## **Original Research Paper**

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## Processing of Cerium Molybdates by Solid Phase Process and their Antiviral Activity

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## Abstract

Single-phase powders of  $\gamma$ -Ce<sub>2</sub>Mo<sub>3</sub>O<sub>13</sub>, Ce<sub>8</sub>Mo<sub>12</sub>O<sub>49</sub>, and Ce<sub>2</sub>Mo<sub>4</sub>O<sub>15</sub> were prepared using the solid-phase method. Then, the antiviral activity of these powders against the non-enveloped virus bacteriophage Q $\beta$  and the enveloped virus bacteriophage  $\Phi 6$  were evaluated. Moreover, H<sub>2</sub>O<sub>2</sub> assay and catalase inactivation test were performed. Regarding the antiviral activity of the prepared samples, the activity against bacteriophage  $\Phi 6$  was higher than the activity against bacteriophage Q $\beta$ . The antiviral activity order for bacteriophage  $\Phi 6$  was correlated with the amount of H<sub>2</sub>O<sub>2</sub> generated and the catalase inactivation rate. It depended also on the eluted Ce/Mo ratio. These results imply that the antiviral activity on bacteriophage Q $\beta$  is influenced by neutralization of the negative charge of capsid by rare earth ions and by adsorption of heteropolyacids to specific sites of the virus. Results suggest that the oxidation reaction of Ce (IV) or of the H<sub>2</sub>O<sub>2</sub>, and the adsorption of heteropolyacids formed by Ce and molybdate ions affect the antiviral activity against bacteriophage  $\Phi 6$ . Among the compounds examined this time,  $\gamma$ -Ce<sub>2</sub>Mo<sub>3</sub>O<sub>13</sub> exhibited the highest antiviral activity against bacteriophage  $\Phi 6$ .

Key-words: Mo, Ce, Antiviral, Solid phase, Virus

## 1. Introduction

The new coronavirus pandemic is continuing worldwide, and seeing the direction of convergence is still difficult. This viral pandemic has underscored the impossibility of halting economic and social activities completely, even under difficult circumstances. To continue certain economic and social activities until vaccines are widely used, technology and knowledge of "prevention" and "suppression of spread" of infection are as important as "development of vaccines and treatment methods". In recent years, antiviral materials have attracted great attention as one technology for that purpose.

Inorganic antibacterial and antiviral materials have a shorter history of development than organic ones, but they are less temperature-dependent, effective against various bacteria and viruses, and make it difficult for viruses to acquire resistance. Conventional inorganic antibacterial and antiviral materials include noble-metal-based materials (Ag, Cu, etc.<sup>1-3</sup>), photocatalysts (TiO<sub>2</sub>, etc.<sup>4,5</sup>), and others (ZnO, alkaline oxides, etc.<sup>6,7</sup>). Nevertheless, several difficulties exist such as cost, deterioration of antibacterial and antiviral activity over time, coloring, and a limited usage environment.

We recently prepared La<sub>2</sub>Mo<sub>2</sub>O<sub>9</sub> (LMO) using the complex polymerization method, which revealed that this rare earth molybdate complex oxide is an antibacterial and antiviral material that overcomes these difficulties<sup>8</sup>). Furthermore, by substituting La of this material with Ce, the antiviral activity against enveloped viruses was improved<sup>9)</sup>. Actually,  $\gamma$ -Ce<sub>2</sub>Mo<sub>3</sub>O<sub>13</sub>, which consists only of Ce and Mo, exhibited extremely high antiviral activity against enveloped viruses including the new coronavirus (SARS-CoV-2)<sup>10)</sup>. From results of these studies conducted to date, it is considered that the antiviral activity of these materials originates from ions or polyacid ions eluted in water. Although oxides of molybdenum exhibit a certain solubility in water, it is suppressed and the sustained release of ions becomes feasible by forming a complex oxide with La or  $Ce^{9}$ . The antiviral activity of  $\gamma$ - $Ce_2Mo_3O_{13}$  against SARS-CoV-2 surpassed that of LMO or MoO<sub>3</sub>, indicating the effectiveness of the combination of Ce and Mo.

Because both Ce and Mo are polyvalent metal elements, cerium molybdate has complex oxides with various element ratios<sup>11)</sup>. Therefore, unless appropriate conditions are set, a single phase powder cannot be obtained. As described in an earlier report<sup>10)</sup>, we prepared single-phase powders of  $\gamma$ -Ce<sub>2</sub>Mo<sub>3</sub>O<sub>13</sub> and Ce<sub>2</sub>Mo<sub>3</sub>O<sub>12</sub> respectively using hydrothermal method and complex polymerization method.

<sup>[</sup>Figures and illustrations] Figures and illustrations published in black and white in the journal can be seen in color at our public website [J-STAGE]. Please make use of it.