

Spin Seebeck Devices Using $Ce_xY_{3-x}Fe_5O_{12}$ Deposited by Metal Organic Decomposition -Influence of Composition and Long Time Annealing-

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Abstract

Spin Seebeck devices with $Ce_xY_{3-x}Fe_5O_{12}$ deposited by metal organic decomposition with various Ce content, $x=0\sim 3$, have been investigated. It was found that as the Ce content is higher the electromotive force is lower for the samples annealed for 3 h at 900 °C, i.e., no Ce content is the best. Then the longer annealing time of 14 h at various temperatures (600 ~ 1050 °C) was carried out for the $Ce_1Y_2Fe_5O_{12}$ film and the 800 °C anneal resulted in the highest electromotive force of 24 $\mu V/50^\circ C$, which is better than that of sample with no Ce content and other reports concerning $Ce_xY_{3-x}Fe_5O_{12}$.

1. Introduction

Recently, studies of thermoelectric conversion technique are actively carried out for wasted heat energy recovery. Efficiency of thermoelectric conversion devices based on Seebeck effect is limited due to the Wiedemann-Franz law, in which the high electron density causes the high thermal conductivity [1]. However, in spin Seebeck effect devices (Fig. 1) are free from this law because magnetic electrically insulating film is used [2, 3]. When magnetic insulating film/metal junction is applied to temperature gradient, spin current is induced by spin Seebeck effect. Then, spin current which injected in metal layer converts into an electric field E_{ISHE} by inverse spin-Hall effect. The electric field E_{ISHE} is expressed as following equation.

$$E_{ISHE} = \theta_{SH}\rho (J_s \times \sigma) \quad (1)$$

where θ_{SH} , ρ , J_s and σ , respectively, represent spin Hall angle, electric resistivity, spin current vector and spin polarized vector [2].

As the magnetic insulation film, $Ce_1Y_2Fe_5O_{12}$ has approximately twice larger Faraday rotation than $Bi_1Y_2Fe_5O_{12}$ (Bi:YIG) [4] and a patent was applied for the spin Seebeck devices [5]. However, no journal paper except from our group [6] has been reported yet. We used metal organic decomposition (MOD) method to deposit $Ce_1Y_2Fe_5O_{12}$ [6]. However, the electromotive force of $Ce_1Y_2Fe_5O_{12}$ is lower than Bi:YIG and other report [5]. In this report we fabricated and evaluated spin Seebeck devices using $Ce_xY_{3-x}Fe_5O_{12}$ in order to increase the power conversion efficiency. We evaluated the relation between long time annealing and its spin Seebeck effect.

2. Electromotive forces of $Ce_xY_{3-x}Fe_5O_{12}$ ($x=0, 1, 2, 3$) films

We used $Gd_3Ga_5O_{12}$ (GGG) (111) as substrate. The recipe of fabricating $Ce_xY_{3-x}Fe_5O_{12}$ film is shown in Table 1. We mixed $Y_3Fe_5O_{12}$ and $Ce_3Fe_5O_{12}$ dip-coating agent by ultrasonic agitation to fabricate $Ce_xY_{3-x}Fe_5O_{12}$ ($x=0\sim 3$). The MOD process of $Ce_xY_{3-x}Fe_5O_{12}$ film was repeated 7 times to produce 280 nm thick film and finally annealed at 900 °C for 3 h in pseudo air ambient ($N_2:O_2=4:1$). The 3D mapping images of the $Ce_xY_{3-x}Fe_5O_{12}$ film by atomic force microscope (AFM) is shown in Fig. 2. As Ce content is higher the

surface roughness is increased and saturates. Then the $Ce_xY_{3-x}Fe_5O_{12}$ film was mechanically polished (MP) using diamond slurry with a diameter of 0.25 μm for 1 min. The 3D mapping images of Ce:YIG film after MP process is shown in Fig. 3. R_a value variation of $Ce_xY_{3-x}Fe_5O_{12}$ ($x=0\sim 3$) before and after MP is shown in Fig. 4. After cleaning the polished surface in acetone and pure water a 10 nm thick Pt was deposited. The result of electromotive force by spin Seebeck effect is shown in Fig. 5. Here, temperature difference between substrate and Pt electrode is 50 °C and intensity of magnetic field is 500 G. As Ce content is higher the electromotive force is decreased. We thought that this reason is that the garnet structure could not be formed when Ce content is over 2 [7].

3. Effect of long time annealing to spin Seebeck effect

The obtained electromotive force of $Ce_1Y_2Fe_5O_{12}$ is lower than other reports (13.4 $\mu V/50^\circ C$) [5], where $Ce_1Y_2Fe_5O_{12}$ was annealed at 680 °C for 14 h. For the above sample annealing was done at 900 °C for 3 h. We thought the crystallinity of $Ce_1Y_2Fe_5O_{12}$ film will be improved by the long time annealing. Then, the $Ce_1Y_2Fe_5O_{12}$ film of 280 nm thick was grown by MOD process and annealed at 600~1050 °C for 14 h in pseudo air ambient ($N_2:O_2=4:1$). X-ray diffraction (XRD) spectra of $Ce_1Y_2Fe_5O_{12}$ film annealed at various temperatures for 3 h and 14 h are shown in Fig. 6. Anneal temperature dependence of XRD peak intensity of $Ce_1Y_2Fe_5O_{12}$ (400) is shown in Fig. 8. $Ce_1Y_2Fe_5O_{12}$ film is crystallized when anneal temperature is over 800 °C and the XRD intensity of $Ce_1Y_2Fe_5O_{12}$ (400) by 14 h annealing is higher than 3 h annealing. Since the surface of $Ce_1Y_2Fe_5O_{12}$ films was rough, it was mechanically polished for 1 min. R_a value variation of $Ce_1Y_2Fe_5O_{12}$ annealed for 14 h before and after MP process is shown in Fig. 7. After cleaning the polished surface a 10 nm thick Pt was deposited. The result of electromotive force by spin Seebeck effect is shown in Fig. 9. $Ce_1Y_2Fe_5O_{12}$ annealed 800 °C for 14 h has the highest performance. The electromotive force is about two times higher than other reports (13.4 μV) [5]. Despite XRD peak intensity of $Ce_1Y_2Fe_5O_{12}$ annealed 1050 °C for 14 h is the best, the electromotive force was not increased so much rather decreased. We thought that the surface of $Ce_1Y_2Fe_5O_{12}$ film annealed 1050 °C for 14 h was modified.

4. Conclusions

We have fabricated spin Seebeck devices using $Ce_xY_{3-x}Fe_5O_{12}$ ($x=0\sim 3$) by MOD annealed at 600~1050 °C for 3 or 14 h. As Ce content is higher the electromotive force is decreased for the samples annealed for 3 h at 900 °C. When the longer annealing time of 14 h is carried out for the $Ce_1Y_2Fe_5O_{12}$ film, the 800 °C anneal was found to be best to obtain a highest electromotive force of 24 $\mu V/50^\circ C$, which is better than that of sample with no Ce content and other reports concerning $Ce_xY_{3-x}Fe_5O_{12}$.

References

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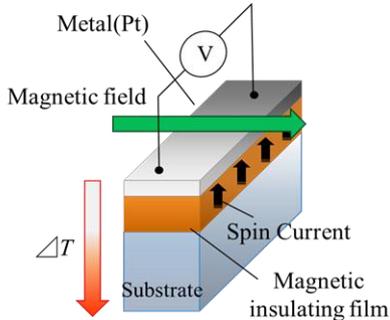


Fig. 1 Structure of spin Seebeck devices.

Table 1 Recipe of fabricating Ce:YIG films by MOD method.

Process	Condition
1. Spin-coating	500 rpm 5 s 3000 rpm 30 s
2. Drying (in air)	150 °C 3 min
3. Temperature rising	35 °C/min
4. Pre-baking (in air)	500 °C 5 min
5. Annealing (N ₂ :O ₂ = 4:1)	• 900 °C 3 h • 600 ~ 1050 °C 14h (Long time annealing)

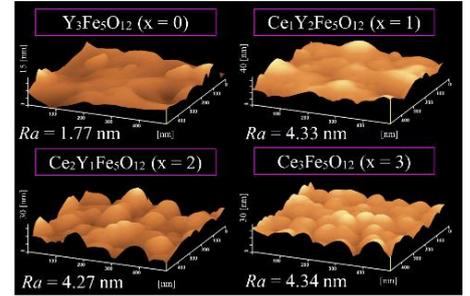


Fig. 2 3D mapping images of Ce_xY_{3-x}Fe₅O₁₂ (x=0, 1, 2, 3) films annealed at 900 °C by AFM (before MP).

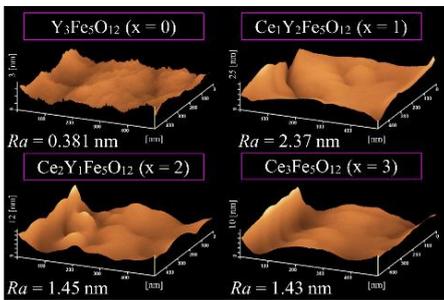


Fig. 3 3D mapping images of Ce_xY_{3-x}Fe₅O₁₂ (x=0~3) films annealed at 900 °C by AFM (after MP).

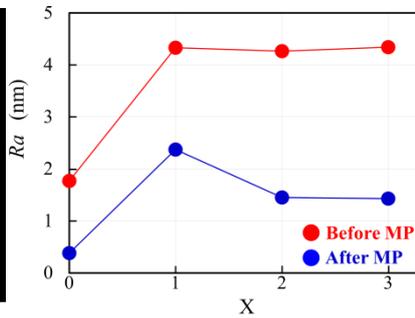


Fig. 4 Ra value variation of Ce_xY_{3-x}Fe₅O₁₂ (x=0~3) after MP process.

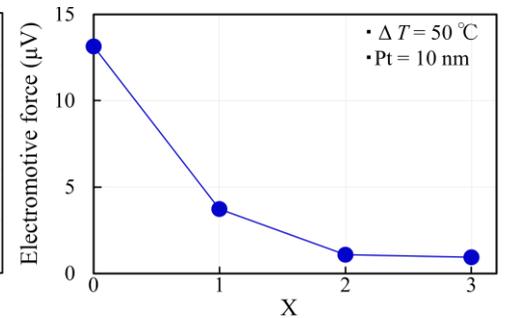


Fig. 5 Electromotive force of spin Seebeck devices using Ce_xY_{3-x}Fe₅O₁₂ (x=0~3).

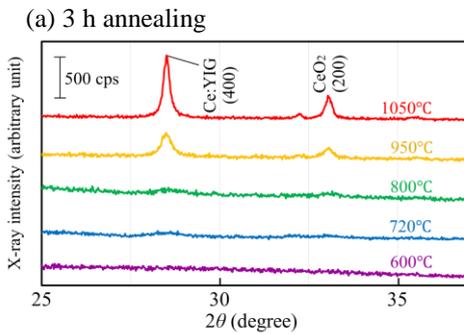


Fig. 6 XRD spectra of Ce₁Y₂Fe₅O₁₂ films annealed at various temperatures. (a) is for samples annealed for 3 h. (b) is for samples annealed for 14 h.

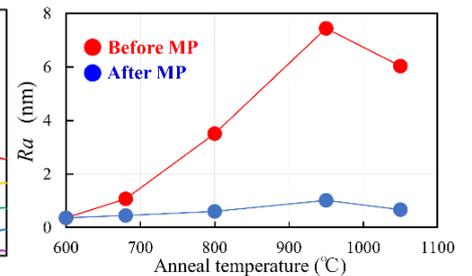
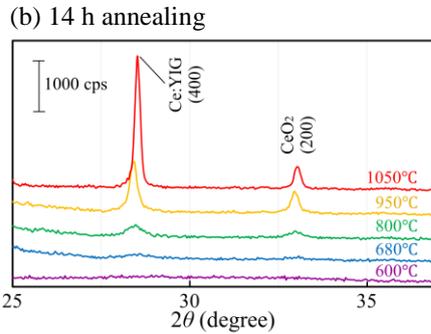


Fig. 7 Ra value variation of Ce₁Y₂Fe₅O₁₂ annealed for 14 h after MP process.

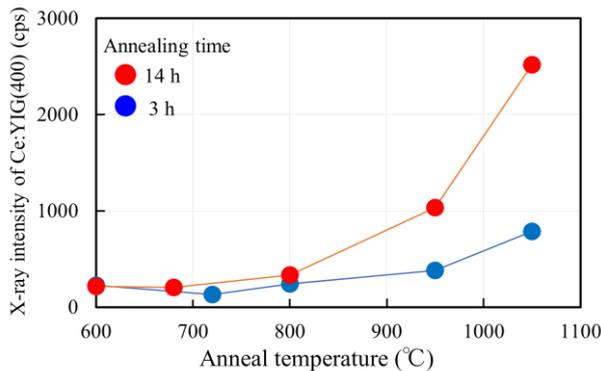


Fig. 8 Anneal temperature dependence of XRD peak intensity of Ce₁Y₂Fe₅O₁₂ (400).

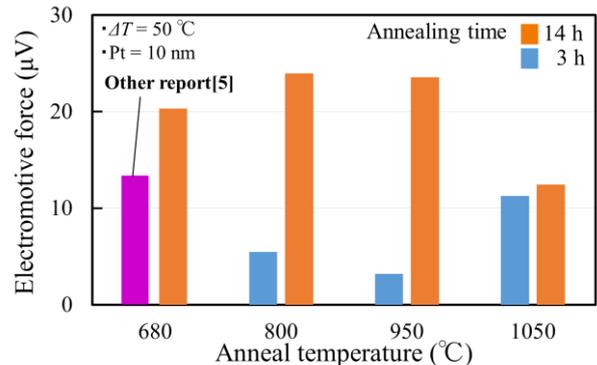


Fig. 9 Electromotive force of spin Seebeck devices using Ce₁Y₂Fe₅O₁₂ annealed for 14 h.