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H2 storage mechanism in fullerides studied with μSR

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When positive muons (μ+) are implanted in insulating materials, they capture electrons to form muonium (Mu), a light isotope of H. This process makes muon spin resonance technique (μ SR) suitable for studying H interaction with matter, for example in hydrogen storage (HS) materials.
Among carbon-based materials, recently metal intercalated fullerides demonstrated to be promising for HS, representing de-facto a novel class of HS compounds: in particular, it has been shown that both lithium and sodium cluster intercalated fullerides can reversibly absorb relevant amount of hydrogen (up to 5 wt % in case of Li6C60), at thermodynamic conditions much milder than what observed in pure C60. However, the hydrogenation mechanism in these systems is not trivial and involves several processes, difficult to disentangle with conventional techniques.
In this work we show how uSR helped us to shed light on the hydrogenation process of these systems. In detail, we performed a μSR investigation of Li6C60 and Na10C60, either as-prepared or after hydrogenation, on the EMU and ARGUS beamlines, at ISIS-RAL. Interestingly, we found that in these compounds the formation of muonium is not inhibited, thanks to the presence of the intercalated partly ionized alkali clusters. Muonium was found to react with C60 to form adduct radicals, appearing as a missing fraction in the muon spin signal. This phenomenon is dependent on temperature and is invariably enhanced on cooling for all the investigated samples.
Such findings indicated that in these systems C60 hydrogenation is already feasible at cryogenic temperatures, with an efficiency even larger than at high T, while the high T needed for hydrogen storage in fullerides is only required to overcome the alkali metals mediated H2 dissociation barrier. Following this hint, we managed to further enhance the hydrogen absorption by co-intercalating transition metals nanoparticles (Pt, Pd) in the fullerides interstices.

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