Preparation of mixed strontium-lead hafnate by wet chemical methods

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The search for new oxides applicable as high-energy radiation detectors is connected to parameters such as high effective atomic number Z_{eff} , high light yield and resolution, and, in some applications, fast response. The first quality - Z_{eff} - is closely related to the material density that, at high values, can be also beneficial in enhancing the compactness and may allow for smaller dimensions of constructed detectors. In the case of mixed oxides, the density is generally related to the density of the constituent oxides. Therefore oxides based on heavy cations, e.g. Lu^{3+}, Bi^{3+}, Pb^{2+}, are explored, among them also HfO_2 and hafnates.

The scientific investigation of strontium hafnate, $SrHfO_3$, started due to its high dielectric constant and large band gap. Additionally, it has a high effective atomic number (Z_{eff} = 63.10) and high density (7.56 g/cm^3) which makes it a promising material for ionizing radiation detection. As activators, Ce^{3+} ions are usually used and Ce:$SrHfO_3$ has been thoroughly studied. In search for other dopants, Pb^{2+} was proposed due to its ionic radius in 12-fold coordination being similar to that of Sr^{2+} (1.58 and 1.63 Å, respectively). Supposedly, as the content of lead increases in (Sr,Pb)HfO_3, the density increases up to 10.2 g/cm^3 in $PbHfO_3$. Additionally, unlike Ce^{3+}, the equal charge of strontium and lead cations does not induce the necessity to compensate the charge imbalance. Moreover, the Pb^{2+} cations emit at about 340 nm thanks to the partially allowed 6s-6p transition.

Historically, a solid state reaction has been the most widely used way to synthesize mixed oxides. However, apart from high energy and time demands (long sintering times at high temperatures), the major drawback of this method is the impossibility to guarantee homogeneity of the samples, which is crucial when the material is being doped with a minor fraction of a dopant cation. Wet chemical methods, on the other hand, thanks to their ability to transfer the homogeneity of a solution to the solid state, have been increasingly successful in the preparation of a huge range of materials. In this work, two wet routes were used to prepare (Pb,Sr)HfO_3: standard Pechini polyesterification and oxidative precipitation using hydrogen peroxide. A series of hafnate samples, in which the ratio of strontium and lead cations was gradually changing according to the stoichiometry $Sr_{1-x}Pb_xHfO_3$ (x = 0-1), were prepared. Their phase composition was determined by XRD and their luminescence properties were measured.

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