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Neutron radiography analysis of a hydride-based hydrogen storage system

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ABSTRACT

A hydrogen container based on the use of a hydride forming material ($LaNi_5$) has been specifically designed for performing in-situ Neutron Imaging experiments. The device allows following the process of heat-induced hydrogen release from the initially fully hydrided material. A reaction front forms in the material around the central heater. As the experiment proceeds, the front expands radially. Later on, heat conduction by the container walls produces the decomposition of the hydride in contact with them. As a consequence, and for the implemented geometry, the front advances at an almost constant radial speed. A simple analysis of the images contrast allows the quantification of the ratio between the hydride and metal phases for the complete experiment.
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Introduction

Neutron Imaging (NI) comprises a series of techniques that transform the interaction between neutrons and an object under study into optical images. Among them, neutron radiography (or *neutrography*) is a transmission visualization technique based on the interaction of neutrons with the elements of a material [1]. It is somehow comparable to X-rays radiography. However, while X-rays interactions depend monotonically on the atomic number of the elements, neutron interaction with matter is related to a cross section that varies not only from element to element but even between different isotopes of an element. In particular, light elements that are virtually invisible to X-rays, like hydrogen or boron, produce strong interactions with neutrons [2]. Conversely, there are heavier elements that produce strong X-rays contrast, like aluminum or zirconium, which are weak neutron attenuators [2]. In addition, neutron penetration of materials is considerably higher than that of X-rays. These characteristics turn neutron radiography (NR) into a useful visualization technique for situations involving strong neutron attenuators such as water, polymers or other hydrogen compounds [3–7].

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The NR technique can be applied to the study of hydride forming materials (HFM) for hydrogen storage, as demonstrated early on by Sakaguchi et al. [8,9]. They used NR to detect hydrogen in Mg_2Ni storage alloys, following the first stages of hydride formation and quantifying the amount of hydrogen in the material. Later on, Jacobson et al. [4] and Wood et al. [10] used a combination of NI techniques to study the reaction pattern of an AB_5 alloy inside a cylindrical container during hydrogen absorption. Their results show that the formation of the hydride phase inside the container is not homogeneous but strongly depends on the material localization. Pranzas and collaborators performed in-situ studies of hydrogen sorption in a massive reactor containing $NaAlH_4/TiF_3$ powder [11,12]. Their results show how the powder, evenly distributed inside the container at the beginning of the experiments, is affected by the hydrogen inflow. The striking images display the formation of a tree-like structure after the first hydrogenation cycle which essentially remains after the following cycles of hydrogen charge/discharge. The non-even powder distribution can negatively affect the container overall performance as a hydrogen storage system.

A particularly interesting example of NI applied to the study of HFM is the work of Gondek et al. [13] which presents a step-by-step observation of the hydride formation process in an AB_5 alloy inside a container. The images show that hydriding occurs first near the container walls, thanks to the higher heat conduction of Al in comparison to the powder.

The processes of hydrogen incorporation and release by a massive hydride-based storage container are usually not observable due to the metallic container walls. However, it has been established that they are largely dominated by the poor thermal conductivity of the hydride forming material [14]. When considering containers design, a great deal of attention is placed on improving heat exchange between the powder material and external heat source/sink. NR may provide useful information about how hydriding and dehydriding reactions take place inside a container. This information could serve to elaborate functional container models, aiding container design for improved heat exchange and reaction kinetics. Considering the NR technique potential, the amount of publications related to hydrogen storage studies is relatively small. One of the reasons is that these studies usually involve managing relatively large amounts of hydrogen at high pressure which could be considered dangerous at NR facilities nearby experimental nuclear reactors or other neutron sources. Another reason is that hydride-based storage containers are usually built with thick steel walls that affect NR contrast, although there are examples of Al made containers [11–13]. Our approach consisted in designing a steel/aluminum container suitable for following hydride reactions by means of NR without losing the usual characteristics of massive hydride-based hydrogen storage devices, i.e. containing a relatively large amount of HFM and keeping the possibility of conducting pressure and temperature triggered processes.

This work is then aimed at using NR as a tool for in-situ following reactions inside a massive hydride-based hydrogen storage container. In particular, we present the container design, describe the specific experimental setup and discuss the results corresponding to the case of thermally induced hydrogen release from the fully hydrided material.

Experimental

Neutron radiography facility

For this work, we have used the NR facility of RA-6 experimental nuclear reactor at Centro Atomico Bariloche, Argentina. The facility provides a 20 cm × 20 cm square neutron beam. By design, the ratio between the source to sample longitude (L) and beam aperture (D) is fixed to 100 (L/D = 100). With the reactor operating at 500 kW, the neutron fluxes are 2.54×10^6 n/cm^{2}s (thermal) and 2.93×10^4 n/cm^{2}s (epithermal). The detection system collects the transmitted beam and consists of a 20 cm × 20 cm scintillation screen made of 6LiF/ZnS doped with Ag, made by Applied Scintillation Technologies The scintillation screen light peak is at $\lambda = 450$ nm. The light is reflected out of the beam path by a set of two front surface mirrors in order to protect the CCD camera from the incoming radiation. A Schneider Kreuznach Xenon condensing lens focuses the image onto the camera. Both, the lens and the camera are placed inside a light-tight box. The camera is a Penguin 600 CLM from Pixera Corporation. It has a maximum resolution of 2776 pixels × 2074 pixels and provides digital depth of 16 bits. The CCD dark current is reduced by a four stage thermoelectric Peltier cooling device.

In NR experiments, the neutron transmittance (T) of a material is usually calculated from the ratio between the transmitted (I) and incident (I_0) intensities as [15]:

$$T = \frac{I}{I_0} = \exp(-\sigma \rho d/M) \tag{1}$$

where σ is the total neutron cross section of the material, d is its thickness, ρ the material density (in g/cm^3) and M the atomic weight (in g/mol). This expression does not take into account the possibility of having multiple scattering effects and it is the simplest description of the experiment.

Hydride-based hydrogen storage device

The container is a cylinder with a stainless steel body and two aluminum lids for mechanical resistance. A detailed description of the container design can be found in Ref. [16]. Here, we only present its main characteristics. The body has been machined from a 304L stainless steel single piece (Fig. 1a). Stainless steel was selected because it does not react with most HFM and has poor thermal conductivity and good temperature resistance in comparison to other metals (e.g aluminum). The cylinder is 14 mm high and has external and internal diameters of 125 mm and 88 mm, respectively. The cylinder floor is 1 mm thick. The floor has a central hole where a small Cu cylinder that hosts a resistive heater was placed and welded. The body cavity is filled with the HFM ($LaNi_5$) and closed by an upper 304L stainless steel lid with a thickness of 3.2 mm that rests over an O-ring. The cavity height and stainless steel floor and lid thicknesses were chosen by considering their impact on the contrast of NR images, a task aided by computer calculation with a home-made program

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