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Dynamics of porous and amorphous magnesium borohydride to understand solid state Mg-ion-conductors

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Rechargeable solid-state magnesium batteries are considered for high energy density storage and usage in mobile applications as well as to store energy from intermittent energy sources. Recently, magnesium borohydride, $Mg(BH_4)_2$, was found to be an effective precursor for solid-state Mg-ion conductors. The mechanochemical synthesis tends to form amorphous $Mg(BH_4)_2$ and it has been postulated that amorphous $Mg(BH_4)_2$ is increasing the conductivity in the Mg-ion conductors. Quasi-elastic neutron scattering (QENS) studies were employed to investigate the dynamics of porous and amorphous $Mg(BH_4)_2$. In general, QENS is needed to understand the local structure and dynamics in the precursor at different temperatures as well as at different energy- and momentum transfers. The results show that the low energy excitation spectrum in $Mg(BH_4)_2$ is strongly dependent on the local structure as can be seen by the comparison of as-received γ-$Mg(BH_4)_2$ and ball milled, amorphous compound. While as-received γ-$Mg(BH_4)_2$ shows almost no quasi-elastic scattering at 310 K, the ball milled version displays a significantly different low energy excitation spectrum and a higher rotational mobility of the [BH4] units. A high rotational mobility is proposed to be a fundamental necessity for high Mg-ion conductivity. This is supported by an almost two orders of magnitude higher conductivity in the ball milled sample compared to the as-received γ-$Mg(BH_4)_2$ at 353 K.

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