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Crystalline silicon gels as anode material for lithium-ion batteries

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Abstract

A silica gel has been synthesized via an efficient sol-gel method under microwave radiation, using tetraethylorthosilicate (TEOS) as the silica precursor and acid-basic conditions during the sol-gel process [1]. This approach makes it possible to obtain mesoporous silica gels in a short time, but amorphous in all cases, with presence of a broad peak at $2\theta = 17–29°$ that corresponds to the formation of amorphous silica according to JCPDS-card 96-900-1582.

Among the reduction methods used to obtain crystalline silicon, the magnesio-thermal reduction process stands out for its speed, control and yields [2,3]. Thus, our amorphous silica gels were mixed with Mg in a Mg:Si molar ratio of 2.5:1 and treated at 750°C under an inert atmosphere (Ar, 300 ml/min). During the heat treatment, different heating ramps were used to avoid overheating of the sample and to maintain the gel structure. The phases produced from the reduction process of SiO_2, such as MgO and Mg_2Si, were eliminated by washing with HCl (1 M) for 4 hours.

The structural properties of the obtained silicon gels were analyzed and measured by SEM, X-ray diffraction (XRD) and high-resolution transmission electron microscopy (HR-TEM). Fig. 1 shows the XRD data for all the reduced silicon gels obtained, illustrating the complete removal of SiO_2, with only Si peaks remaining in the structure. The major diffraction peaks at $2\theta = 28.4°$, 47.4° and 56.2° are presented at (111), (202) and (131) planes, respectively, which can be attributed to high-purity silicon gel according to JCPDS-card 96-901-3109. However, such peaks reduce their intensity when the heating ramp decrease. Through SEM analysis we can appreciate variations on the final morphology of the reduced species. In sample treated with ramp at 1°C/min the nodular morphology is well maintained, whilst in sample reduced at 50°C/min fused nodules can be observed. The electrochemical performance of the reduced silicon gels as anodes for lithium ion batteries have been also investigated by prolonged galvanostatic cycling vs. Li/Li^+.

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Figure 1. XRD data of the reduced silicon gels