, pyridine structure conserved carbon nano-sheet also was successfully synthesized using controllable glow like solution plasma. Selective decomposition efficient of hydrogen increased by controlling of coulomb energy using low pass filter circuit. The increase of the excited alpha hydrogen with 26 percent was explained by the decrease of instantaneous Coulomb force with 22 percent. The increase of the excited alpha hydrogen means to remove a bonding of C-H in pyridine. However, CN bonding was conserved as glow like solution plasma was generated. Nitrogen conserved carbon nano-sheet could be synthesized to keep pyridine structure without heat treatment. Lattice spacing and ring pattern of {0004}, {10-10}and {11-20} was investigated using high resolution transmission electron microscope image and diffraction pattern. Nitrogen bonding with carbon was confirmed from FT-IR results. In addition, carbon nano-sheet was synthesized by interruption of energy transition. The glow like solution plasma produced ten to thirteen layered carbon sheet with the smallest average thick size, 2.6 ± 0.8 nm and sheet shape.