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Toward understanding the real mechanical robustness of composite electrode impregnated with a liquid electrolyte

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ABSTRACT

The mechanical robustness of highly loaded composite electrodes is important for ensuring the long-term reliability of high-energy-density secondary batteries. Considering that in real state, the electrodes in batteries are completely impregnated with electrolyte, the swelling of the polymeric binder must be carefully observed and controlled to maintain the electric connectivity within the electrode. However, the decrease in the cohesion/adhesion of electrodes caused by electrolyte impregnation has not been directly measured due to the absence of appropriate tools. Here, the surface and interfacial cutting analysis system and a specifically designed sample holder are well combined to realize this breakthrough measurement. When electrode is impregnated with a liquid electrolyte, not only the 12% increase in electrode thickness but also the greater than 74% decrease in cohesion/adhesion, which is caused by the swelling of the amorphous phase of the polymeric binders, is clearly observed. The large decrease in cohesion/adhesion can be greatly ameliorated by controlling both the degree of crystallinity and crystallite size of the polymeric binder through a simple annealing process. Thus, it believes that the measurement of the real cohesion and adhesion of composite electrodes can provide an innovative and practical way to secure the reliability of high-energy-density batteries.

1. Introduction

To obtain secondary batteries with increased energy density but the same active material, it is essential to increase the electrode loading level (21 mg cm^{-2}) and lower the composition of inactive components such as polymeric binders and conducting materials[1–10]. However, as long-term reliability of secondary batteries is required and is as important as their initial performance, the mechanical robustness of composite electrodes under harsh operating conditions, which include wider voltage [11–15] and temperature ranges [16–20], higher current density [21–25], etc., has to be ensured., However, the fact that electrolyte impregnation into polymeric binders tends to weaken the mechanical robustness of composite electrodes has been overlooked so far owing to the absence of appropriate analysis tools. In other words, the real cohesion and adhesion of composite electrodes completely wet with the liquid electrolyte have not been measured reliably and systematically. Alternatively, this measurement is frequently conducted by comparing the surface or cross-sectional morphology of composite electrodes before and after dipping them in the liquid electrolyte. At most, the conventional peel test has been used to compare the adhesion or cohesion of composite electrodes before and after electrolyte swelling [26,27]. However, as is well known, the peel test can provide only the cohesion or adhesion of the weakest regions in composite electrodes [27–31]. Recently, Komaba et al. reported only the real adhesion of composite electrodes at the interface in Na-ion batteries in support of improving the adhesive capability of new binders [32]. However, since they focused on the development of new binder materials, a simple comparison was reported without in-depth insight into phase changes of the polymeric binders in the impregnated composite electrodes.

Therefore, without revealing the real cohesion and adhesion of composite electrodes impregnated with liquid electrolytes, it is almost impossible to determine the best binder candidates and their compositions in relation to liquid electrolyte type and operating conditions. Especially from this point of view, conventional polyvinylidene fluoride (PVdF)-type binders are easily swelled owing to the existence of an amorphous phase; [10,33,34] thus, quan-

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