

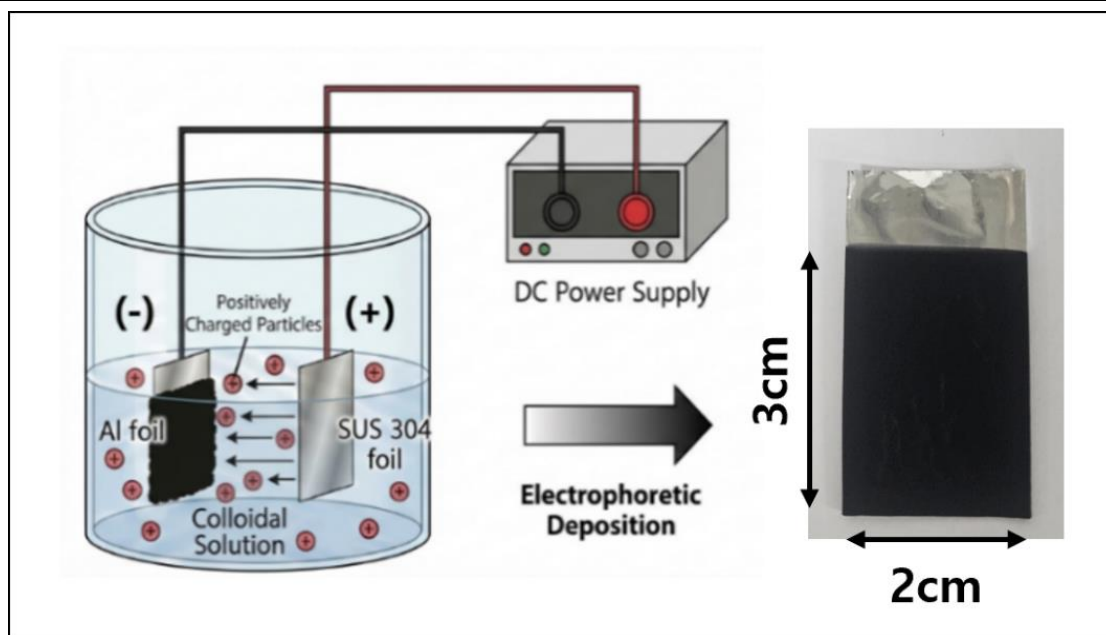


Figure S1. Schematic illustration of the cathodic electrophoretic deposition (EPD) process and an optical photograph of the resulting as-deposited electrode. The schematic depicts the migration of positively charged colloidal particles toward the Al foil (working electrode) under an applied DC electric field. The optical image on the right demonstrates the successful fabrication of a macroscopically uniform composite electrode with a defined area of 2 cm × 3 cm.

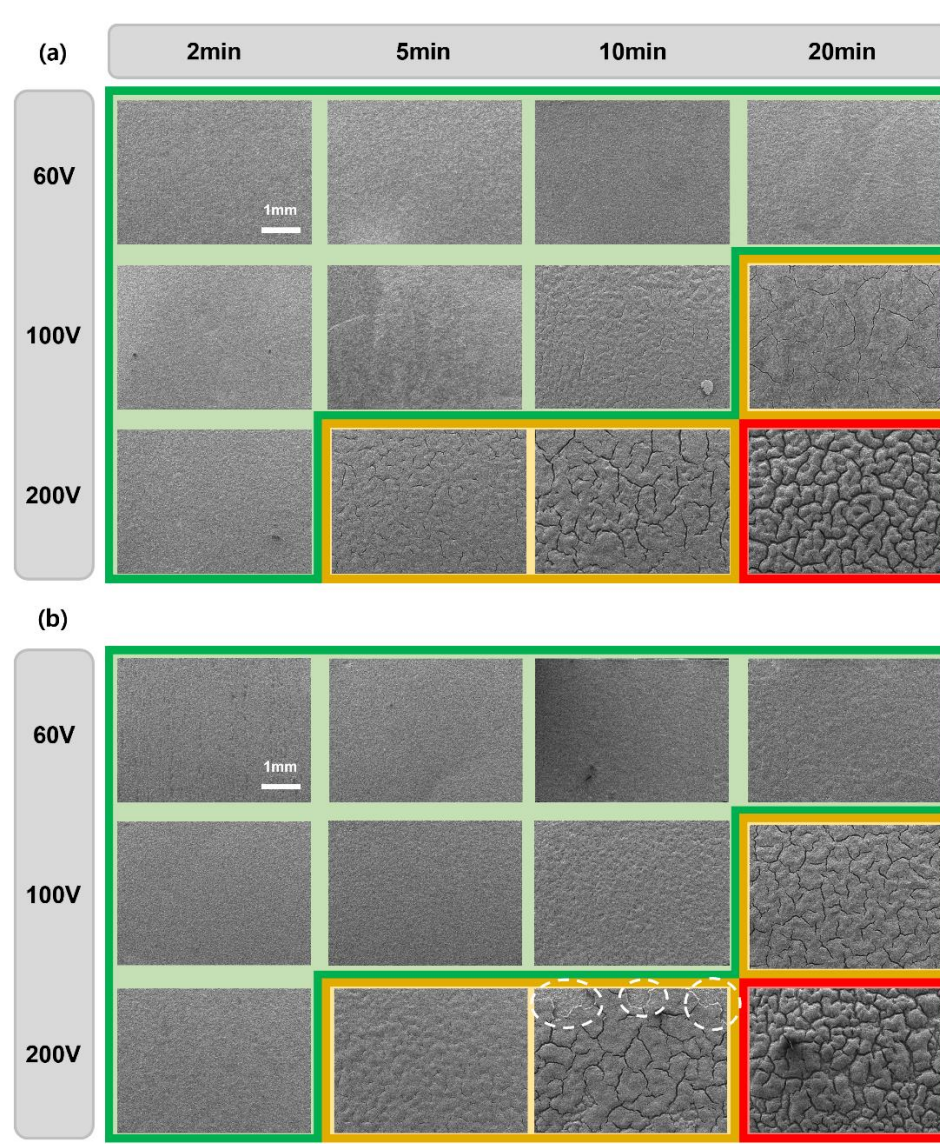


Figure S2. Top-view SEM images demonstrating the morphological evolution of the (a) Ti-0 and (b) Ti-1 electrodes under various EPD conditions (applied voltage: 60–200 V, deposition time: 2–20 min). The colored borders categorize the structural integrity of the electrodes based on the mass loading: uniform and stable surfaces without visible defects (green region), the onset of surface micro-cracks (yellow region), and significant structural degradation including severe cracks and macroscopic delamination (red region). In particular, the specific areas where delamination occurred are highlighted by white dotted circles. Consistent with the trend described in Figure 2, these results indicate that excessive applied voltages and prolonged deposition times lead to a critical film thickness, triggering mechanical failure in the EPD electrodes.

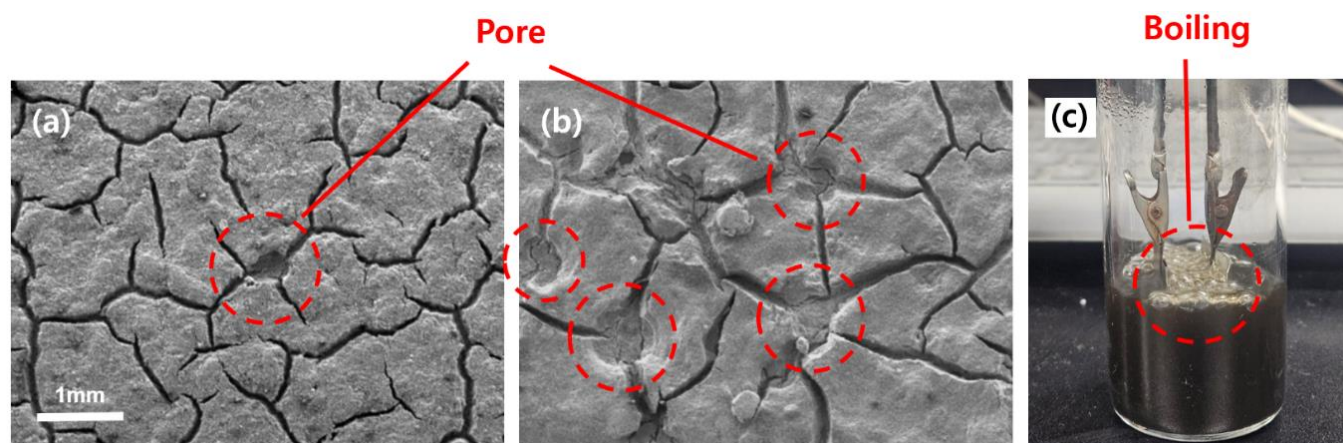


Figure S3. Identification of EPD process limitations and morphological defects formed under extreme conditions. (a, b) Top-view Scanning Electron Microscope (SEM) images of the electrode surface after a prolonged deposition time of 30 min (200 V). Red dotted circles indicate surface pores formed typically by gas evolution at the electrodes. Severe surface cracking is also visible due to excessive thickness. (c) Digital photograph demonstrating solvent boiling within the EPD cell when a high voltage (300 V, 5 min) is applied. The boiling phenomenon, highlighted by the red dotted circle, is induced by severe Joule heating due to high current density, defining the upper limit of the applied voltage for a stable EPD process.

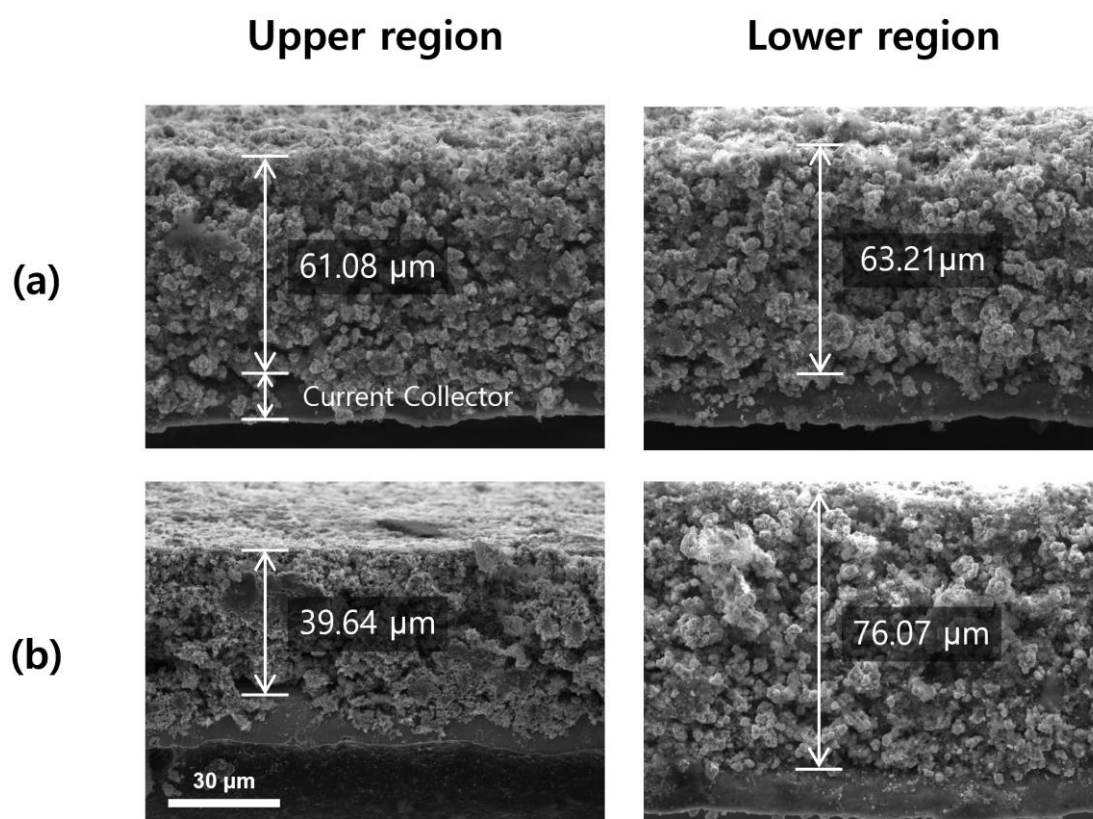


Figure S4. Cross-sectional SEM comparison of EPD electrodes prepared (a) without and (b) with the intermediate pause-and-stirring procedure during a total deposition time of 10 min. The electrode deposited without intermediate stirring shows more pronounced thickness variation, whereas the electrode prepared with intermediate stirring exhibits a more uniform coating layer without a clear interlayer boundary.

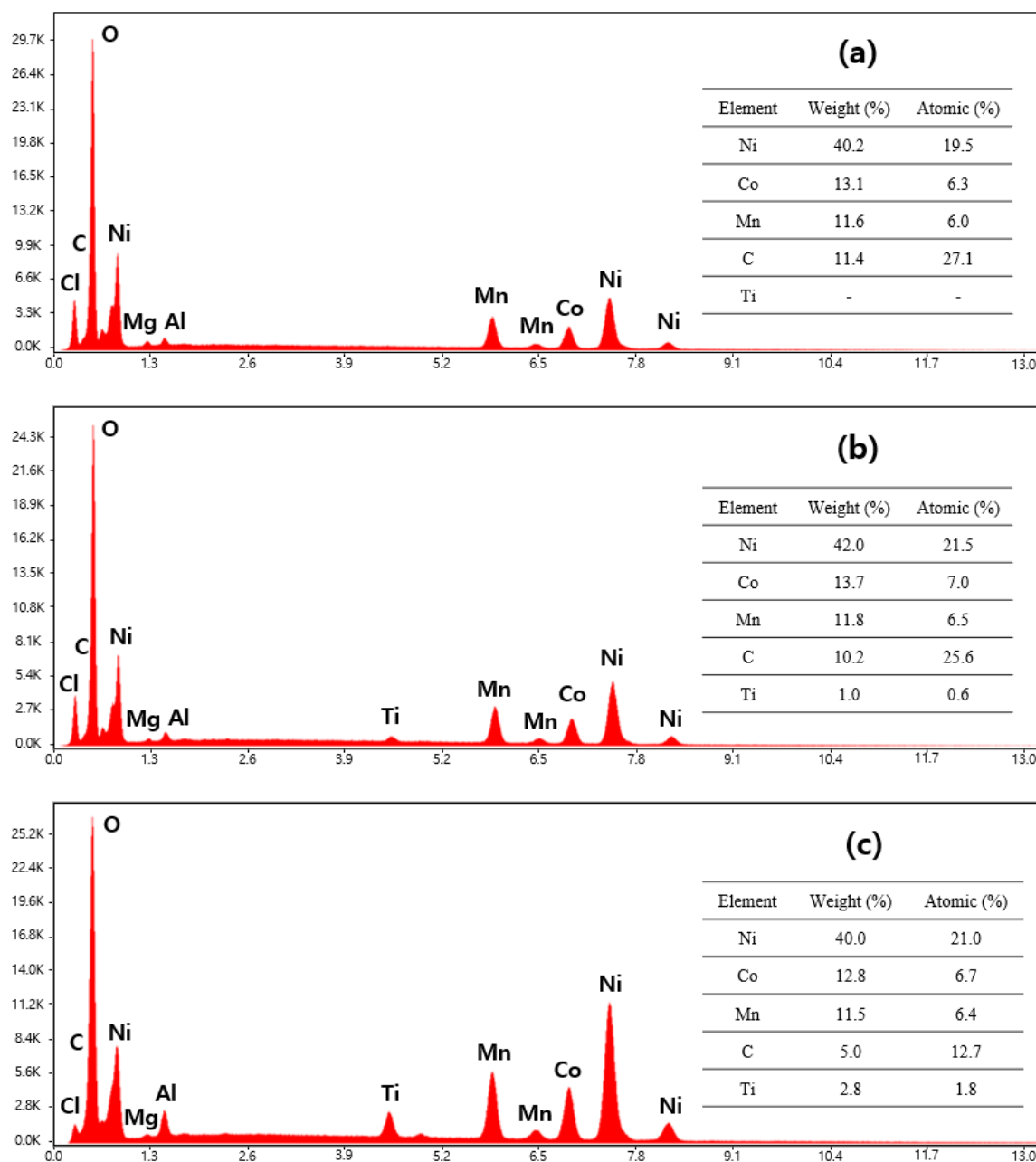


Figure S5. Energy-dispersive X-ray spectroscopy (EDS) spectra and the corresponding quantitative elemental analysis tables for the EPD electrodes prepared with different initial suspension formulations: (a) Ti-0, (b) Ti-1, and (c) Ti-5. The characteristic peaks indicate the presence of elements associated with the co-deposited NCM622 active material (Ni, Co, Mn) and conductive additives (Ti, C) within the final electrodes, and the compositional ratio (wt%) of each element was determined from the quantitative EDS analysis. Based on these quantitative elemental analyses, the phase ratios of NCM622:Carbon:Ti₄O₇ in the deposited electrodes were estimated as 90.5:9.5:0 for Ti-0, 90.6:8.2:1.2 for Ti-1, and 91.9:4.3:3.8 for Ti-5. Note that the minor peaks of Al, Mg, and Cl are attributed to the underlying aluminum foil current collector and trace residues from the EPD suspension additives, respectively.

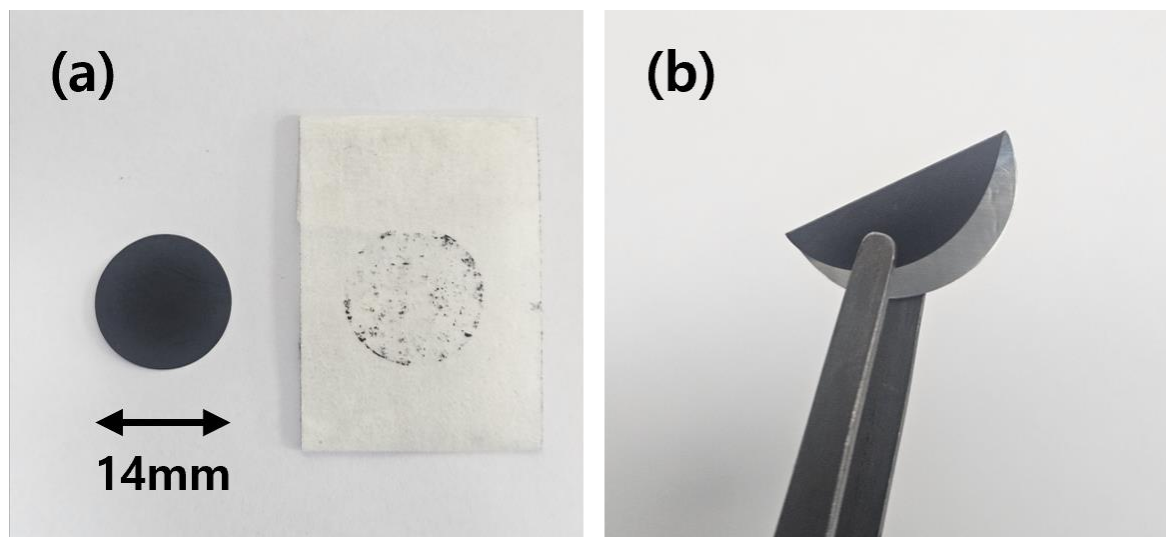


Figure S6. Macroscopic mechanical stability and flexibility of the prepared EPD electrode. (a) A 3M tape peeling test performed on a punched electrode (14 mm in diameter), confirming strong interfacial adhesion to the Al substrate with minimal active material detachment. (b) A 180° bending test using tweezers, demonstrating excellent mechanical flexibility without any visible surface cracks or delamination.