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Effects of cerium and tungsten substitution on antiviral and antibacterial properties of lanthanum molybdate

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1. Introduction

The history of inorganic antiviral materials is not as long as that of organic antiviral materials. However, studies of inorganic antiviral materials are increasing gradually because one such material can affect various viruses under a wide temperature range with only a small probability of resistance development by a virus. Various materials have been investigated. Antiviral properties have been reported for metals [1–3] (e.g. Ag and Cu), photocatalysts (e.g. TiO\textsubscript{2} [4,5]), and other materials (e.g. ZnO and CaO [6,7]). In fact, some of these materials have already been applied for practical use. Nevertheless, these materials entail several difficulties such as coloration or inactivation because of oxidation, and usage environment restrictions (requirement of light illumination, or alkalization). Development of new inorganic antiviral materials has been demanded to overcome these and other difficulties.

We specifically examined the hydrophobicity of rare-earth oxides [8–12] and antibacterial effects of molybdenum [13–16]. Then we developed the complex oxide La\textsubscript{2}Mo\textsubscript{2}O\textsubscript{9} (hereinafter LMO) using polymerizable complex method [17]. This material exhibited hydrophobicity and simultaneous antibacterial effects against gram-negative (Escherichia coli, E. coli) and gram-positive (Staphylococcus aureus, S. aureus) bacteria, and antibacterial effects against non-envelope (bacteriophage Q\textsubscript{β}, hereinafter denoted as Q\textsubscript{β}) and envelope (bacteriophage Φ\textsubscript{6}, hereinafter denoted as Φ\textsubscript{6}) viruses [18]. Moreover, both antibacterial and antiviral activities were found to be greater than two orders after 6 h in the dark. However, the material’s antiviral activity against Φ\textsubscript{6}, which is similar to influenza and COVID-19 viruses, was less than that against E. coli, S. aureus, or Q\textsubscript{β}.

Given this background, we tried to substitute different atoms into LMO to improve its antiviral activity against Φ\textsubscript{6} type viruses. We selected cerium (Ce) and tungsten (W) because CeO\textsubscript{2} exhibits antibacterial activity and because it is a readily available, low-cost rare-earth oxide [19–22]. Tungsten, located below Mo in the periodic table, forms a solid solution with Mo in a wide chemical composition range [23,24]. Our preliminary experiments conducted using these two elements revealed that single-phase powder was difficult to obtain for high Ce concentrations. As the W concentration increases, the antibacterial performance of the material is degraded. Therefore, for this study, we replaced 10% of La with Ce (La\textsubscript{1.8}Ce\textsubscript{0.2}Mo\textsubscript{2}O\textsubscript{9}, hereinafter denoted as LCMO) and 50% of Mo with W (La\textsubscript{2}(Mo, W)O\textsubscript{9}, hereinafter denoted as LMWO). These powders were prepared using the same polymerizable
complex method as that used for an earlier study [18]. Then we compared their performance with that of LMO.

2. Experiment procedure

2.1. Powder synthesis and characterization

As starting materials for this study, we used lanthanum (III) nitrate hexahydrate (La(NO₃)₃·6H₂O, 99.9%), hexa-ammonium heptamolybdate tetrahydrate ((NH₄)₆Mo₇O₂₄·4H₂O, 99.9%), ammonium tungstate pentahydrate (NH₄)₆W₁₂O₄₁·5H₂O, 85%), and cerium (III) nitrate hexahydrate (Ce(NO₃)₃·6H₂O, 99.9%), all from Fujifilm Wako Pure Chemical Corp., and all with no further purification.

For LMO, La(NO₃)₃·6H₂O (2.50 g) and (NH₄)₂MoO₄·2H₂O (1.02 g) were each dissolved into distilled water (10 ml). Then these solutions were mixed and set to Ca:Mo = 1:1. For LMWO, (NH₄)₂MoO₄·2H₂O (0.51 g) and (NH₄)₂W₁₂O₄₁·5H₂O (0.754 g) were each dissolved into distilled water (20 ml). After La(NO₃)₃·6H₂O (2.50 g) was dissolved into distilled water (10 ml), these solutions were mixed to set La:Ca:Mo = 1:0.5:0.5. For LCMO, La(NO₃)₃·6H₂O (2.25 g) and Ce(NO₃)₃·6H₂O (0.250 g) were each dissolved into distilled water (8 ml for La(NO₃)₃·6H₂O and 2 ml for Ce(NO₃)₃·6H₂O). In addition, (NH₄)₂MoO₄·2H₂O (1.02 g) was dissolved into distilled water (10 ml). These solutions were then mixed to set (La + Ce):Mo = (0.9 + 0.1):1.

Subsequently, 2.31 mol/l of citric acid (C₆H₈O₇, 99.5%; Fujifilm Wako Pure Chemical Corp.) aqueous solution was added to the mixed solution. The molar ratio of metal ion (La + Ce + Mo + W) to citric acid was 2:3. This solution was stirred in a water bath at 80 °C for 6 h to facilitate esterification. Then precursor gels were obtained. The obtained gels were dried at 200 °C in ambient air. The dried gels were milled for 10 min using a mortar and pestle. After calcination at 500 °C, 550 °C for La(NO 3)3·6H2O and 2 ml for Ce(NO 3)3·6H2O). In addition, (NH₄)₂MoO₄·2H₂O (1.02 g) was dissolved into distilled water (10 ml). These solutions were then mixed to set (La + Ce):Mo = (0.9 + 0.1):1.

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2.2. Antiviral and antibacterial activity measurements

2.2.1. Sample preparation

For this study, antiviral and antibacterial activity measurements were taken not only for prepared powders but also for La₂O₃ (99.99%; Fujifilm Wako Pure Chemical Corp.), CeO₂ (99.5%; Fujifilm Wako Pure Chemical Corp.), WO₃ (> 99%; Kanto Chemical Co. Inc., Tokyo, Japan), and MoO₃ (99.0%; Fujifilm Wako Pure Chemical Corp.). All these oxide reagents were washed in distilled water (powder:water = 5 g:500 ml) and were dried at 100 °C for 12 h before measurements. These powder samples were dispersed into ethanol (sample: ethanol = 1 mg:1 ml; C₆H₈O₇, 99.5%; Fujifilm Wako Pure Chemical Corp.). The suspension (0.15 ml) was loaded uniformly onto a glass substrate (25 mm × 25 mm) and was dried at 100 °C for 30 min. After the loading–drying cycle was repeated three times, the substrate was coated with the sample powder (0.45 mg total amount).

2.2.2. Dissolved ion contact method. As described also for an earlier study, we evaluated the ion concentration when dissolved into 1/500 NB. To elucidate the contribution of dissolved ions to the overall antiviral activity, we took antiviral activity measurements using the solution after filtering the powder samples. Each prepared powder was mixed with 1/500 NB by the same solid/liquid ratio as that used for the film adhesion method. Then the mixture was shaken at 100 times/min for 2 h. Each suspension was filtered to exclude the powder. The pH value of the filtrate was measured. In addition, the amount of the dissolved ion (La, Ce, W, and Mo) was measured using ICP-OES. The filtrate solution was used for antiviral activity measurements against Qβ and Φ6. The filtrate solution, the suspensions of Qβ or Φ6 (2.2 × 10⁷ PFU/ml), and the distilled water were mixed at a ratio of 8:1:1. This mixture was stirred and incubated at room temperature (ca. 25 °C) in the dark room for 0, 2, 4, or 6 h. After incubation, the reactions were stopped with SCDLP medium. The medium was diluted with 0.01 M PBS. Then the antiviral activity was evaluated using the same procedure as that used for the film adhesion method. The initial virus concentration before contact with the filtrate solution including dissolved ions was set as the 0 h value. The control data were obtained using 1/500NB instead of the filtrate solution. Therefore, the ratio of 1/500NB, the virus suspensions, and the distilled water was 8:1:1; then, the same measurements were conducted. Hereinafter, this method is described as the dissolved ion contact method.

2.2.3. Antibacterial activity measurement

Evaluation of the antibacterial activity was conducted according to a film adhesion method described for ISO 17094, with minor modifications. For this study, we used E. coli (NBRC 3972) and S. aureus (NBRC 12732). Each had been precultured on nutrient agar (NA; Nissui Materials Science & Engineering C 117 (2020) 111323
Pharmaceutical Co. Ltd., Tokyo, Japan) at 37 °C for 18 h and had been suspended in 1/500 NB. The concentrations of these bacteria were fixed to ca. 2.0 × 10^6 colony-forming units (CFU)/ml. A 50 μl (=10^5 CFU) bacteria suspension was pipetted onto a substrate loaded with the sample powder. After the substrate was covered with a transparent film to contact the bacteria suspension with the particles, it was incubated under a humid condition at room temperature (ca. 25 °C) in a dark room. After a certain period (0, 2, 4, and 6 h), the bacteria were harvested by shaking with 5 ml SCCLP for 2 min to halt the incubation. The bacteria in SCCLP were diluted with 0.01 M PBS. Each 1 ml of the diluted bacteria suspension was mixed in NA and was incubated at 37 °C for 48 h to produce bacterial colonies. The concentration of viable bacterial cells at each time point (N) was calculated by multiplying the number of colonies and the dilution ratio. The control cell concentrations were calculated using the same procedure as that used for a pristine glass substrate. The initial cell concentration for each sample was presented as the cell concentration at 0 h. The colony assay was conducted twice for each point.

2.2.4. Cytotoxicity test

Cytotoxicity confirmation was conducted using Madin–Darby canine kidney cells (MDCK cells, CCL-34) purchased from American Type Culture Collection (Manassas, VA, U.S.A.). Cells were maintained using modified Eagle’s medium containing 10% fetal bovine serum and were incubated at 35 °C under a humidified 5% CO₂ atmosphere in an incubator. The prepared sample glass was put in a Petri dish into which was poured 5 ml of PBS. The extraction solutions of sample powders were prepared by shaking 100 times/min for 2 h. The extraction solution (50 μl) and MDCK cell solution (4.0 × 10^5 cells/ml, 150 μl) were mixed and incubated at 35 °C under a humidified 5% CO₂ atmosphere in an incubator for 4 days. After incubation, an adenosine triphosphate detection reagent (Viral ToxGlo™ Assay; Promega Corp., U.S.A.) was added (100 μl). A spectrophotometer (infiniteM200; Tecan Group Ltd., Austria) was used to measure the emitted light intensity. The number of living cells was calculated. Control data were obtained using the same experimental procedure with a pristine glass plate used instead of a glass plate with a sample powder.

3. Results and discussion

3.1. Powder characterization

Fig. 1 presents XRD patterns of the prepared powders. The obtained powders were almost single phase; peaks in the patterns were identified as La₂Mo₂O₉ (card No. 28-0509). Fig. 2 portrays the corresponding SEM micrographs. The primary particle sizes of the powders were 50–100 nm. No specific microstructure such as micro domains was observed in these particles by TEM observation (see Fig. SI-1 in Supporting information). XRD patterns and SEM micrographs of oxide reagents are presented in Supporting information (Figs. SI-2 and 3). They were larger than the prepared powders. Although La₂O₃ was the mixture of La₂O₃ (card No. 5-0602, major phase) and La(OH)₃ (card No. 5-0585, minor phase), others were single-phase oxides. Their peaks were identified respectively as CeO₂ (card No. 34-0394), MoO₃ (card No. 5-0508), and WO₃ (card No. 32-1395).

Subsequent ICP analysis revealed that all prepared powder samples possess the chemical composition ratio as charged (La:Mo = 1:1 for LMO, La:Ce:Mo = 1.7:0.2:2.0 for LCMO, and La:Mo:W = 1.9:1.0:1.0 for LMWO, respectively). In the wide scan XPS spectra, only constituent elements and carbon were detected. Narrow scans were conducted on La3d, Ce3d, Mo3d, and W4f, respectively (see Fig. SI-4 in Supporting information). Surface compositions for the prepared powder samples obtained by XPS were La₂Mo₂O₉ (card No. 5-0602, major phase) and La(OH)₃ (card No. 5-0585, minor phase), others were single-phase oxides. Their peaks were identified respectively as CeO₂ (card No. 34-0394), MoO₃ (card No. 5-0508), and WO₃ (card No. 32-1395).

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The dissolved ion concentration in 1/500 NB solution, with specific surface areas of prepared powders and oxide reagents.

| pH | SSA [m²/g] | La [µmol/l] | Ce [µmol/l] | Mo [µmol/l] | W [µmol/l] |
|----|------------|-------------|-------------|-------------|------------|
| Initial | 7.80 | – | – | – | – |
| LMO | 5.26 | 6.8 | 168 | N.D. | 299 | N.D. |
| LCMO | 5.77 | 10.4 | 75 | 0.43 | 112 | N.D. |
| LMWO | 5.84 | 12.0 | 12 | N.D. | 16 | 1 |
| La₂O₃ | 6.60 | 0.6 | 2 | N.D. | N.D. | N.D. |
| MoO₃ | 3.35 | 1.7 | N.D. | N.D. | 3992 | N.D. |
| CeO₂ | 5.46 | 3.3 | N.D. | 4 | N.D. | N.D. |
| WO₃ | 4.87 | 4.8 | N.D. | N.D. | 87 | |

was identified as Ce(III):Ce(IV) = 56:44 by the deconvolution of Ce3d peaks [28,29].

Table 1 presents results of pH and dissolved ion concentrations from sample powders immersed in 1/500 NB, and presents the specific surface areas of samples. The surface area values were 7–12 m²/g for prepared powders. The bar graphs for pH and dissolved ion concentrations presented in Table 1 are displayed in Supporting information (Figs. SI-5 and SI-6). The initial pH value for 1/500 NB before ion dissolution from the powders was 7.8. Despite their small specific surface areas, the dissolved ion concentrations from MoO₃, CeO₂, and WO₃ were greater than those from prepared powders. The solubility of MoO₃ in water is 3.4 mmol/l at 28 °C [30]. Results obtained for La₂O₃, CeO₂, and WO₃ are quite low. They can indicate that the materials are almost insoluble [31,32]. The dissolved ion concentration found for La₂O₃ was the lowest among oxide reagents. Dissolved amounts of Ce, W, and Mo decreased because they formed complex oxides with La. This result implies that the dissolution of Ce, W, and Mo was suppressed, and that the sustained release of these elements is attributable to the formation of complex oxides with La because of its low solubility. The dissolved ion amount ratios between La and Mo are of almost equal order, which suggests that the dissolution of Mo induces La dissolution. An inverse trend was found between pH values and the Mo dissolution amount. As described in an earlier report [18], that result is expected to be attributable to the resultant increase of the hydronium ion concentration by Mo dissolution. The pH value order for oxide reagents was La₂O₃ > CeO₂ > WO₃ > MoO₃. By forming complex oxides with La, the pH values of prepared powders became higher than those of simple oxide reagents.

3.2. Antiviral activity

Fig. 3 presents results of antiviral activity against Qβ for prepared powders (Fig. 3(a) and (b)) and oxide reagents (Fig. 3(c) and (d)) obtained by film adhesion method (Fig. 3(a) and (c)) and dissolved ion contact method (Fig. 3(b) and (d)). The antiviral activity achieved using dissolved ion contact method was higher than that achieved using film adhesion method for LMO, LCMO, and LMWO. The trend found for LMO and LMWO was similar to that found for oxide reagents. Although little antiviral activity was found for CeO₂, La₂O₃ exhibited clear antiviral activity against Qβ. The antiviral activity order by film adhesion method was MoO₃ > WO₃, and La₂O₃ > CeO₂; that by dissolved ion contact method was La₂O₃, WO₃ > MoO₃ > CeO₂.

The contributions of dissolved ions, especially those of ions of La, W, and Mo, are large for antiviral activity against Qβ because the activity achieved using dissolved ion contact method was higher than that achieved using film adhesion method. Before this study, we conducted a preliminary study of LaVO₄, for which we replaced Mo by V in LMO. This material exhibited a small pH value (4.71) under the same experimental conditions. Because the dissolved amount of V is much greater than that of La, ion exchange between VO₄³⁻ and OH⁻ were expected for charge compensation. Nevertheless, this material exhibited little antiviral or antibacterial activity (see Table SI-1, Figs. SI-7 and SI-8 in Supporting information). Consequently, differences of antiviral activity cannot be attributed simply to the different pH values.

The dissolved W and Mo form polyacids in this pH range. They might form heteropolyacids with other ions because the solution contains various ions such as phosphorous. The polyacids have a negative charge in aqueous media and contribute to antiviral activity [33–36]. Judd et al. demonstrated that polyacids adsorbed at the cation site of lysine residue in the active site of reverse transcriptase by electrostatic interaction and demonstrated that they inactivate human immunodeficiency virus (HIV) [37]. Reportedly, lysine residue is necessary for bonding with proteolytic enzymes in neuraminidase, which contributes to desorption from the host cell [38]. These polyacids might contribute to inactivation by inhibiting virus desorption from the host cell. Although HIV is a virus with an envelope, lysine residue also exists on the surface of non-envelope viruses at the active site of a spike, which contributes to bonding with the host cell [39]. Actually, lysine residue exists in the structure of Qβ [40]. Consequently, these polyacids can be expected to contribute also to inactivation of non-envelope viruses such as Qβ.

Several reports have described adsorption or interaction between Mo or W ion (strictly speaking, poly type anions of molybdate (MoO₄²⁻) or tungstate (WO₄²⁻) ions) and lysine under different pH [41–43]. The adsorption amount of Mo and W polyacids to lysine depends on the lysine-metal ratio. The adsorption amount of W exceeds that of Mo when the ratio becomes higher than a certain value [43]. This ratio might engender antiviral activity difference between WO₃ and MoO₃ in dissolved ion contact method. Pérez et al. demonstrated that La(III) does not enter the host cell. It blocks Ca(II) entry induced by rotavirus (non-envelope type virus) infection [44]. Wengler et al. also demonstrated that lanthanide ion (such as La(III) and Ce(III)) treatment blocks the activity of the host cell to support the replication of flavivirus (envelope type virus) RNA. A similar effect of La(III) is expected to contribute to the inactivation of Qβ [45]. The slight antiviral activity shown by CeO₂ might be attributable to the major Ce ion valence (not III but IV).

Based on the relation between the dissolved ion amount and antiviral activity against Qβ, we can infer that the activity order of prepared powders depends on the dissolved ion amount, and especially on the amounts of La and Mo. The dissolved La(III) amount was LMO > LCMO > LMWO > La₂O₃, whereas antiviral activity by dissolved ion contact method was La₂O₃, WO₃ ≈ LMO > LCMO > LMWO ≫ CeO₂. The activity difference between WO₃ and LMWO is attributable to the dissolved W ion concentration difference. However, the activity order of LMO, LCMO, LMWO, and La₂O₃ does not simply follow the order of the dissolved La amount. Although the reason for this result remains unclear, one reasonable explanation is the interaction among La, W, and Mo ions. The antiviral effects of La ion against Qβ might weaken when W or Mo ions co-exist in the solution by formation of heteropolyanion; alternatively, they might weaken for some other reason.

Fig. 4 presents the results of antiviral activity against φ6 for prepared powders (Fig. 4(a) and (b)) and oxide reagents (Fig. 4(c) and (d)) by film adhesion method (Fig. 4(a) and (c)) and by dissolved ion contact method (Fig. 4(b) and (d)). The antiviral activity achieved by dissolved ion contact method was lower than that achieved by film adhesion method for LMO, LCMO, and LMWO. The antiviral activity order shown by film adhesion method was LCMO > LMWO > LMO. That by dissolved ion contact method was LCMO > LMO > LMWO. The activity order of oxide reagents was MoO₃ > WO₃ ≫ La₂O₃ > CeO₂. This order was the same for both methods. Although the antiviral activity of WO₃ by dissolved ion contact method is higher than that by film adhesion method, other oxide reagents exhibited similar antiviral activity from these two methods.

Dissolved Mo and W ion also contribute to the overall antiviral activity against φ6 to some degree, although their contributions are not...
as strong as that of Qβ. The antiviral activity by dissolved ion contact method was lower than that by film adhesion method for prepared powders. Therefore, we can infer that the effects of direct contact of the virus to the powder surface also exist for these powders. This trend differs from Qβ. The ion concentration is expected to be high around the powder surface. Therefore, this difference might be attributable to the difference of virus characteristics such as the resistance against dissolved ions. Resistance against La ion might also be related to this difference. The addition of Ce ion shows positive effects on antiviral activity against Φ6. In fact, LCMO exhibited the highest activity among the prepared powders, even though the dissolved ion amount is less than that for LMO. Because CeO₂ possesses little antiviral activity, some synergetic effect is expected between Ce and Mo ions.

To date, several investigations have been conducted to elucidate the antiviral activity of polyacids combined with rare-earth elements [46–48]. Liu et al. investigated antiviral activity against influenza viruses (envelope type viruses similar to Φ6) of rare-earth bortungstate heteropolyoxometalates. They demonstrated that heteropoly blues (a part of W in polyacid that is reduced) containing Ce exhibits the strongest inhibition activity against influenza virus. They inferred that this substance has a molecular size that is suitable to interfere and destroy the virus or virus’ chemical components [47]. Recently, Shiohara et al. and Kato et al. demonstrated that a combination of multi-valence elements such as Mn and Ni enhances the Mars – van Krevelen mechanism [49,50], which derives from the strong oxidation power of Ce [51,52]. Actually, when LCMO was washed with ethanol, the ratio of Ce(IV) decreased and that of Mo(V) increased (see Fig. SI-9 in Supporting information). Therefore, the oxidation power of Ce might also enhance antiviral activity against Φ6. Further investigations must be conducted to identify the dominant mechanism of this system. However, the effect of Ce is more remarkable on Φ6 than on Qβ. The combination of Ce ion and Mo-based polyacids is expected to be advantageous for similar viruses such as influenza and COVID-19.

For this study, the advantage of W for antiviral activity by film
adhesion method (LMWO > LMO) is remarkable. However, a different trend was obtained when dissolved ion contact method (LMO > LMWO) was used. The dissolved ion amount for LMWO is much smaller than that of LMO, probably because of the decrease of the atomic ratio of Mo with high solubility. Near the powder surface, the ion concentration increases. For that reason, the effect of direct contact of the virus to the surface becomes remarkable; LMWO exhibited higher antiviral activity than LMO by film adhesion method. Therefore, if the direct contact situation between powder and virus is designed effectively, then W addition might also be effective for increasing antiviral activity against Φ6.

Fig. 5 displays photographs of the change of plaque number for Φ6 that occurs when using film adhesion method. The number of plaques was decreased remarkably. Almost all plaques were eliminated by LCMO within 6 h.

3.3. Antibacterial activity

Fig. 6(a) and (b) presents results of antibacterial activity against E. coli for prepared powders (Fig. 6(a)) and for oxide reagents (Fig. 6(b)). The activity order for the prepared powders was LMO > LCMO ≈ LMWO. That for oxide reagents was MoO3 > La2O3 ≫ WO3, CeO2. Antibacterial activity was not obtained from WO3 or CeO2. These trends are almost identical to that found for E. coli, which suggests that substitution of Ce or W for the part of LMO is ineffective for antibacterial activity improvement. However, both LCMO and LMWO retain a certain degree of antibacterial activity (three order decrease in 6 h). Fig. 6 displays the cytotoxicity test results for LMO, LCMO, and LMWO. None of these powders possesses cytotoxicity. This result was the same as that obtained from our previous study [18]. It is noteworthy that various experiments can be used to assess the toxicity of the materials. Further various experiments are necessary to confirm the cytotoxicity of these materials.

The dissolved amount of ions from prepared powders is greater than that for oxidized silver (Ag2O, 0.86 μmol/l [59]), but less than that for MoO3. All these powders are white (LMO and LMWO) or light yellow (LCMO). The probability of spoilage design is low. Although MoO3 is effective for antiviral and antibacterial use in the initial stage, its activity is expected to deteriorate quickly because of its high solubility. Actually, La-based complex oxides such as LMO, LCMO, and LMWO are difficult to deactivate. For that reason, they retain their activity longer than MoO3, releasing Mo-polyacids slowly with higher amounts of minimal inhibitory concentration (MIC) value (0.17–1.7 μmol/l for E. coli [60]).

This study indicated LCMO as the most promising material with high antiviral activity against Φ6. We confirmed that this material possesses not only antiviral and antibacterial properties but also hydrophobicity similar to that of LMO (see Figs. SI-10, SI-11, and SI-12 in Supporting information). Because of its hydrophobicity, this complex oxide is expected to have good affinity with organic compounds. It is noteworthy that the method of measuring antiviral and antibacterial properties used for this study does not include the contribution of materials’ hydrophobicity. Several earlier studies demonstrated the benefits of hydrophobicity for antiviral and antibacterial performance [61–63]. Detailed investigations of the relations between hydrophobicity and antiviral or antibacterial performance of this material must be undertaken in future work.

Unlike LMO, LCMO has a UV-shielding property that originated in Ce [64–66] (Fig. 9). However, photocatalytic decomposition activity was not obtained from this material. Therefore, this material might be applicable to UV shielding coatings for windows or cosmetics.

4. Summary

For this study, we partially replaced La and Mo of La2Mo2O9 (LMO) by Ce or W. We then investigated their antiviral and antibacterial properties using Qβ, Φ6, E. coli, and S. aureus. The powdered sample was prepared using polymerizable complex method. The obtained
powders, which were almost single phase, exhibited both antiviral and antibacterial performance. The dissolved ions strongly affect antiviral activity against Qβ. A certain contribution of direct contact between the powder surface and virus was inferred, aside from the effects of dissolved ions for antiviral activity against Φ6. Results suggest that Mo and W form polyacids in the solution. Partial substitution of Ce for La improved the antiviral activity against Φ6. All prepared powders inactivated ALP enzyme proteins, suggesting that one mechanism accounts for their antiviral and antibacterial performance.

CRediT authorship contribution statement

Takumi Matsumoto: Investigation, Data curation, Formal analysis
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Takeshi Nagai: Methodology
Toshihiro Isobe: Investigation, Supervision
Sachiko Matsushita: Investigation
Hitoshi Ishiguro: Methodology, Data curation, Formal analysis

Akira Nakajima: Conceptualization, Project administration, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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