Supplementary Information

Tantalum oxide nanomesh as one-nanometre-thick electrolyte

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First, the TaO_3 nanosheets were deposited on the surface of a silicon wafer where they could be easily seen. The suspensions with different nanosheet concentrations were spread over the Si wafers under the conditions described in the main text. The deposited nanosheets were observed using an atomic force microscope (SPA400 AFM system, Seiko Instruments). Fig. S1a–c shows the AFM images of the samples prepared from the suspensions with TaO_3-nanosheet-concentrations of 0.4, 0.8 and 1.6 g·dm^{-3}, respectively. Although thicker crystallites, which would be unexfoliated particles, are found in the images, there is no significant overlapping of the nanosheets. The images were converted into the histograms of relative height, as shown in Fig. S1d, to determine the coverage of the nanosheets. Each histogram is composed of two peaks. When the peak corresponding to the uncovered area is set at the relative height of 0 nm, the other peak is located at *ca.* 1.0 nm, which agrees well with the thickness of the TaO_3 nanosheet. Therefore, it can be concluded that the peak at 1.0 nm comes from the region covered with a monolayer of TaO_3 nanosheets. From the areas of the peaks, the coverages are found to be 20%, 40% and 60% for the samples prepared from the suspensions with the concentrations of 0.4, 0.8 and 1.6 g·dm^{-3}, respectively. When the TiO_2 nanosheets were used in place of the TaO_3 nanosheets, the coverages for the suspensions with the TiO_2-nanosheet-concentrations of 0.2 and 0.4 g·dm^{-3} are 25% and 50%, respectively. The coverage is unchanged even on the $LiCoO_2$ thin films as confirmed by two kinds of microscopy, conducting AFM and low-voltage scanning electron microscopy (SEM), because the surface of the films deposited on polycrystalline Pt plates was not flat enough to clearly observe the nanosheets in conventional AFM images.

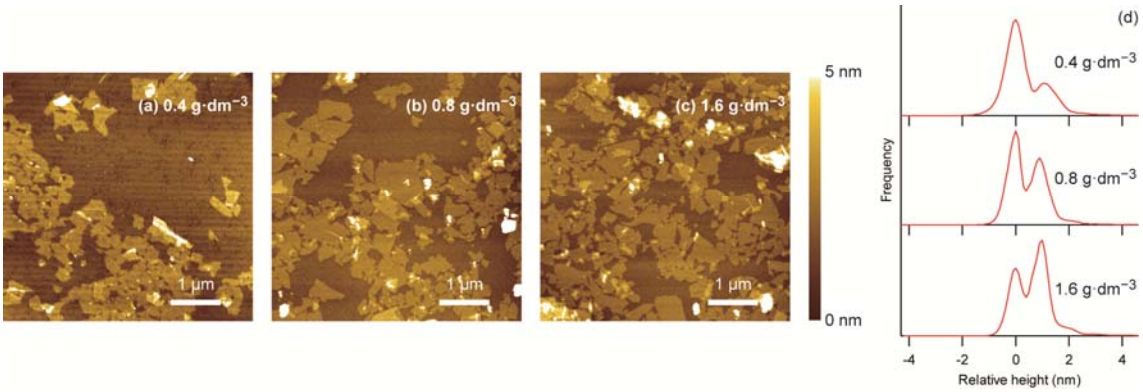

Figure S1. AFM images of TaO_3 nanosheets deposited on Si wafers by spin-coating method (a)–(c). Concentrations of the nanosheets in the suspensions used in the spin-coating are indicated in the images. The images were converted to the histograms of relative height in (d), in which the first peaks were positioned at a relative height of 0 nm.

Since the nanosheets are insulating for electronic conduction due to the wide band gap, they can be detected as a change in resistance under conducting AFM. Conducting AFM measurements were carried out using an SII Nanotechnology E-Sweep microscope in a vacuum under a base pressure of less than 1×10^{-6} Torr to avoid contamination. We used a conductive 0.1–0.2 $N\cdot m^{-1}$ silicon cantilever coated with a Pt overlayer to measure I–V curves. The tip radius of the cantilever was about 30 nm. The applied load was within 1–2 nN. We measured a current image simultaneously with a topographical image, by changing the applied voltage. In the conducting AFM images, the nanosheets stand out as areas where very small current flows, as shown in Fig. 2 in the main text.

We also confirmed the unchanged coverage by scanning electron microscopy (SEM). MULTI SCAN LAB with UHV GEMINI system (Omicron Nanotechnology, Germany) was used as the scanning electron microscope[S1] and operated at low primary electron energy of 200 eV so as to prevent electrons from penetrating the nanosheets. Fig. S2 shows a typical micrograph taken for one of the samples. The nanosheets, seen as dark pieces in the images, are found to be uniformly spread over the $LiCoO_2$ surface. These images indicate that the coverage on the $LiCoO_2$ films is almost the same as that observed on the Si wafers.

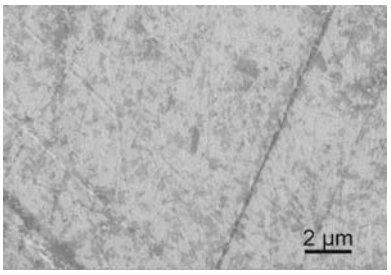

Figure S2. A low-voltage SEM image of a $LiCoO_2$ thin film with its surface covered with TaO_3 nanosheets. The nanosheet concentration of the suspension for preparing the sample was 0.8 g·dm^{-3}.

References

S1 K. Kumagai and T. Sekiguchi, *Ultramicroscopy*, 2009, **109**, 368–372.