## **Supporting Information**

In addition to the active material for a secondary rechargeable battery, we carried out supplementary experiments with a negative active material. The slurry for the anode electrode was prepared by employing graphite, styrene-butadiene rubber (SBR), and carboxymethyl cellulose (CMC) as the negative active material and binder compound, respectively. The average particle size of graphite was 15  $\mu$ m, and the composition ratio of the elements was 98:1:1. The constituents were mixed in deionized water. The resulting slurry was coated on an aluminum foil, and dried for 24 hr at 80 °C in a vacuum oven. After then, the sample was compressed with a hot presser. Also, the prepared slurry was freeze-dried for morphological analysis.

Figure S1 shows a sample in a plastic container right after finishing the freeze-drying experiment. It was verified that the slurry components were successfully immobilized during freeze-drying.



**Figure S1**. Sample image right after the freeze-drying experiment: (a) top view and (b) front view



Figure S2. FE-SEM image of the freeze-dried specimen: (a) 500x and (b) 1000x

Figure S2 presents the FE-SEM image of the freeze-dried slurry sample. Similar to the positive active material shown in Figure 6, the negative active material forms a network structure. Keep in mind that the purpose of this study is to analyze the internal structure of a multi-component slurry system in a liquid state before coating and compression processes for rechargeable secondary battery. The internal structure of the slurry, which is determined by the mixing process, significantly affects the quality of the following coating process. On the other hand, we also took into account more realistic battery preparation processes by coating and compressing the slurry sample for an anode electrode on the aluminum foil. Figure S3 displays the image of the slurry sample after the coating and compressing procedures. It is found that the internal structures shown in Figure 2S and 3S are quite different. Therefore, we may need to further investigate both the coating and compressing processes in a bid to understand the change in the internal structure of the slurry components.



**Figure S3**. FE-SEM image of the specimen compressed on the aluminum foil: (a) 500x and (b) 1000x