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Watching the formation of metal hydrides in situ

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In the quest for low-cost, low-weight, high-capacity reversible hydrogen storage media, real-time in situ studies are gaining more attention recently. This is because it is now commonly accepted that the lack of reversibility is one of the key obstacles for the use of low-weight hydrogen stores. In order to follow the formation of crystalline solids both during the hydrogenation and the dehydrogenation process and including the hydrogen positions, in situ neutron powder diffraction is a powerful method.

We have constructed a gas pressure cell for in situ neutron powder diffraction of solid-gas reactions such as hydrogenation (deuteration). The sample holder is based on a 10 cm long free standing sapphire crystal tube [1]. By proper orientation of the single crystal Bragg peaks of the container material can be avoided, resulting in a very clean background as compared to most other designs for gas-pressure cells made e. g. of silica or metals. Using a laser heating and gas pressure controller, the hydrogenation (deuteration) of intermetallics can be studied in real time routinely up to 100 bar gas pressure and 700 K at present. At a time resolution in the order of one minute high quality diffraction data can be collected suitable for detailed Rietveld analysis in most cases.

This development allowed getting a deeper insight into reaction pathways of the hydrogen uptake in intermetallics and the formation of metal hydrides. In palladium the $\alpha \longrightarrow \beta$ transition of the hydride (deuteride) could be followed, yielding in a detailed picture of the H(D) distribution during the whole process. The hydrogenation of palladium rich intermetallics MPd3 (M = Mg, In, Tl) proceeds through metastable intermediates, which could be fully structurally characterized [2]. Moreover, a complete reaction mechanism was proposed, which allowed the planned syntheses of designed metastable intermetallic compounds and metal hydrides [2]. Further examples include the hydrogenation of Dy5Pd2, Zintl phases like SrGa2 and the light-weight hydrogen storage material Li3N, illustrating the potential and limitations of the sapphire single crystal based gas pressure cell for in situ neutron powder diffraction on solid-gas reactions.

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