Impurity-governed lighting of rare-earth doped HfSi_xO_yN_z thin films grown by reactive magnetron sputtering

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Hafnia-based nanocomposites being doped with rare-earths show promising properties for optical applications due to high refractive index, high optical transparency in the ultraviolet-infrared spectral range and low probability of non-radiative phonon assisted relaxation. In spite of this, only few works report on optical characterization of Eu or Tb doped HfO₂ nanotubes prepared by sputtering via PVP template and Er-doped sol-gel SiO₂-HfO₂ waveguides. Recently, magnetron co-sputtering has been applied to produce HfSiO_x films doped with Pr³⁺ or Er³⁺ [1,2]. It was shown that contribution of rare-earth ions and host defects (oxygen vacancies, band tail states) in lighting process can be comparable.

In this work, the effect of deposition conditions and annealing treatment on the structural and optical properties of undoped and rare-earth doped $HfSi_xO_yN_z$ films was investigated by means of FTIR, XRD, TEM, Auger spectroscopy and photoluminescence (PL) methods. The sputtering was performed in Ar-N₂ plasma on Si substrate kept at 45-400°C. The films were annealed at 700-1100°C in nitrogen flow for 15-60 min. Obtained results revealed a phase separation and formation of Si-rich and Hf-rich phases in the films accompanied by an appearance of PL emission in the visible-near-infrared spectral range. It was found also that the matrix crystallinity affects significantly the shape of rare-earth PL band. An enhancement of rare-earth emission under non-resonant excitation was attributed to an effective energy transfer from Si nanoclusters and/or host defects. Obtained results offer multifunctional applications of elaborated films as luminescent materials for traditional phosphors applications.

1. Y.-T. An, C. Labbé, L. Khomenkova, M. Morales, X. Portier, F. Gourbilleau, Nanoscale Research Letters, 8, 43 (2013).

2. L. Khomenkova, Y.-T. An, D. Khomenkov, X. Portier, C. Labbé, F. Gourbilleau, Physica B, 453, 100 (2014).