Light Emitting Diodes (LEDs): Seeing the Luminescence Centers in Silicon Aluminum Oxynitrides by Advanced Electron Microscopy

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Solid-state lighting (SSL) is a technology in which traditional incandescent and fluorescent lamps are replaced by light-emitting diodes (LEDs) for general lighting purposes. An SSL tool generates visible light due to the propagation occurring in different wavelengths as a result of stimulation of the crystal structure of the specially formulated semiconductor-based diode material. Today, applications of LED based lamps can be seen in many areas such as traffic lighting, electric board, motorized vehicle and interior–exterior building lighting. The main advantages of LED lamps compared to conventional incandescent and fluorescent lamps are high energy conversion efficiency, long service life and they are not including hazardous substances, and ultraviolet-infrared radiation. Therefore, it is the main crystal structure of the semiconducting material that allows the device to illuminate in the desired color by flaring with the passing of an electric current through the most important component of the LED lamps. Silicon aluminum oxynitrides (SiAlONs) are new generation solid-solution ceramics derived from the same crystal structures of α-β/Si_3N_4. Depending on the sintering dopants, the desired engineering properties of SiAlONs for especially white light emitting diodes (LEDs) can be tailored by microstructure, primarily consisting of two distinct polymorphs (α and β), triple junction phases and grain boundaries. Therefore, unveiling the luminescence centers at atomic-scale is a key step in further developing and designing the LEDs from SiAlONs.

Here, we report the atomic-scale imaging and spectroscopy of gas pressure sintered Yb, Ce, Yb-Ce and Sm containing single and double rare-earth cation doped α/β-SiAlONs as well as TiN reinforced β-SiAlON ceramics using different type of state of the art aberration-corrected STEMs and EELS. Based on the results, it was demonstrated that: (i) Yb and Ce dopants were preferentially incorporated into the β-SiAlON crystal structure at the atom-specific lattice locations, with higher solubility for Yb than Ce; (ii) this observation was also confirmed in the Yb-Ce co-doped and Sm systems; (iii) Ce atoms without any co-doped cation were present in the triangular-like host sites of α-SiAlON unit-cell, accommodating much more atoms than Ce-doped β-SiAlON; (iv) Yb and Ce atoms were periodically and differently arranged in the grain boundaries, explaining that they are in semi-crystalline nature, not completely amorphous; and (v) Ti atoms were incorporated into the β-SiAlON unit-cell. Atomic-resolved advanced microscopy and spectroscopy results show the direct visualization of sites and solubility of rare-earth and transition metal atoms in α-β SiAlON unit-cells and grain boundaries. This capability offers new atomic-level engineering insights into how appropriate rare-earth types and SiAlON host polymorphs should be chosen for nanoscopic tailoring of both next-generation SiAlON-LEDs. We expect our observations of impurity sites and solubilities in SiAlON polymorphs and grain boundaries represents the first step towards

a new paradigm for atomic-scale guidance in the production of rare-earth and transition metal atoms doped LEDs.