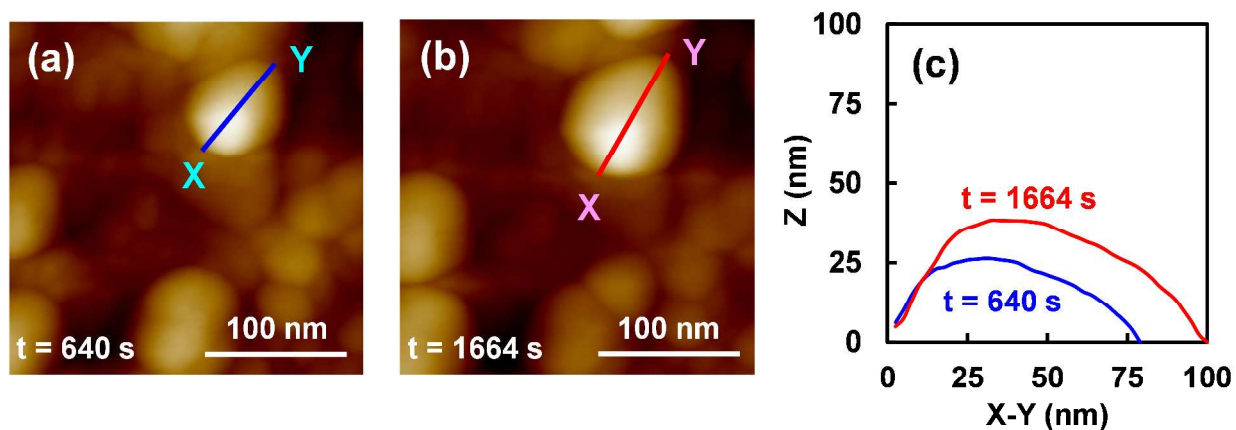


Supporting Information of

Real-time observation of Li deposition on a Li  
electrode with operant atomic force microscopy and  
surface mechanical imaging

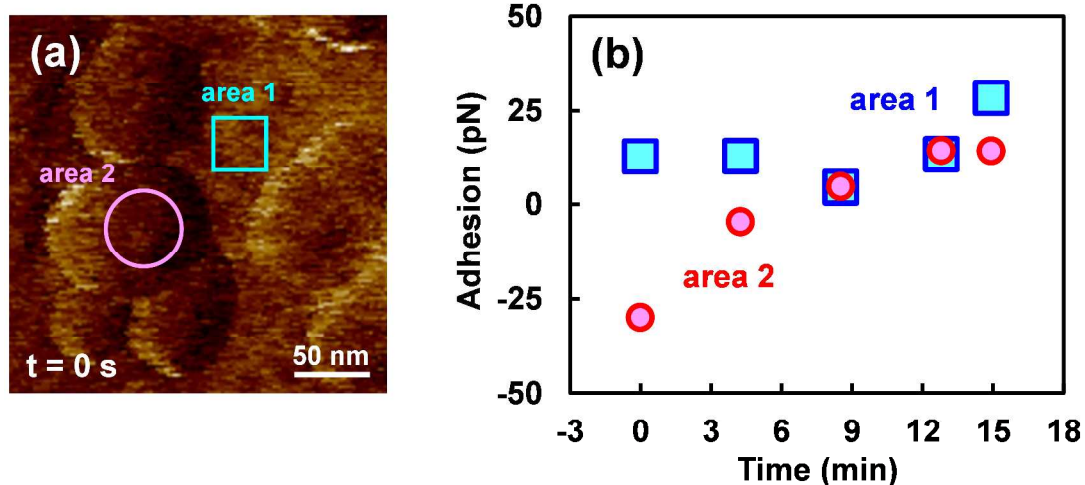
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**Figure S1**

Line profiles of a deposited Li object for two different deposition times. (a) and (b) show the deposited Li protrusion at  $t = 640$  s and  $t = 1664$  s, respectively. The corresponding line profiles are presented in figure (c). As can be seen in the line profiles, the deposited Li object can be considered as a hemisphere at both times. Consequently, we estimate the volumes of the deposited Li objects using a hemisphere model, i.e.,  $V = 2/3\pi r^3$ .



**Figure S2**

Time alternation of adhesion force on the deposited Li surface with OCP condition. (a) Area of adhesion analysis. The original pristine Li surface is marked with the blue rectangle and the surface of deposited Li is denoted with a purple circle. The time evolution of the adhesion forces of each analyzed area is plotted on figure (b) (area 1 with blue rectangles and area 2 with red circles, respectively). It can be clearly seen that the adhesion forces at the deposited Li surface increase with time, while the adhesion forces for the original Li surface remain almost constant. The adhesion forces at the surface of deposited Li seem to be saturated after around 15 min and the values approach an adhesion force value near to the value of original pristine Li surface. Usually the electrolyte is decomposed and forms a surface film (which is intrinsically linked to the mapped adhesion forces) on the Li surface due to its high reduction potential. Once the surface film has formed, it will prevent the further decomposition of electrolyte and, this way eventually lead to a

saturated thickness of the surface film, which in turn explains a saturation of the related adhesion forces.