Experimental report

13/09/2019

Proposal: 6-05-995 **Council:** 4/2018

Title: Hydrogen induced glass-to-glass transition in Mg-based metallic glasses

Research area: Materials

This proposal is a new proposal

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Samples: Mg based metallic glasses Mg70Ni20-xCe10Cux x=0,5,10 and 15
Mg based metallic glasses Mg70-xNi20Ce10Cux x=0,5,10 and 15

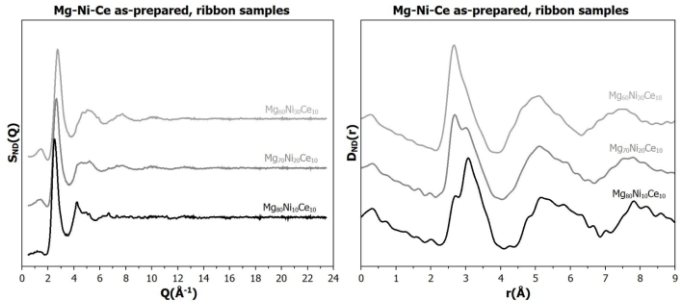
Instrument	Requested days	Allocated days	From	To
D4	5	4	17/10/2018	19/10/2018
			31/08/2019	02/09/2019

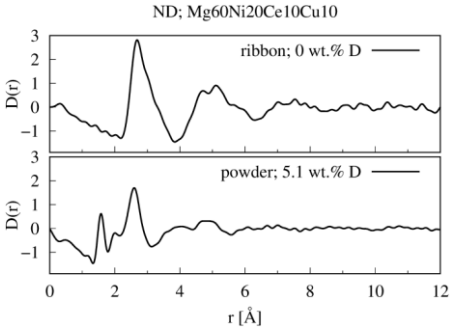
Abstract:

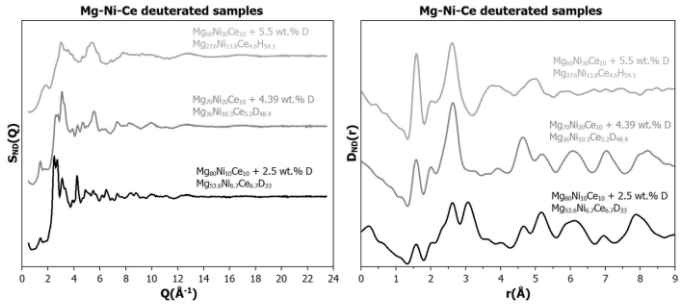
The exploration of favourable hydrogen storage materials is of great importance for the realization of a sustainable hydrogen energy society. Metallic glasses exhibit glass-to-glass transitions that may provide a number of opportunities for hydrogen storage.Their chemical compositions can be easily tuned by adding elements to alter their chemical interaction with hydrogen, hence leading to tunable hydrogen storage capacity, kinetics and thermodynamics. Mg-based MGs are of particular interest because of high hydrogen density of up to 7.6 wt% for MgH2. The de-/hydrogenation temperatures of Mg-based MGs, however, are far above their crystallization temperatures. Recently evidence of reversible hydrogenation and dehydrogenation at room temperature of the Mg-Ce-Ni metallic glass have been reported. We developed new Mg based glassy alloys which can store 4 to 5 wt.% of H2 (8 to 10 wt.% D2). To the best of our knowledge, this is the highest hydrogen storage capacity obtained in amorphous alloys to date. The alloys passes interesting hydrogen induced glass-to-glass transition which we want to investigate in detail by the proposed neutron experiment.

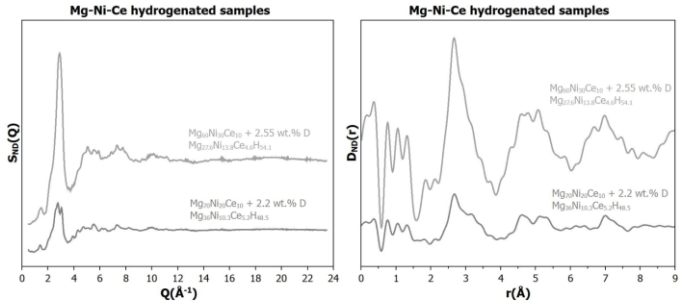
Experimental report 6-05-995

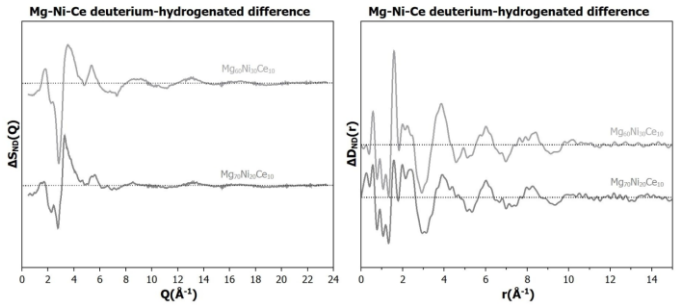
During the experiment we have measured and extract high quality neutron structure factors from the Mg-Ni-Ce and Mg-Ni-Ce-Cu metallic glasses in as-prepared and also deuterated and hydrogenated states. Figure down shows all the Mg-Ni-Ce structure factors normalised on absolute scale.



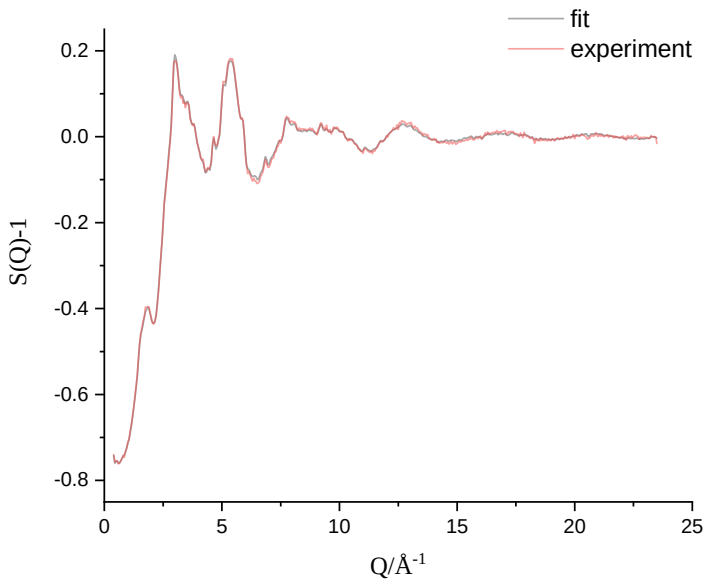




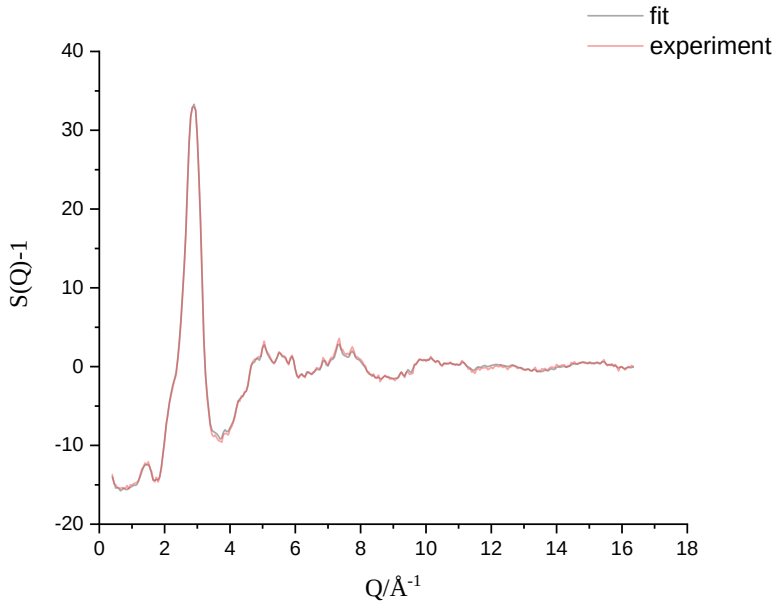




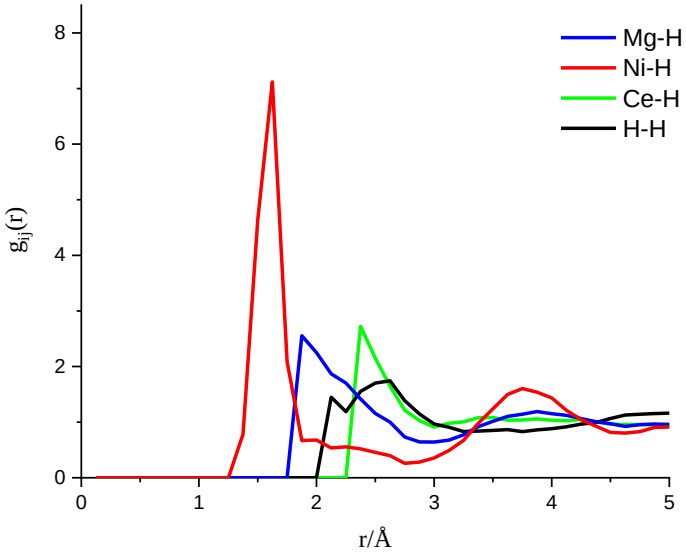
First results from atomic structure modelling



Experimental structure factor and RMC fit of $Mg_{60}Ni_{30}Ce_{10}$-D obtained by fitting simultaneously the $Mg_{60}Ni_{30}Ce_{10}$-D and $Mg_{60}Ni_{30}Ce_{10}$-H structure factors



Experimental structure factor and RMC fit of $Mg_{60}Ni_{30}Ce_{10}$-H obtained by fitting simultaneously the $Mg_{60}Ni_{30}Ce_{10}$-D and $Mg_{60}Ni_{30}Ce_{10}$-H structure factors



Hydrogen-related partial pair correlation functions of fully loaded $Mg_{60}Ni_{30}Ce_{10}$ (metal-to-hydrogen ratio 1:1.22)

Preliminary simulations revealed the presence of strong Ni-H correlations. The mean Ni-H nearest neighbour distance is about 1.59 Å. The corresponding average Ni-H coordination number is 2.51 ± 0.4. The peak of $g_{MgH}(r)$ is at about 1.9 Å. This distance distribution is strongly asymmetric. The reason for this may partly be the overlap of Mg-H, Ce-H and H-H peaks that makes their separation a difficult task. The situation may be improved after adding XRD and EXAFS data to the reverse Monte Carlo simulation (experiment at the DESY Germany in Septemver and October 2019). Those measurements are not sensitive directly to the H-related correlations the rearrangement of metal-metal g(r) functions may have an impact on the H-H and H-metal ones.