

**京都大学教育研究振興財団助成事業
成 果 報 告 書**

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会 長 藤 洋 作 様

所属部局・研究科 工学部・材料化学専攻

職 名・学 年 博士後期課程・二回生

氏 名 GAO YUAN

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成 果 の 概 要	タイトルは「成果の概要／報告者名」として、A4版2000字程度・和文で作成し、添付して下さい。「成果の概要」以外に添付する資料 <input checked="" type="checkbox"/> 無 <input type="checkbox"/> 有()	
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当財団の助成について	自身の研究発表以外にも、世界的に著名な研究者の講演や、世界最大規模の会議だからこそ幅広いテーマの研究発表を聴講することができ、知見や興味を広げることができた。この貴重な経験は、今後の研究生活に十分に活かしていきたい。今回の国際会議への参加は私にとって非常に有意義なものとなりました。このような機会と多大なるご支援をいただきました貴財団に深くお礼申し上げます。	

成果の概要

GAO YUAN

1. 国際会議の概要

The *35th International Japan-Korea Seminar on Ceramics* was held in Gangneung, Korea during November 21st to 24th, 2018. In its 35-year history, this is the first visit of the Japan-Korea Seminar on Ceramics to the city of Gangneung in Korea. Gangneung City is a beautiful city located in the northern part of the east coast of Korea, about 300km from the Seoul, and asset of winter olympic game held in 2018. This seminar is the one of most famous in sector of ceramics between Japan and Korea. Japan and Korea hosted the annual meeting alternatively to promote the active communication of researchers and students in terms of ceramic science & technology also human resources. This conference covers broadband subject, such us fuel cell, batteries, secondary cells, solar cell, thermoelectrics, electronic ceramics, magnetic ceramics, piezoelectrics, sensor materials, structure and 3D printing, including power process, soft material, bio-applications, basic science of ceramics, glasses, nanoceramics, thin films, coatings, industry ceramics and so on. During this event, there are 146 papers were scheduled, including 33 invited, 54 oral presentations and 59 poster presentations made by many of the world's most prominent researchers from academia and industry.



Figure 1 opening ceremony

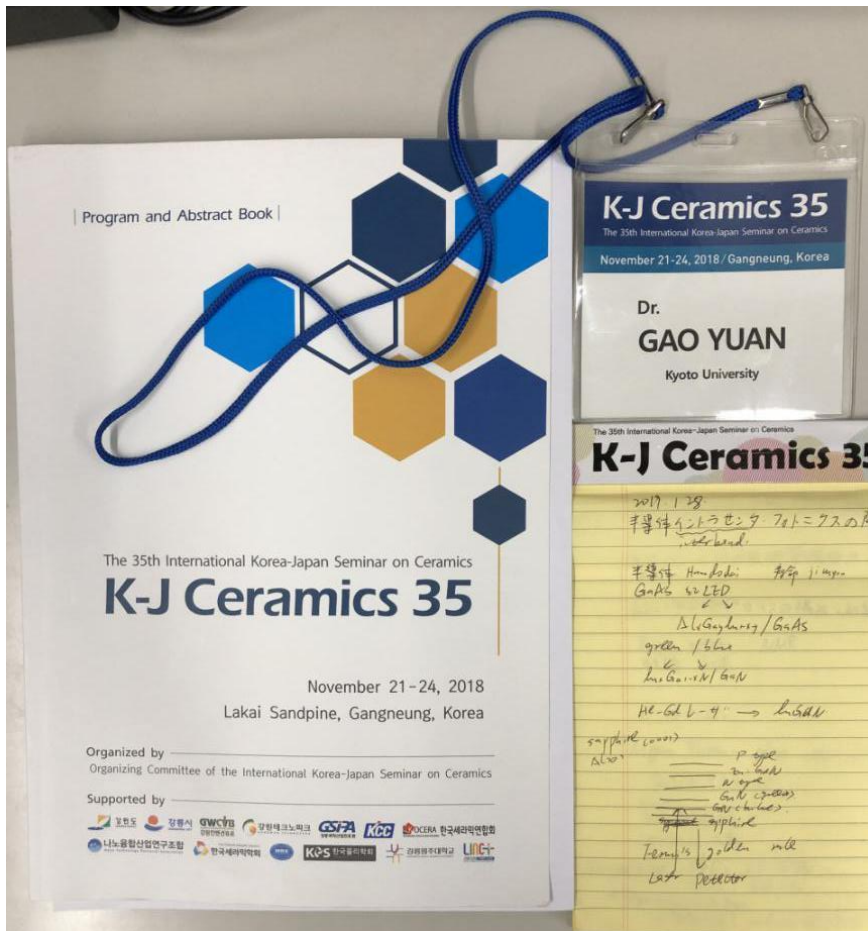


Figure 2 Documents

2. 研究テーマと討論内容

In this meeting, my talk presentation was performed in the symposium G-01, which is about nanoceramics general sessions, on November 23 at 13:00-13:15. I presented my research with the title as "Achieving Spatially- Selective Dopant Distribution in Glass Ceramics for Warm Light Emitting Diodes". As well known, white light-emitting diodes (WLEDs) are used extensively in backlights, flashlights and handlights as replacements for conventional incandescent and fluorescent lamps because of their high energy efficiency, durability, reliability, and capacity for use in products with various sizes and more eco-friendly components. Phosphors have been widely investigated for use in phosphor-converted WLEDs. The most widely used WLED combines the yellow phosphor $Y_3Al_5O_{12}:Ce^{3+}$ (YAG) with a blue InGaN LED chip. However, this LED presents a low color rendering index (CRI) ($R_a < 70$) and high correlated color temperature (CCT > 6000 K) because of the absence of red emission in its spectrum. In this study, we have fabricated Eu-doped oxy-fluoride glass-ceramics containing ~~two kinds of crystals $Na_5Gd_9F_{32}$ and $NaAlSiO_4$ nano- and microcrystals~~ by melt-quenching and post-annealing for warm WLEDs. ~~Annealing above $640^\circ C$ lead to the two crystalline phases precipitated in the glass matrix (Fig. 3(a)), and the glass-ceramics exhibited green broad emission at 540 nm and sharp red emission at 610 nm. The analysis including STEM EDX, SEM CL, and confocal PL~~

microscopy revealed that the emission came from the selective distribution of Eu^{3+} and Eu^{2+} in $\text{Na}_5\text{Gd}_9\text{F}_{32}$ and NaAlSiO_4 phases, respectively. The glass ceramics excited with InGaN UV chip ($\lambda_{\text{ex}}=395\text{nm}$) showed warm white emission (Fig. 3(b)) with CCT of less than 3000 K and CRI of over 70, while photoluminescence quantum yield was over 35%. Furthermore, we examined thermal quenching of photoluminescence at 573K; the photoluminescence integral intensity is still about 58% of that measured at room temperature. We believe that the glass-ceramics we found are applicable to high power W-LEDs and displays.

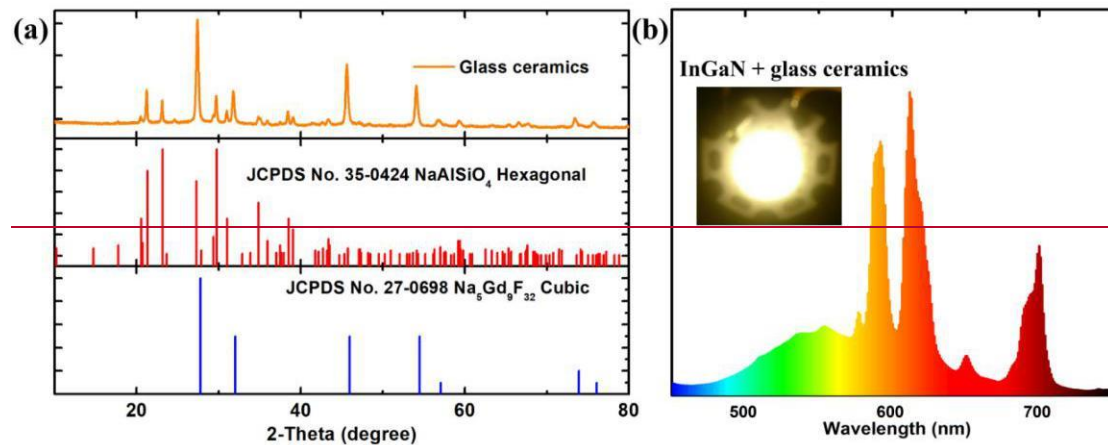


Figure 3 (a) X ray diffraction pattern of glass ceramics. (b) Photoluminescence spectrum excited with an InGaN UV chip.

I made my 10 minute-oral presentation, with 5 minutes questions-and-answers time. Approximately 20 people attended my talk. I received a couple of questions. Q#1: You were able to control the color of your glass ceramics sample under 393 nm UV source excitation from pink region to yellowish region, does it mean that the photoluminescence color can be adjusted between two kinds of emission spectra?", "A#1: The photoluminescence color changed dramatically with the formation of NaAlSiO_4 -containing Eu^{2+} , especially, with the heat treatment temperature increased from 620 to 640 degree C, which led to the precipitation of second phase of NaAlSiO_4 microcrystals, where Eu^{2+} primarily selectively distributed, there is a close relationship with the formation of NaAlSiO_4 second phase and the shift of photoluminescence color. So, in this case, it would be possible in principle for someone to control the photoluminescence color by changing the concentration of NaAlSiO_4 . However, under high temperature, we have difficulty in precisely controlling the phase formation process volume by changing the concentration of NaAlSiO_4 due to the fact that the rate of crystallization is very fast." "Q#2: Since the glass ceramics is opaque, how can it be used in illumination for WLEDs?", "A#2: Actually, the opaque glass ceramics was good for this purpose because it absorbs more UV excitation, and converts it to white light more so than that of transparent one. Moreover, UV light is harmful for human eyes". A couple of people got interested in my talk and we were able to make meaningful discussions with each other in our research fields, after the talk.

3. 国際会議に出席した成果（コミュニケーション・国際交流・感想）

The international Japan-Korea annual seminar on ceramics is a well-organized and deeply impressive conference, where I truly learned something new and broadened my horizons. In this meeting, there are presentations in various fields as well as my session, participating in announcements and demonstrations in different research fields has become a big stimulus for themselves. Also, after both oral and poster presentations, there was opportunity for discussion with researchers who came from in the same and different research fields, and it was very meaningful to obtain knowledge about future research activities. This is my second times to attend Japan-Korea seminar in ceramics, I attend this seminar at Hamamatsu, Japan in November, 2017. In my first time to attend this conference in the 34th Japan-Korea seminar in ceramics, the communication with researchers from all over the world is greatly inspired and unforgettable. Attending the international conference was a great experience for me and I was able to gain an incredible experience to meet the atmosphere of the international conference and people from oversea researchers. I answered the question I received as much as I could.



Figure 34 Banquet of Conference.

In this conference, During and after my presentation, I received some comments and questions, which are ~~our~~ important for our future work. Besides learning some new knowledge about optical measurement and manufacture, closely related my research field, I learned many technologies in fabrication method, temperature measurement and analysis ways et al. It was very impressive. At first day (22th November) of my travel, I listened invited speakers talk at plenary session. The speakers from Korea University, Shizuoka University, Zhejiang University et al, look very powerful and

those people gave me vitality to do my research more structural, more beneficial and also more goal-seeking. Especially, Professor Jong-Heun Lee who come from Korea University, tasking title is "Highly Sensitive, Selective, and Robust Oxide Semiconductor Gas Sensors: New Opportunities and challenges", which include the use of hollow and hierarchical nanostructures with high gas accessibility, microspheres with tri-modal porosity, p-type oxide semiconductors with high catalytic activity, yolk-shell micro-reactors to promote the reforming of inactive gases to more reactive forms, bilayer sensing film consisting of nano-scale catalytic overlayer, and hetero-nanostructures as sensing materials or platforms. I think all the attendees enjoyed this keynote speech and were touched by his rigorous attitude. At the same day, I also enjoyed poster session, including these title of "Synthesis of Mn^{4+} doped red phosphor by using atmosphere furnace", "Novel Inorganic Pigment with Ce^{3+} as a Coloring Source", "thick-film phosphor-in-glass with Nd^{3+} -ion doped phosphate glass for white LED with wide color gamut" and "Up-conversion studies on $\text{Er}^{3+}/\text{Yb}^{3+}$ co-doped fluorophosphate glasses" and so on. There are some poster presentation was interested and deep impressive in my research, such as Masashi MAEDA gave a poster title with "Synthesis of Mn^{4+} doped red phosphor by using atmosphere", in such work, he clarified the mechanism of high emission intensity, using various microscale to nanoscale characterization techniques: X-ray diffraction, a scanning electron microscope, and X-ray absorption fine structure spectroscopy. The emission intensity was closely related to the ratio of Mn^{4+} and structure of the phosphor. I learn various kind of measurement method and its mechanism, and I received information that next international Japan-Korea seminar in ceramics will be held in Tottor-shi of Japan in 2019 with further deeply communication, and sometimes we can use ing both English and Japanese. Moreover, the title is "thick-film phosphor-in-glass with Nd^{3+} -ion doped phosphate glass for white LED with wide color gamut" made by Yoon Hee NAM, a female student who come from Kongju University of Republic of Korea. In her study, a glass containing Nd^{3+} -ion and fabricated a phosphor-in-glass thick film on a glass substrate which dose not require further processes were developed Strong absorption at 580 nm due to Nd^{3+} -ion was introduced to adjust spectral shape of the green and red phosphor. Thick-film phosphor-in-glass with 50 μm in thickness were obtained via simple screen printing process. The Nd^{3+} -ion doped glass frit and $\text{Lu}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$ as green phosphor, and $\text{CaAlSiN}_3:\text{Eu}^{3+}$ as a red phosphor were mixed to obtain white light varying their contents. Doping concentration of Nd^{3+} -ion within the glass matrix was also optimized for color gamut improvement. Various optical properties were measured after mounting on top of the blue LED by an integrating sphere such as color coordinates, correlated color temperature, color rendering index and color gamut. I also enjoyed talk with other student for their own country culture and exchange e-mail of each other. In the second day, I present my research. Before my talk, I felt kind of fear that someone could not understand my research, if I received unfamiliar question that I couldn't give answer and so on. But after that, I understood that there was no need to feel heartbeat, because everyone was trying to exchange knowledge and learn new things. In addition, I enjoy talking with many top students of the field of inorganic ceramics. Many provided and inspired me to their

research with novel methods and materials. This was my first international conference on overseas in my doctor course. I was amazed by the profound discussions and sharing of knowledge of researchers during presentations to improve each research.



Figure 45 Picture with Korea student.

Finally, let me take this opportunity to express my deep gratitude for the support and grant of the international conference attendance from The Kyoto University education-research-foundation. I am honored and thankful to have been chosen for this financial support. Thus, I would like to express my seiesincerenee gratitude to my supervisor Prof. Shunsuke MURAI and Prof. Katsuhisa TANAKA for their help and guidance.



Figure 56 The Mascots of winter Olympic in 2018.