Synthesis of Nanostructured Bi_2Te_3 by Controlling Metal Species in Aqueous Solution

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1. Introduction

Bismuth telluride (Bi_2Te_3) is considered to be best materials for near room temperature thermoelectric (TE) applications. A value of dimensionless figure-of-merit (ZT) is required to be ~3 for a practical application. Recently, many efforts have been made to increase the ZT value of Bi_2Te_3 materials. It is reported that nanostructured TE materials in low dimensional form show a higher ZT value compared with bulk TE materials. There have been many researches for the synthesis methods of nanostructured Bi_2Te_3 such as solvothermal, hydrothermal, electrochemical method and sonochemical method. Since these methods normally require high temperature, high pressure, toxic regent and special equipment, it was difficult to achieve the practical use of TE. Therefore, what is of vital importance today is to develop a synthesis method in an aqueous solution to synthesize nanostructured Bi_2Te_3 for a green and low-cost synthesis method.

On the other hand, we reported that well crystallized and uniform metal alloy NPs could be synthesized by controlling metal complex in the aqueous solution based on metal complex calculations using the critical stability constants[1]. Needless to say, redox potential of metal ion and/or complex in solution varies according to their condition. This means that the reduction route could be controlled by controlling the metal complexes.

In this work, Bi and Te species for making homogenization of redox potentials are controlled by adding complex agent into aqueous solution and redox potentials of reducing agent are optimized by adjusting the concentration of reducing agent and pH for the synthesis of nanostructured Bi_2Te_3. In this method, nanostructured Bi_2Te_3 are synthesized at near room temperature, in the aqueous solution, without toxic reagents.

2. Experimental

Bi_2Te_3 was synthesized by using $Bi(NO_3)_3$, Na_2TeO_3, and ethylenediamine-N,N,N',N'-tetraacetic acid disodium salt (2NaEDTA) as metal precursor salts and complex reagent, respectively. Ascorbic acid was used as a reducing agent. The concentrations of metal species in the aqueous solution at various pH were calculated by using the critical stability constants. Metal species in the solution were identified by ESI-TOF-MS. The electrochemical analysis was performed to examine reduction potentials of metal species. The samples synthesized at each pH were characterized by XRD, SEM and TEM.

3. Results and discussion

Figure 1 and 2 show the XRD profile and TEM micrograph of the sample synthesized at pH10. White precipitations generated in the mixing solutions of Na_2TeO_3, $Bi(NO_3)_3$ and EDTA below pH 9 and above pH 11. In former case (<pH 9), TeO_3^{2-} is considered to change to TeO_2 precipitations, while Bi-OH complexes are formed and Bi_2O_3 precipitates in later case (>pH 11).

On the other hand, as shown in Fig.1, Bi_2Te_3 with rhombohedral phase (PDF#15-0863) was successfully synthesized. The morphologies of nano-structured Bi_2Te_3 are plate- like particles with a typical diameter 30nm and a thickness 10nm. At pH 10, TeO_3^{2-} ion and formed Bi-EDTA complex are stable. Then the stable Te ion and Bi complex can be reduced to nanostructured Bi_2Te_3 at pH10 by adding ascorbic acid. Therefore, the pH value plays a significant role in the formation of Bi_2Te_3 alloy.

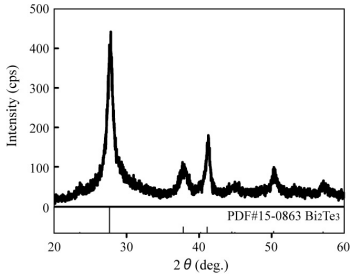

Fig.1 XRD profile of materials synthesized at pH10

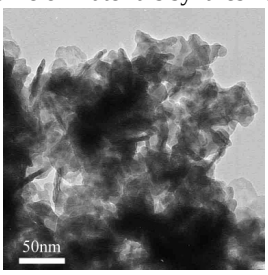

Fig.2 TEM micrograph of materials synthesized at pH10

In our presentation, detailed results, such as reduction potentials of metal species, optimization of reducing agents, from the viewpoint of electrochemical and ESI-TOF-MS analysis for the synthesis of nano-structured Bi_2Te_3 will be introduced. In addition, we will suggest a large-scale and low-cost synthesis method for nanostructured Bi_2Te_3.

4. Conclusion

Nanostructured Bi_2Te_3 can be synthesized by controlling metal species and optimizing the reducing agent in the aqueous solution at near room temperature, without toxic reagents.

Acknowledgement

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Reference

[1] H. Takahashi, N. Konishi, H. Ohno, K. Takahashi, K. Asakura, A. Muramatsu, Applied Catalysis A: General 392, 80-85 (2011)