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題目(和文)	近赤外光応答性アップコンバージョンナノ粒子／アナターゼ型TiO2複合エアロゲルに関する研究
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著者(和文)	LiFu-Chih
Author(English)	Fu-Chih Li
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論文の要約
Thesis Outline

論文題目:

Study on NIR-Responsive Upconversion Nanoparticles/Anatase TiO_2 Composite Aerogel
（近赤外光応答性アップコンバージョンナノ粒子／アナターゼ型TiO_2複合エアロゲルに関する研究）

Environment contamination has been a serious problem in the world. The photocatalyst which utilizes one of renewable solar energy to decompose organic pollutions is considered as a solution for solving the environment pollution problem. Hence, I design a UCNPs/TiO_2 composite aerogel with high specific surface areas and wider absorption spectrum under solar irradiation as photocatalyst. The main aim of the present study is establishing a synthesis process for combining TiO_2 aerogel of 3-dimensional interconnected network structure with UCNPs which are able to convert the light from low energy NIR region to high energy UV region. The UCNPs/TiO_2 composite aerogel is synthesized by a sol-gel process followed by a supercritical ethanol drying process, and this novel UCNPs/TiO_2 composite aerogel is first reported in the present study.

Although TiO_2 has been reported for few decades, the effective synthesis process for synthesizing TiO_2 composite with high specific surface area and extending absorption spectrum has not drawn much concerned. The previous synthesis process for TiO_2 aerogel was not suitable for synthesizing composite due to strong acid and base as catalyst and required post heat treatment, which generally leads to the decrease of specific surface area. Besides, the $NaYF_4$:Yb,Tm@TiO_2 core shell structure has been reported and $NaYF_4$:Yb,Tm NPs could convert the NIR light to UV region for TiO_2. However, only extending light absorption spectrum of TiO_2 was discussed. The specific surface area which is one of the essential factors for photocatalytic activity was not investigated. Hence, as originality in the present study, TiO_2 aerogel was synthesized with high specific surface area by altering the temperature of sol-gel process to control the condensation rate of TiO_2 network structure formation. Additionally, a synthesis process for combining TiO_2 aerogel of 3-dimensional interconnected network structure with UCNPs which are able to convert the light from low energy NIR region to high energy UV region was established.

The optimum synthesis process to synthesize the anatase TiO_2 aerogel with high specific surface area using a green and easy chemical process is investigated for further fabrication of UCNPs/TiO_2 composite aerogel and described in Chapter 2. In Chapter 3, the synthesis process of UCNPs/TiO_2 composite aerogel with converting the NIR light to visible and UV regions was successfully established. UCNPs of $NaYF_4$:Yb,Ho which can convert the NIR light to visible region are doped into TiO_2 aerogel structure to synthesize NIR-responsive UCNPs/TiO_2 composite aerogel. Besides, in the literature, the $NaYF_4$:Yb,Tm NPs were proved to successfully convert light from NIR region to UV region. The $NaYF_4$:Yb,Tm/TiO_2 composite aerogel with converting the NIR light to UV region is synthesized and characteristics are investigated. The upconversion luminescence capability of $NaYF_4$:Yb,Tm NPs that exist in the TiO_2 aerogel matrix structure is proved to be able to convert NIR light to UV region after the synthesis process of the composite aerogel. The photocatalytic activities of pure TiO_2 aerogel under UV light irradiation, and $NaYF_4$:Yb,Tm/TiO_2 composite aerogel under UV and NIR light irradiation were discussed in Chapter 4.

In the present study, I provide new insights for designing and synthesizing new type of photocatalyst and this way is favor to enhance phtocatalytic activity of TiO_2 photocatalyst. The results obtained in the present study are summarized, and general conclusions are described in Chapter 5.

In this dissertation, anatase TiO_2 aerogel with high specific surface area is successfully synthesized by altering the temperature of sol-gel process to control the condensation rate of TiO_2 network structure formation. In addition, synthesis process of NIR responsive UCNPs/TiO_2 composite aerogel which is based on synthesis of TiO_2 aerogel is established. The characteristics and synthesis conditions of UCNPs/TiO_2 composite aerogel also are discussed. The main conclusions of this study are summarized in the following points.

1. The anatase phase of TiO_2 aerogel with mesoporous 3-dimensional interconnected network structure and high specific surface area is successfully synthesized by sol-gel process followed by supercritical ethanol drying process. The supercritical ethanol drying process is able to not only remove solvent and unreactants from gel but also transform the amorphous phase to anatase phase of TiO_2 aerogel.
2. Altering the reaction temperature during sol-gel process is able to control condensation rate and the condensation rate has effects on the formation of TiO_2 network structure. At lower temperature, the condensation rate is slower and the TiO_2 solution needs more gelating time to form TiO_2 gel so the TiO_2 network structure could be formed completer. Hence, this TiO_2 gel is easier to maintain the TiO_2 network structure during the supercritical ethanol drying process. In addition, at higher temperature, the condensation rate is faster and the TiO_2 solution only needs 3 h to form the TiO_2 gel; the network structure of TiO_2 aerogel is hard to maintain during supercritical ethanol drying process if the structure is not generating well, leading to specific surface area decreased.
3. The synthesis process for synthesizing UCNPs/TiO_2 composite aerogel was successfully established. The crystal structure, crystal size and morphology of UCNPs can be maintained after the UCNPs doping into TiO_2 aerogel structure.

The UCNPs existing in the TiO_2 matrix were proved to transfer the NIR light to the visible region under 980 nm excitation by photoluminescence analysis.

4. In terms of synthesis of UCNPs/TiO_2 composite aerogel, altering the reaction temperature during sol-gel process is able to avoid precipitation of UCNPs before the UCNPs/TiO_2 composite gel formed. This precipitated situation had an effect on the formation of TiO_2 aerogel structure and it hindered the formation of 3-dimensional interconnected network structure of TiO_2 aerogel.
5. The $NaYF_4$:Yb,Ho NPs that exist in TiO_2 aerogel structure can convert NIR light to visible region and the $NaYF_4$:Yb,Tm NPs which exist in TiO_2 aerogel structure can convert NIR light to visible and UV regions.
6. The more UCNPs exists in the TiO_2 aerogel matrix, the more NIR light could be absorbed by the composite aerogel and transferred to the UV region. The larger UCNPs doping into the TiO_2 aerogel, it could generate extra space between TiO_2 crystal structure and UCNPs so that the specific surface area and average of pore size are increased.
7. The high specific surface area TiO_2 aerogel with more available sites for capturing useable light and more available pores for chemical reaction exhibited high photocatalytic performance.
8. The anatase TiO_2 aerogel still maintained the photocatalytic ability after the $NaYF_4$:Yb,Tm NPs doped into the structure under UV light irradiation. The TiO_2 aerogel can absorb UV light which transferred from $NaYF_4$:Yb,Tm NPs to degrade MB under NIR light irradiation.