Liquid scintillators enriched with high-Z metal oxide nanoparticles for rare events search

P. Moiraghi[1], F. Cova[1], S. Mostoni[1], M. Fasoli[1], L. Gironi[2,3], and M. D'Arienzo[1]

[1]*Department of Materials Science, University of Milano-Bicocca, 20125 Milan, Italy*
[2]*Department of Physics, University of Milano-Bicocca, 20126 Milan, Italy*
[3]*National Institute of Nuclear Physics (INFN), Milano-Bicocca section, 20126 Milan, Italy*
Corresponding Author Email: p.moiraghi@campus.unimib.it

Liquid scintillators (LSs) have emerged in recent years as promising alternatives to conventional solid-state scintillators thanks to their easy scalability to high volumes, the high degree of purity and overall homogeneity, and the reduced costs of production and implementation. For these reasons, LSs in various formulations are widely used in large-scale neutrino physics experiments such as JUNO, SNO+ and KamLAND-Zen [1]. LSs are also applied on a smaller scale to investigate radioactive materials or isotopes that can be directly dissolved in the solvent; this gives a significant advantage for detecting and counting low-level β activity and in the search for the neutrino-less double-β (0νββ) decay, with the possibility of easily dispersing candidate isotopes in the scintillator itself [2].
However, the organic composition of LSs leads to a comparatively low effective atomic number (Z_{eff}) and density; this causes incident high-energy (in the order of MeV) γ photons to preferably undergo Compton scattering, leading to incomplete energy deposition in a relatively small detector (in the order of a few cm^3). A strategy to enhance the probability of the primary interaction with γ photons and their complete energy deposition in LSs is to load them with high-Z materials, thereby increasing the Z_{eff} of the resulting mixture and improving their detection efficiency. Thanks to their cost-effective scalability and easy incorporation of high-Z species in large amounts, LSs offer a very promising and versatile alternative both to large inorganic crystals, whose growth can become a difficult and expensive process, and to nanocrystals-loaded plastic scintillators, which can exhibit aggregation phenomena and uneven distribution of dispersed species.
In this work, the loading of a commercial LS (Ultima Gold AB, Revvity) with non-luminescent high-Z tungsten-based nanoparticles (NPs) is investigated as a potential strategy to enable a more efficient detection of γ-rays by increasing the Z_{eff} and density of the mixture. Various synthetic strategies are explored; microemulsion and precipitation approaches using WCl_6 and NH_3 as precursors result in bare tungsten-based NPs, while a wet impregnation route of pre-synthesised and monodispersed Stöber silica NPs (~ 30 nm) with Na_2WO_4 yields supported tungsten-based NPs. These are subsequently functionalized to improve the compatibility with the LS and prevent aggregation. The influence of synthetic parameters, such as the precursor molar ratio (WCl_6/NH_3), on the composition, morphology and size of the NPs and, in turn, on the optical and scintillation properties of the loaded LS, is investigated by means of structural and spectroscopic techniques. Diffuse reflectance spectroscopy reveals a strong dependence of the nature of bare NPs on the WCl_6/NH_3 ratio, with a threshold around 1:6, whereby NPs synthesised at ratios of 1:6 or lower exhibit an absorption onset at 2.6 eV, while NPs synthesised at higher ratios show a shifted onset located at 3.4 eV. Moreover, TEM micrographs show a uniform coverage of silica NPs with tungsten-based species. Under soft X-ray excitation, dispersions in the LS prepared both with bare NPs synthesised with a high WCl_6/NH_3 ratio and with silica-supported NPs show a promising increase in the radioluminescence intensity with respect to the unloaded LS with increasing loading fraction. Measurements of the scintillation response through excitation with ^{22}Na, ^{57}Co, and ^{241}Am sources with monochromatic γ-ray emissions are performed to reconstruct the spectra of energy deposited in the loaded LS and investigate the shift in the position of the Compton edge with respect to the unloaded LS.

1. S. Schoppmann, "Review of Novel Approaches to Organic Liquid Scintillators in Neutrino Physics", *Symmetry*, **15**, 1 (2023).
2. S. Arai et al., "Development of liquid scintillators loaded with alkaline earth molybdate nanoparticles for detection of neutrinoless double-beta decay", *J. Ceram. Soc. Japan*, **127**, 1 (2019).