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Deseada Díaz Barrero
Tritium Laboratory of Karlsruhe-Institute Astroparticle Physics
Karlsruhe Institute of Technology

MONITORING OF THE SYNTHESIS AND TEMPORAL EVOLUTION OF TRITIATED METHANE MIXTURES BY RAMAN SPECTROSCOPY

D. DIAZ BARRERO[1], T. L. LE[1], M. SCHLÖSSER[1], H. H. Telle[2],

[1] *Tritium Laboratory Karlsruhe (TLK), Institute for Astroparticle Physics (IAP), Karlsruhe Institute for Technology (KIT)*
[2] *Departamento de Química Física Aplicada, Universidad Autónoma de Madrid, Campus Cantoblanco, 28049 Madrid, (Spain)*

Tritium, the radioactive isotope of hydrogen, is an indispensable reagent for future tokamak-type nuclear fusion reactors (like the International Thermonuclear Experimental Reactor, ITER). It also is the key component for the Karlsruhe Tritium Neutrino mass experiment (KATRIN) at the TLK of the Karlsruhe Institute of Technology, which aims to obtain neutrino masses with higher precision. Using tritium in large quantities, radiochemical reactions take place during their circulation through the system, leading to molecular products, such as water isotopologues, Q_2O, and tritiated methane species, CQ_4 normally accumulating in low concentration (where Q can be protium (H), deuterium (D) and tritium (T).

Raman spectroscopy has become an essential analytical tool for the in-line monitoring of these gaseous mixtures. However, spectroscopic knowledge of tritiated methanes is rather incomplete, due to the very low concentrations used in the investigations to date. For the results presented here the CQ_4 compounds were synthesised in large quantities using a stepwise enrichment procedure based on catalytic isotope exchange reactions to increase the concentrations to more than 20%, as well as the rate of tritiation of the methane molecules. Therefore, several vibrational bands could be identified that had not been observed experimentally so far.

The resulting mixtures were composed of the family $CT_xH_{(4-x)}$, as well as the hydrogen molecular isotopologues H_2, T_2 and HT. The composition of the samples has been evaluated by depolarisation Raman spectroscopy, allowing the semi-quantification through the $\nu_1(Q_1)$ bands of the different species of the isotope-substituted methane family. The spectroscopic data also reveal that the composition of the samples evolve over time. Complementary mass spectrometry measurements showed that several of the observed Raman signals correspond to molecules of up to 5 carbon atoms. Our preliminary analysis indicates that the radiochemical processes produce tritiated hydrocarbons with a predominance of fully tritiated ethylene.

Keywords — catalysis, small molecules, Raman, dynamics/kinetics