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#11-180 Lead Halide Perovskite Nanocomposite Scintillators

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Lead halide perovskites (LHPs) have many characteristics that are of interest when considering their use as scintillators: due to the high atomic number of lead, LHPs have high X-ray stopping power; their luminescence wavelength may be tuned throughout the visible region through adjustments to their composition (this wavelength could be chosen to fit the application of the detector, e.g. to match the maximum quantum efficiency of a given photomultiplier tube); LHP nanocrystals are solution-processable at room temperature, which allows for low-cost manufacture; these materials have also been shown to have a fast response time as well as a high light yield. During the last three years, articles relating to the development of LHP nanocrystal scintillators have been published by several groups. In each of these cases, all-inorganic nanocrystals were set into films of inert plastic that were thinner than 2 mm. The development of thicker composites with high nanocrystal loadings is typically limited by transmission losses caused by scattering of light from the nanocrystals. The negative effect of this scattering increases with particle size, and is therefore exacerbated by the formation of nanocrystal aggregates within the composite scintillators.

Here, nanocomposites up to 1 cm thick have been produced by loading nanocrystals of either formamidinium lead bromide ($FAPbBr_3$) or caesium lead halide ($CsPbX_3$) into plastic scintillator. The resultant samples combine the scintillation properties of both components, while protecting the nanocrystals from environmental degradation. A chemical ligand has been used to suppress aggregation of the nanocrystals, paving the way for production of nanocomposites with high loadings of LHP nanocrystals. Ultimately, the aim of this work is to produce a plastic scintillator in which the inclusion of a large proportion of high-Z LHP nanocrystals allows for the observation of photoelectric absorption peaks.

Hybrid organic-inorganic $FAPbBr_3$ nanocrystals have been synthesised using a room-temperature ligand-assisted reprecipitation method. A solution of perovskite precursors is injected into a second solution which contains chemicals that serve to stabilise the perovskite and to prevent excessive crystal growth, as well as an antisolvent which causes the yellow nanocrystals to precipitate. This method produces particles of a range of sizes, which may then be separated into two groups using a centrifuge. The smaller set of particles are typically less than 30 nm in size and are small enough to demonstrate quantum confinement effects, while the larger particles are typically 300 - 500 nm in size and are stable in a toluene dispersion for a period of more than a year.

Photoluminescence spectra were measured at room temperature using a 20 mW, 405 nm laser. For $FAPbBr_3$ nanocrystals in a toluene dispersion, luminescence maxima were recorded at 525.2 nm for the smaller set of nanocrystals, and 541.5 nm for the larger set of particles. The shorter emission wavelength measured for smaller nanocrystals is an effect caused by quantum confinement, which also leads to brighter emission for the small nanocrystals. The Stokes shift (the difference between emission and absorption maxima, which provides a quantitative measure of self-absorption) for LHP materials is small, but also has a size-dependence: consistent with the literature, we measured Stokes shifts of 70 meV for our smaller nanocrystals, and 10 meV for our larger nanocrystals. This means that there is reduced self-absorption in the smaller nanocrystals, which will reduce losses in the finished nanocomposite. The photoluminescence decay time of the FAPbBr3 nanocrystal dispersion was measured to be 36.1 ns, consisting of two components with time constants of 8.2 ns and 60 ns respectively.

A PVT-based plastic scintillator was used a basis for the nanocomposites, produced from the Eljen Technology EJ-290 casting kit. Prior to casting the scintillator, a concentrated dispersion of LHP nanocrystals was mixed into the partially-polymerised resin. The mixture was poured into moulds and then cured in a water bath.

In order to prevent aggregation of nanocrystals within the nanocomposites, a chemical ligand was attached to the nanocrystals after synthesis. The ligand allows bonds to form between the LHP nanocrystals and the PVT

polymer chain during the casting of the scintillator, hence preventing the nanocrystals from aggregating. This ligand-assisted strategy had previously been used by other groups to achieve aggregate-free loading of up to 60% in composites of inorganic quantum dots in PVT. However, the ligand had never before been applied to perovskites.

Initially, nanocomposite scintillators were produced that contained a low (1%) loading of larger nanocrystals either with or without the chemical ligand. The ligand was successful in preventing aggregation of nanocrystals, and the increased uniformity of the nanocrystal loading could be seen visually in the samples. Elemental mapping of the scintillators was also carried out, using energy dispersive x-ray spectroscopy. This confirmed the location of nanocrystals within the surface layer of the composites, providing an image of the differences in nanocrystal aggregation between the two types of sample.

Photoluminescence and radioluminescence spectra have been recorded for these nanocomposites. In these, a luminescence peak resulting from the nanocrystals can be clearly seen, replacing the five luminescence peaks that would normally result from the un-loaded plastic scintillator. The LHP emission peak recorded in radioluminescence was weaker than that for photoluminescence, which is a consequence of the opacity of the sample: while in photoluminescence measurements the luminescence is generated at the surface of the sample and easily escapes to the detector, in radioluminescence measurements luminescence is generated deeper into the sample and suffers losses from scattering and re-absorption while travelling out of the scintillator.

In future work, nanocomposites will be produced which contain smaller nanocrystals (<30 nm size) as well as the aggregate-reducing ligand. Our calculation of the effect of Mie scattering with different particle sizes leads us to believe that this improvement in the nanocrystal size will improve the efficacy of the ligand and will have a significant effect on scattering. This development will therefore increase the transparency of the composites, improving the light output and allowing for higher loadings of nanocrystals.

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