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Performance evaluation of the developed in-situ ortho-to-parahydrogen fraction measurement system for the ESS cryogenic moderator system

At the ESS target, high energy spallation neutrons are generated by impinging a 2 MW proton beam on the tungsten target. The proton beam is pulsed with a repetition of 14 Hz and a pulse length of 2.86 ms. The moderator system consists of a water pre-moderator and two liquid hydrogen cold moderators, which are optimized to achieve a high cold neutron brightness. The neutronic performance of the cold moderators degrades rapidly when parahydrogen fraction falls below 99.5%. The neutron collisions in the cold moderators increase the orthohydrogen fraction unless compensated by the ortho- to parahydrogen conversion driven by the catalyst system. Therefore, the Cryogenic Moderator System (CMS) is equipped with an ortho-parahydrogen (OP) catalyst to sustain the desired high parahydrogen fraction. The ortho-to-parahydrogen ratio of liquid hydrogen at the inlet and outlet of the moderators will be measured by the in-situ measurement system (OPMS) using a Raman spectroscopy. A Raman optics system with an accuracy of 0.1% parahydrogen fraction is essential to detect any undesirable shift towards a higher orthohydrogen fraction induced by para-to-ortho back conversion driven by the neutron collisions in the cold moderators. The developed Raman optical probe system has successfully achieved our primary goal of 0.5% accuracy in measurement, as it can detect the fourth parahydrogen peak (J= 4) of normal hydrogen gas at 1240 cm-1.

In this study, Raman spectra of normal hydrogen at ambient temperature were measured while varying hydrogen pressures from 0.10 MPa (0.0827 kg/m3) to 1.1 MPa (0.896 kg/m3). The intensities at J= 0 to 4 were proportional to the hydrogen density. For the pressures higher than 0.7 MPa, the ortho-to-parahydrogen ratios can be measured within an accuracy of ±0.1%. We measured Raman spectra of hydrogen gas passing through a catalyst cooled by liquid nitrogen. The measured ortho-to-parahydrogen ratio was 0.9879±0.0044 and matched closely with the theoretical values of 0.9967. Furthermore, the ortho-to-parahydrogen ratio of hydrogen obtained from the J-PARC CMS under the nominal condition at 21 K was also measured at a pressure of 0.7 MPa and a temperature of 293 K. The parahydrogen fraction was measured to be 99.74 within an accuracy of ±0.11%. It can be expected that the developed Raman optical probe system would achieve the required accuracy of 0.1% for measuring parahydrogen in liquid hydrogen at the ESS CMS, given the significantly higher density of liquid hydrogen.

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