39-9 Synthesis and Photoluminescence Properties of Layered Oxides Intercalated with Rare-Earth Cations by Electrostatic Self-Assembly Methods

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Various rare-earth cations were intercalated into the interlayer of the exfoliated $H_{0.76}Ti_{1.81}\square_{0.19}O_4 \cdot nH_2O$ ($Ti_{1.81}O_4$-nanosheet) and $H_2Ti_4O_9 \cdot nH_2O$ (Ti_4O_9-nanosheet) by the electrostatic self-assembly deposition (ESD), where a nanosheet colloidal solution was mixed with an aqueous solution of a rare-earth cation solution at a certain pH value. Rare-earth cations in the interlayer exist in their 7-10 coordinated aqua ion forms according to XRD and thermal analysis data. The interlayer distance of 6-7 Å is large enough to accommodate a rare-earth aqua ion. Heat treatment by 300^{o}C resulted in the shrinkage of the interlayer distance to lower values corresponding to the radius of bare rare-earth cations. Intercalation was also succeeded by the layer-by-layer self-assembly (LBL) method. Sequential deposition of nanosheets and lanthanide cations on PEI-treated substrate resulted in the formation of thin layered oxide films intercalated with rare-earth cations. Eu^{3+} in interlayer of $Ti_{1.81}O_4$-nanosheets and Tb^{3+} in interlayer of Ti_4O_9-nanosheets showed strong red and green luminescence at room temperature, respectively. It was found that the luminescence is mainly contributed by the band gap excitation in the host nanosheet layer. Heat treatment and humidity controlled experiments revealed the importance of the surrounding water molecules for the high luminescence. The films treated with high humidity showed strong luminescence while heat-treated or dehumidified films showed relatively very weak luminescence. The mechanism was suggested that electrons and holes migrating in the host layer move simultaneously through the surrounding water molecules to interlayer rare-earth cations to yield emission rather than giving radiationless quenching via energy transfer to OH vibrating molecules.

[2006 the 10th International Symposium on Advanced Display Materials and Devices Abstract(ADMD 2006), 2006.6]