A kinetic study of the catalytic conversion of ortho- to parahydrogen for the ESS liquid hydrogen moderator

A summary of the thesis work by Emil Karlsson

High parahydrogen content is an important factor for the ESS liquid hydrogen moderator to maximize the neutron brightness. Parahydrogen is obtained by catalytic conversion of orthohydrogen and this thesis work is dedicated to the kinetic study of the orthohydrogen conversion for two types of catalysts. This will provide useful information for the selection of the catalyst to be used by ESS.

The conversion reaction for the IONEX® catalyst (composed of an iron oxide) followed first order kinetics, which is in agreement with the literature [1]. The data could be fitted to an exponential curve, showing the decrease of orthohydrogen content. The reaction using OXISORB® (silica doped with chromium oxide) on the other hand exhibited kinetics represented by two first order reactions, which are dominant during different stages of the catalytic process. The results show that the amount of OXISORB® catalyst needs to be approximately seven times greater, in order to obtain conversion times corresponding to the experiments of the IONEX® catalyst.

The European Spallation Source (ESS) is building the world's largest neutron source providing a neutron brightness which is about 3.5 times greater than existing sources, to lead advanced materials research in the world. The remarkable efficiency of the ESS facility is due to the ground breaking design of the liquid hydrogen moderator, which meets the specific demands of the structure and concentration of the moderating medium.

The hydrogen molecule exists in two forms- ortho and parahydrogen, which is a result of symmetry restrictions of the hydrogen atoms. Parahydrogen is favored at lower temperatures and is about 99.8% at 20K (-253°C). Natural conversion of ortho- to parahydrogen is extremely slow, but can be accelerated by the presence of a catalyst.

Optimal efficiency of the ESS moderator is achieved for concentrations of parahydrogen above 99.5% in the liquid hydrogen. Concentrations below this limit results in a significant drop in neutron brightness. Orthohydrogen is formed during the operation of the moderator which results in a decrease of the parahydrogen content. The circulation of liquid hydrogen between the moderator and the catalyst reconverts orthohydrogen into parahydrogen. The optimal catalyst corresponds to and facilitates a parahydrogen concentration above the lower limit of 99.5% and result in the use of a lesser amount of the catalyst and a smaller hydrogen storage, which is favorable from a safety perspective.

Information regarding the catalytic conversion of ortho- to parahydrogen under the operating conditions of the ESS moderator is not available for the IONEX® and OXISORB® catalysts. This work aims to provide some preliminary kinetic data for the choice of catalyst.

The parahydrogen content is monitored using Raman spectroscopy, which is a new method for the ESS facility. The thesis work includes the design and configuration of the analysis setup. The present results aid in the implementation of a qualitative analysis method for the continuous online measurement of parahydrogen content in the moderator.

This thesis work has been a collaboration between the European Spallation Source (ESS) and the Materials Engineering Division at Lunds Tekniska Högskola (LTH).

References

[1] I. F. Silvera, "The solid molecular hydrogens in the condensed phase: Fundamentals and static properties," *Rev. Mod. Phys.*, vol. 52, no. 2, pp. 393–452, Apr. 1980.