Solid conversion behaviors of Indium oxide gel comprising of hybrid clusters with thermal- and/or ultraviolet-treatments for low temperature processing

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This study reports on the investigation of the thermal and UV-irradiation solidifications of In-O cluster gel which was made from an In-acac solution using PrA as a solvent.

It is found that the thermal solidification process was strictly defined by the temperature. Thus, the gel state is maintained from 100 °C to 270 °C with constant desorption of the residual solvent, and then the organic ligand starts to decompose at 270 °C generating strong exothermic reaction to convert the gel to an In-O solid. It was confirmed that the well-defined structure of the cluster gel enables us to have a predictable solidification in the thermal treatment. More precisely, it was possible to see what happens in each temperature stage from solution to solid can be strictly defined. It was noticed that even the amount of the residual solvent in gel was a function of temperature. In the case of thermal pyrolysis, however, the solidification did not fully complete even by annealing at 500 °C, but some amount of gel elements remained in In-O solid. That was proofed quantitatively by the chemical composition analysis, which showed that the remaining carbon was 11%. Consequently, although the obtained semiconductor film by the thermal annealing at 500 °C had a moderate mobility, it contained a lot of defects.

As for the UV-irradiated solidification, it was demonstrated that the decomposing ability of organics was high enough to solidify gel even at room temperature. However, since most UV energy has to be consumed to remove residual solvent in UV treatment at RT, In-O cores in a gel did not grow, but remained in small-sized ordered clusters whose volume was twice as that of the original In-O core. In the case of the 200 °C-UV treatment, the most UV energy can be effectively used to decompose ligand molecules, because heat promptly removes the residual solvent in an early stage. As a result, the UV energy causes necking reaction of In-O cores to ensure the growth of fine crystals. Organics were completely carbonized during UV irradiation as strongly suggested by FT-IR analysis, Vth value of TFT, and TDS analysis. The results provided by these analyses showed that carbon atoms terminated defects of the film. In conclusion, we showed that In-O cluster gel can be converted into a good semiconductor film even at temperature as low as 200 °C by using a combined method of UV irradiation and thermal treatments.

In addition, I applied UV irradiation technique to eco-friendly pattering and low temperature processing for fabrication of oxide materials. UV irradiation and re-dissolving (UV-RD) pattering is one of the patterning method for oxide gel material which was developed in my master's study. UV-RD patterning is a patterning method using the change of re-dissolving ability by UV irradiation to the oxide gels without a vacuum system and photo-resist material. In this study, I applied the UV-RD patterning to some hybrid-cluster oxide gel materials; LaRuO, LaZrO, InGaO, and InSnO. After patterning of each materials, I applied the UV-RD patterning to all solution processed TFTs fabrication. Patterning of each materials was succeeded and all solution-processed TFTs were successfully fabricated by UV-RD patterning. Moreover, UV irradiation technique was applied to low temperature processing of oxide materials. By combination of 200 °C-UV treatment and hybrid cluster gels, the solution-processed oxide TFTs were successfully fabricated and operated at the process temperature below 200 °C on glass substrate. The characteristics of fabricated TFTs were a field-effect mobility of 0.5 $cm^2V^{-1}s^{-1}$. Using the TFTs, an electrophoretic display was successfully fabricated and operated. Finally, the solution-processed oxide TFTs were demonstrated on the flexible substrate.

In this study, the solidification process of hybrid-cluster InO gel by thermal treatment and UV treatment was investigate by various analytical methods in the first part. In the second part, the applications of UV treatment were introduced. As described above, combination of UV irradiation and hybrid cluster gels enables low temperature processing and eco-friendly of oxide material. By improving these to the stage where they can be put to practical use, it can greatly contribute to the realization of a sustainable society.

Key words; solution process, oxide semiconductor, hybrid cluster gel, ultraviolet irradiation, thin-film transistor