Spectro-Pyrometric Measurements on Solid and Liquid Uranium-Americium Mixed Oxides near Their Melting Transition

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Radiance spectra of uranium-americium mixed oxides in the visible and near-infrared have been measured using a fast multi-channel spectro-pyrometer and a single-wavelength pyrometer operating near 650 nm. The disk-shaped samples were heated up to and beyond their melting transition using a powerful Nd:YAG laser. Quasi-containerless conditions were established by limiting the molten area to a spot smaller than the sample, with the surrounding solid forming a kind of "self-crucible". The experiments were performed in an autoclave filled with argon at a slight overpressure and with precise control of any oxygen traces. The autoclave is mounted inside a glovebox for radiation protection purposes with all laser optics and other instruments placed outside. In addition to the radiometric calibration of the pyrometers, uranium dioxide was used as a high-temperature secondary reference. The radiance spectra were converted into temperature based on the typical gray-body behavior of uranium-, plutonium-, and minor-actinide oxides. Thus, the melting temperatures of uranium-americium mixed oxides with an americium mass fraction up to 20 % were obtained. The high-quality samples were synthesized using two different liquid routes, ensuring homogeneity at the molecular level (as opposed to blending or infiltration processes used in the past). Thus, for the composition containing americium mass fractions of 1 and 5 %, the well-established ammonia co-precipitation procedure followed by thermal decomposition was applied. For the higher americium contents (10 and 20 %), the innovative hydrothermal conversion of mixed oxalate process was applied.