Project No. 02319/ Grant No. JPMXP0723833151

Report form of Joint Research Project at ZAIKEN (FY2023)

Title of Project	Development of efficient phosphor materials with transition element dopings		
Priority Area	I-B(Prolongation of service life & Structures), I-C(Prolongation of service life & Properties), III-B(Energy saving & Structures), III-C(Energy saving & Properties)		
New proposal			
Name of Main Applicant	Farhod Rahimi		
Institution	National Academy of Sciences, Republic of Tajikistan	**title**	President (Professor)

Aim of the research project

Phosphor materials have gained extensive attentions because of their important applications in our life. Many of the current phosphor materials are prepared by doping of dilute amount of rare-earth or transition metal ions, which act as an emission center, in matrix materials. Among such phosphors, rare-earth doped compounds, such as nitrides, oxides and fluorides, have been widely used, since such materials have good luminescence properties. However, due to the limitation of the rare-earth elements in the earth and the cost for the sources of rare-earth elements, rare-earth free phosphor materials have been strongly demanding, and therefore such materials have been extensively investigated these years. In these phosphor materials, red emission phosphor is especially important, since the conventional white light emitting diode (w-LED) made with combination of blue LED and yellow phosphors has low color rendering property and high color temperature. In the current project, we propose development of efficient red-emitting phosphors with transition element dopings. By collaborating with Professor Yamamoto, advanced analyses such as X-ray diffraction, SEM-EDX, AES, XPS, UV-Vis, PL, ESR will be performed at Waseda. From the results of the current project, we will obtain new materials with longer service life contributing to the energy saving.

Contents and results of the research

The first principles calculations for various kinds of oxides and fluorides doped with Mn^{4+} ions have been performed and compared with the experimental results in the collaborating project with Prof. Yamamoto. Some of the results have been already reported as joint papers in international journals [1-3]. In the current project, the earlier research has been expanded to various kinds of matrix materials and perform a series of analyses, i.e., X-ray diffraction for structure analysis, UV-Vis for electronic structure analysis, photoluminescence (PL) analysis, to search the efficient rare-earth free Mn-doped red-emitting phosphors.
Owing to the collaborations with Prof. Yamamoto, samples were provided by his group, and cutting-edge analyses have been carried out mainly at ZAIKEN. The first principles calculations within a density functional theory have been also conducted for the analysis of the local structure around doped 3d transition ions such as Cr^{3+}, Mn^{4+} and Ni^{2+}.

Followings are the items we conducted in this fiscal year.

1) Samples
Samples were prepared with the conventional solid-state reaction method by the group of Prof. Yamamoto.
2) X-ray diffraction at ZAIKEN
Phase purity and crystal structures of the sample powders provided by Prof. Yamamoto have been examined by the X-ray diffraction technique. Rietveld analysis will be also carried out.
2) UV-Vis analysis at ZAIKEN
The electronic structures of the sample powders were examined by the diffuse reflectance spectra using integration sphere equipped with the UV-Vis. In these measurements, change in band-gap and absorption states created by the Mn doping were determined.
3) Photoluminescence analysis at Yamamoto Lab.
Photoluminescence spectra were measured in excitation and emission modes, which enable us to know the efficiency and emission wavelength, i.e., color, of the sample powders.
4) First-principles calculations at Yamamoto Lab.
We have performed the first-principles calculations with vasp code, in which both meta-GGA and hybrid functionals were employed for the electron-electron correlation functional. Dr. Dilshod Nematov in our group visited Waseda University and partially performed the above 1)-4), during his stay at ZAIKEN. Dr. Amondulloi Burkhonzoda performed first-principles calculations remotely on the server settled in ZAIKEN. We plan to summarize the results of the current joint research, which will be submitted to the international journal.

References
[1] M. Subhoni, U. Zafari, Ch-G. Ma, A. M. Srivastava, W. W. Beers, W. Cohen, M. G. Brik, M. Piasecki and T. Yamamoto, *Materials,15*, 613, 2022
[2] M. Subhoni, U. Zafari, A. M. Srivastava, W. W. Beers, W. Cohen, M. G. Brik and T. Yamamoto, *Opt. Mater.* 115,110986, 2021.
[3] M. Sagayama, U. Zafari, M. Subhoni, W. W. Beers, W. E. Cohen, M. Brik, T. Yamamoto, *ECS J. Solid State Sci. Technol*. 10, 076004, 2021.

Outputs of the project (publications, presentations, patents)

We have successfully published the following two papers in the impacted international journals.

1) Z. Umar, M. S. Kurboniyon, O. Khyzhun, T. Yamamoto, C.-G. Ma, M. G. Brik, M. Piasecki

First-Principles Calculations of the Electronic Structure and Mechanical Properties of Non-Doped and Cr^{3+}-Doped K_2LiAlF_6 Under Pressure, J. Lumin. 266 (2024) 120278

2) Z. Umar, O. Khyzhun, T. Yamamoto, P. Bragiel

Ab initio calculations of two-site occupation for substituting ions in $MgTa_2O_6$:Cr^{3+} phosphor: The geometric and electronic structures and optical transition energies, Opt. Mater. 140 (2024) 115057