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## Scattering influences in quantitative fission neutron radiography for the in situ analysis of hydrogen distribution in metal hydrides



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### ABSTRACT

In situ neutron radiography allows for the time-resolved study of hydrogen distribution in metal hydrides. However, for a precise quantitative investigation of a time-dependent hydrogen content within a host material, an exact knowledge of the corresponding attenuation coefficient is necessary. Additionally, the effect of scattering has to be considered as it is known to violate Beer's law, which is used to determine the amount of hydrogen from a measured intensity distribution. Within this study, we used a metal hydride inside two different hydrogen storage tanks as host systems, consisting of steel and aluminum. The neutron beam attenuation by hydrogen was investigated in these two different setups during the hydrogen absorption process. A linear correlation to the amount of absorbed hydrogen was found, allowing for a readily quantitative investigation. Further, an analysis of scattering contributions on the measured intensity distributions was performed and is described in detail.

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### 1. Introduction

Neutron radiography (NR) is known to be a unique tool for non-destructive testing and analysis, used for a number of applications. In ideal NR, the neutron beam intensity  $I$  measured in the detector plane is described by the fundamental Beer's law:

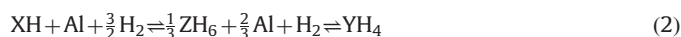
$$I = I_0 \cdot \exp(-\Sigma_{tot}d) \quad (1)$$

where  $I_0$  is the initial beam intensity without any attenuation. For a known thickness  $d$ , the sample specific total attenuation coefficient  $\Sigma_{tot}$  provides information about the quantitative amount of elements contributing to the total beam attenuation. While this method is widely used for the study of water transport phenomena, batteries and fuel cells [1,2], so far it has not been widely used in the field of hydrogen technology. In this paper we investigate the applicability of NR to the quantitative study of hydrogenation as it takes place within, e.g., a metal hydride bed. Considering such a hydride on hydrogenation, the total attenuation coefficient of the system changes and a quantitative NR requires the knowledge of the attenuation coefficient for hydrogen throughout the whole sorption process. Moreover, the influence of scattering on the measured intensity distribution in the detector plane is known to

cause a deviation from the ideal Beer's law, which needs to be corrected for [3–5]. Within this study, we use fission neutrons at the instrument NECTAR at FRM II to study the neutron beam attenuation caused by hydrogen during the sorption process inside the metal hydride sodium alanate. The latter was chosen as a model system, the evaluation methods and results are expected to be valid for any other metal hydride material or host system in general. Moreover, the results provide the basis for a future time-resolved and quantitative analysis of the hydrogen distribution in such systems. Furthermore, the effects of scattering contributions on the measured beam intensity are investigated.

### 2. The effect of hydrogen absorption on the neutron beam attenuation and time dependency of scattering

In the field of metal-hydride based hydrogen storage alanates form a class of so-called complex hydrides that operate in the medium temperature regime of  $T \in [100, 180]^\circ\text{C}$ . Starting from a desorbed state, the hydrogenation of alanates usually proceeds in the following two-step reaction [6]:



where X is an alkali metal and Y represents aluminum. The first reaction step is accompanied by the formation of a hexa-alanate,

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the alkali–aluminum compound is labeled here by Z, and with the completion of the second reaction step and the formation of tetra-alanate the material is fully hydrogenated. The corresponding alkali–aluminum compound is labeled by Y. Among the different alanate systems, sodium alanate is the one that has been most widely studied and we use it here as a model material to study the applicability of NR for the quantitative investigation of metal hydride systems. The procedure shown here can be readily transferred to more simple cases of one-step reactions as well as more complex reaction pathways with different reactants and/or products.

In general, for an in situ investigation of a hydrogenation of a metal-hydride by ideal NR, the total attenuation coefficient  $\Sigma_{tot}$  of (1) is 4-dimensional: the 3 dimensional information is related to the spatial information of the material distribution and the additional dimension is a time dependency due to the absorption of hydrogen by the metal hydride. Instead of considering the additional energy dependency of the attenuation coefficient related to the energy spectrum of the instrument we work with an averaged attenuation for reduction of complexity. For taking a ratio as it is described below and using large propagation distances between sample and detector spectral shifts in the projected volume of the metal hydride that are caused by moderation or beam hardening can be neglected in first approximation. This is discussed later on in the following sections. If the attenuation by the material can be assumed to be constant in time referred to the pixel area, the attenuation coefficient separates into a material and a hydrogen contribution,  $\Sigma_{tot}(r, t) = \Sigma_M(r) + \Sigma_{H_2}(r, t)$ , where only the latter carries the time-dependent information. This assumption is crucial to the separation of contributions and Eq. (3). Its validity within this study is checked later on. It is remarked that the attenuation coefficient of the material subsumes the metal hydride material itself as well as any enclosing material, which are storage tanks made of steel and aluminum used in this study. To investigate only the effect of the hydrogen absorption, the logarithm of the ratio of the intensities of two images is taken

$$\Delta\Omega(t) = \Omega_{H_2}(t) - \Omega_{H_2}(t_0) = -\ln\left(\frac{I(t)}{I(t_0)}\right) \quad (3)$$

where  $I(t)$  is the measured intensity at  $t$  during the absorption process and  $I(t_0)$  is the reference image of the material distribution before the hydrogen uptake. All images are corrected by a dark field measurement before. The difference  $\Delta\Omega^1$  is the attenuation that is caused only by the absorbed amount of hydrogen within the time interval  $\Delta t = t - t_0$ . It is combined with the corresponding amount of absorbed hydrogen for the same time interval to study the type of correlation between the two quantities. For a quantitative analysis of the hydrogen distribution inside the metal hydride material, the knowledge of this absorbed hydrogen vs. time dependency is a basic requirement.

Simultaneously to the evolution of  $\Delta\Omega$ , a time-dependent scattering influence has to be considered as well, as hydrogen is a strong scatterer in comparison to other elements. The scattering in total leads to an increased background intensity. In first approximation this background intensity is assumed to be spatially constant on the detector plane for large neutron propagation distances from the sample to the detector. In general, an effect of scattering can be denoted as a perturbation of the ideal Beer's law with almost no spatial dependency for propagation distances above 10 cm, which was shown in particular for fission neutrons in [7]. Thus, the perturbation is assumed to be spatially invariant, but time-dependent due to the increasing amount of hydrogen

during the sorption process. It is

$$I(r, t) = I_0 \cdot \exp(-\Omega(r, t)) + I_{sc}(t). \quad (4)$$

Here,  $I_{sc}(t)$  denotes the scattering background for a given initial beam intensity. Accordingly, the time evolution of the scattering contribution can be investigated by taking the difference of  $I(t) - I(t_0)$  in areas for a time constant attenuation, assuming a time-constant distribution of the initial beam intensity.

### 3. Results

#### 3.1. General

The measurements were performed at the instrument NECTAR at FRM II using fission neutrons [8]. A lead filter of 10 cm thickness for suppression of the gamma ray background was used. For conversion of fission neutrons into visible light a ZnS scintillator embedded in propylene was used. The acquisition of images was performed with a CCD-ANDOR DV-434BU camera with a pixel array of  $1024 \times 1024$  at a pixel size of  $0.27 \text{ mm} \times 0.27 \text{ mm}$ . Two setups have been used: 50.1 g of desorbed sodium alanate ( $\text{NaH} + \text{Al} + \text{additive}$ ) stored inside a steel storage tank has been investigated with a camera readout frequency of 5 MHz and a total time between two succeeding images of 252 s. Additionally, the threefold amount of the same material stored in a tank made of aluminum has been studied using a less collimated beam and a readout frequency of 1 MHz for reduction of exposure time with a total time between two succeeding images of 138 s. In this second setup an additional filter was installed in the beamline setup consisting of 1 cm of  $B_4C(50\%)$  embedded in epoxy. In order to avoid a strong scattering influence, the distance between the sample and the detector was set to 15 cm for both setups. For the reduction of the influence of image artifacts caused by radiation directly hitting the detector, a filtering algorithm for fission neutron imaging was applied in combination with an additional median filter [9]. To correlate the attenuation by the absorbed hydrogen with its corresponding amount inside the metal hydride material, the hydrogen flow from an external gas supply to the sample was measured by a mass flow controller. Additionally, the temperature and pressure at the sample inside the two storage tanks were recorded. These thermodynamic quantities allow for the calculation of the amount of absorbed hydrogen by the metal hydride material. By time-synchronization to the image acquisition, each image can be allocated to the respective amount of absorbed hydrogen. The performed absorption was the first absorption after the manufacture of the raw material. As an initial pressure application is known to cause a macroscopic structure deformation for loose powder [10,11], the metal hydride material in both storage tanks was pre-compacted so that the circular structure remains macroscopically constant. During the absorption process, the material structure can be assumed to remain macroscopically constant as well: for compacts of our model material sodium alanate, the volumetric expansion within the first absorption is negligible, since it is below 3% in radial dimension [12]. The change in axial direction is less important as the beam attenuation is integrated for this direction resulting in a projection of the volume on the detector area. However, the expansion in the axial direction is as well less than 3%. Additionally, no significant change of the material's diameter was observed by NR during the measurement.

#### 3.2. Time-dependent scattering component

Before processing the images in the way described in the first section to derive only the attenuation by hydrogen according to

<sup>1</sup> In the following we use the attenuation  $\Omega$  for the attenuation of the neutron beam on its path along the distance  $d$  inside the sample instead of  $\Sigma \cdot d$ .

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