【Grant-in-Aid for Specially Promoted Research】

Science and Engineering

Title of Project : Creation of multi-element high entropy nano-alloys by non-equilibrium synthesis methods

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Keyword : Multi-element alloy, High entropy, Non-equilibrium synthesis, Catalysis

【Purpose and Background of the Research】

In this research, we will develop a new nano-sized solid solution alloys with many elements by the multi-element high entropy effect, and create an innovative catalytic functionalities. The supercritical solvothermal continuous flow synthesis method, which was originally developed by our research group, alloys various elements at the atomic level and we explore for new fusion-elements, new chemical substances, and new materials. Specifically, no one has succeeded so far in 1) fabrication of high-entropy solid-solution nanoalloys composed of multiple elements of precious metals, 2) high-entropy solid-solution nanoalloys composed of the elements of precious metals and base metals, 3) high-entropy solid-solution nanoalloys composed of noble, non-precious, and light elements.

Furthermore, by applying process-informatics, we will develop a comprehensive and innovative chemical process. Through this research and development, it is expected that we will be able to learn "advantages and characteristics of the elements" related to individual catalytic reactions through machine learning, which cannot be obtained from human experience.

【Research Methods】

We will establish solid-solution nano-alloys technology that makes full use of methods such as non-equilibrium synthesis, nanosizing, hydrogen process, and solvothermal continuous flow synthesis. So far, we have conducted research and development to mix metallic elements that never mix in the bulk state at the atomic level by using nanosize and non-equilibrium synthesis methods. However, in the method of reducing metal ions by spraying a metal precursors solution, there has been a limit to the increase in reduction speed, and it is difficult to instantly and simultaneously reduce metal ion species having largely different redox properties, so that they uniformly mixed was not successful. Therefore, by applying the ultrafast reduction continuous synthesis method (solvothermal continuous flow synthesis method) using hydrothermal and solvothermal reactions, we will establish a synthetic technology of multi-element solid-solution alloys in a non-equilibrium state in supercritical and subcritical. Solvothermal synthesis is a method of synthesizing a solid materials using a high-temperature and high-pressure solvent (or supercritical fluid), and when the solvent is water, it is called hydrothermal synthesis. Using this method, a solvent in which various metal ions are dissolved is instantly transferred to a supercritical/subcritical fluid, and each metal ion is instantly reduced and together alloyed into metal atoms under high temperature and high pressure, and the temperature is instantly decreased into room temperature. It is a method that enable the synthesis of a 1 nm class solid-solution alloy. With the exception of metal oxides such as ceria, there was no example of the synthesis of alloy nanoparticles until a few years ago. The features of the solvothermal continuous flow synthesis method originally developed by this project are 1) pressurized and heated up to 40 MPa and 450 °C, 2) even ethanol can be used as a reducing agent up to 300 °C, 3) metal ions can be reduced even when diluted with water to 10% ethanol, 4) three liquids injection at once is available, and 5) stable synthesis and mass production of solid-solution nanoalloys are possible.

【Expected Research Achievements and Scientific Significance】

Through this research and development, it is expected that we will be able to learn "advantages and characteristics of the elements" related to individual catalytic reactions through machine learning, which cannot be obtained from human experience.

【Publications Relevant to the Project】

- Platinum-group-metal High-entropy-alloy Nanoparticles, D. Wu, K. Kusada, T. Yamamoto, T. Toriyama, S. Matsumura, S. Kawaguchi, Y. Kubota, H. Kitagawa, ***J. Am. Chem. Soc.***, 142, 32, 13833-13838 (2020).
- New Aspects of Platinum Group Metal-based Solid-solution Alloy Nanoparticles: Binary to High-entropy Alloys, K. Kusada, D. Wu, H. Kitagawa, ***Chemistry - A European Journal***, 26, 5105-5130 (2020).

【Term of Project】 FY2020-2024

【Budget Allocation】 486,100 Thousand Yen

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