

## SUPPLEMENTARY INFORMATION

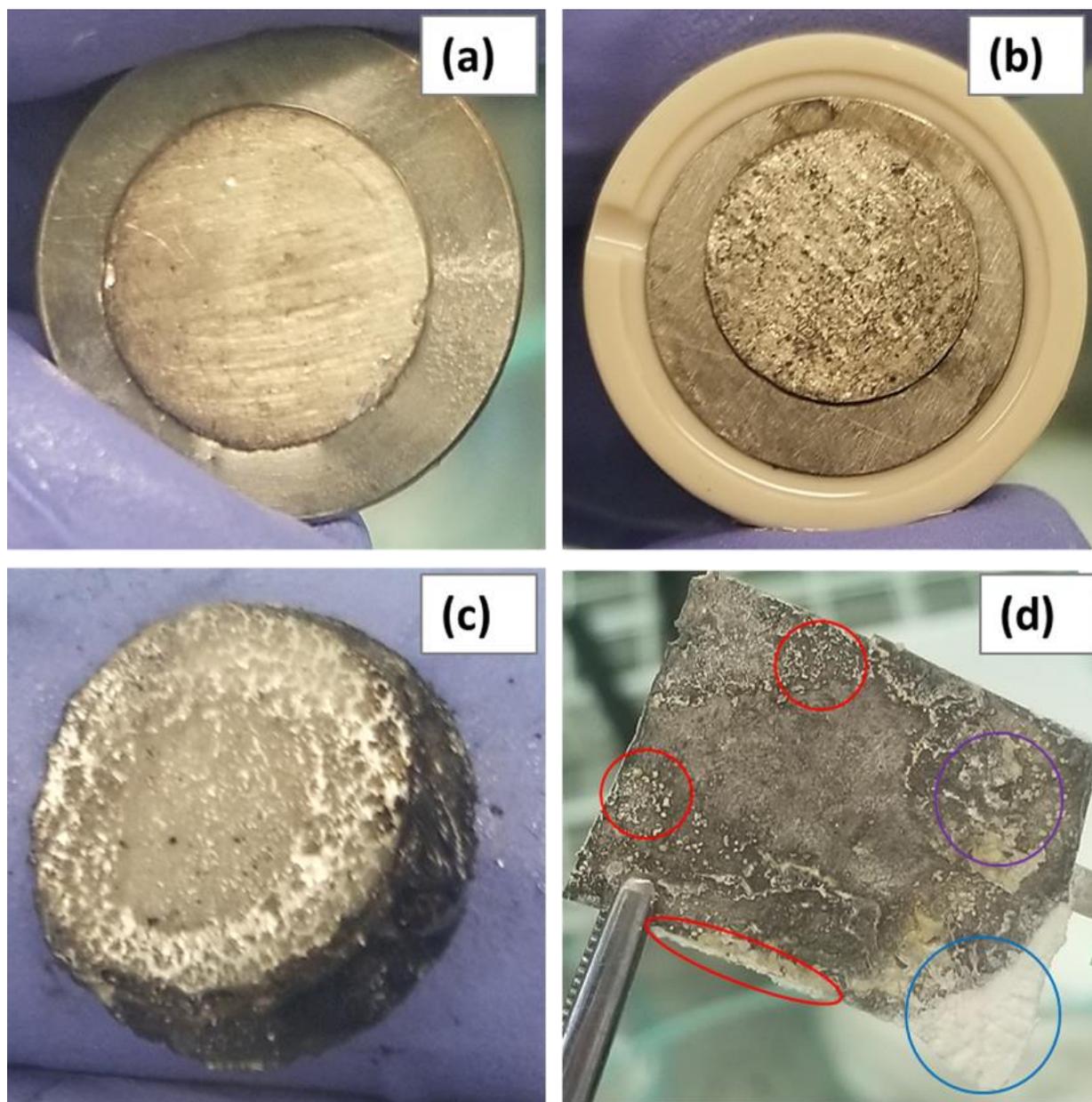


Figure S1: Images of representative cycled Li metal anodes from the different cycling conditions. (a) 4.0V+LT, (b) 4.3V+LT, (c) 4.0V+HT, (d) 4.3V+HT. Presence of lithium plating in all conditions, and combination of dead Li possibly forming dendrites (red circles), thick Li plating regions (purple regions), and white regions from decomposed electrolyte salts (purple circle) are observable for 4.3V+HT.

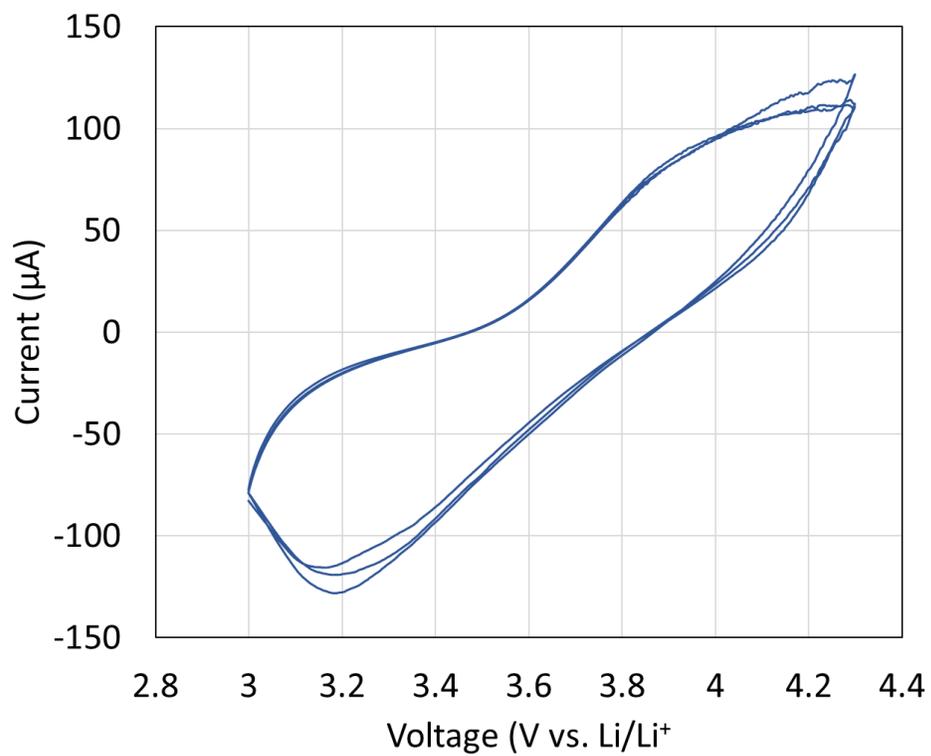


Figure S2: Cyclic voltammogram for NMC811 cathode cycled at 4.3 V vs Li/Li<sup>+</sup> and 25 °C

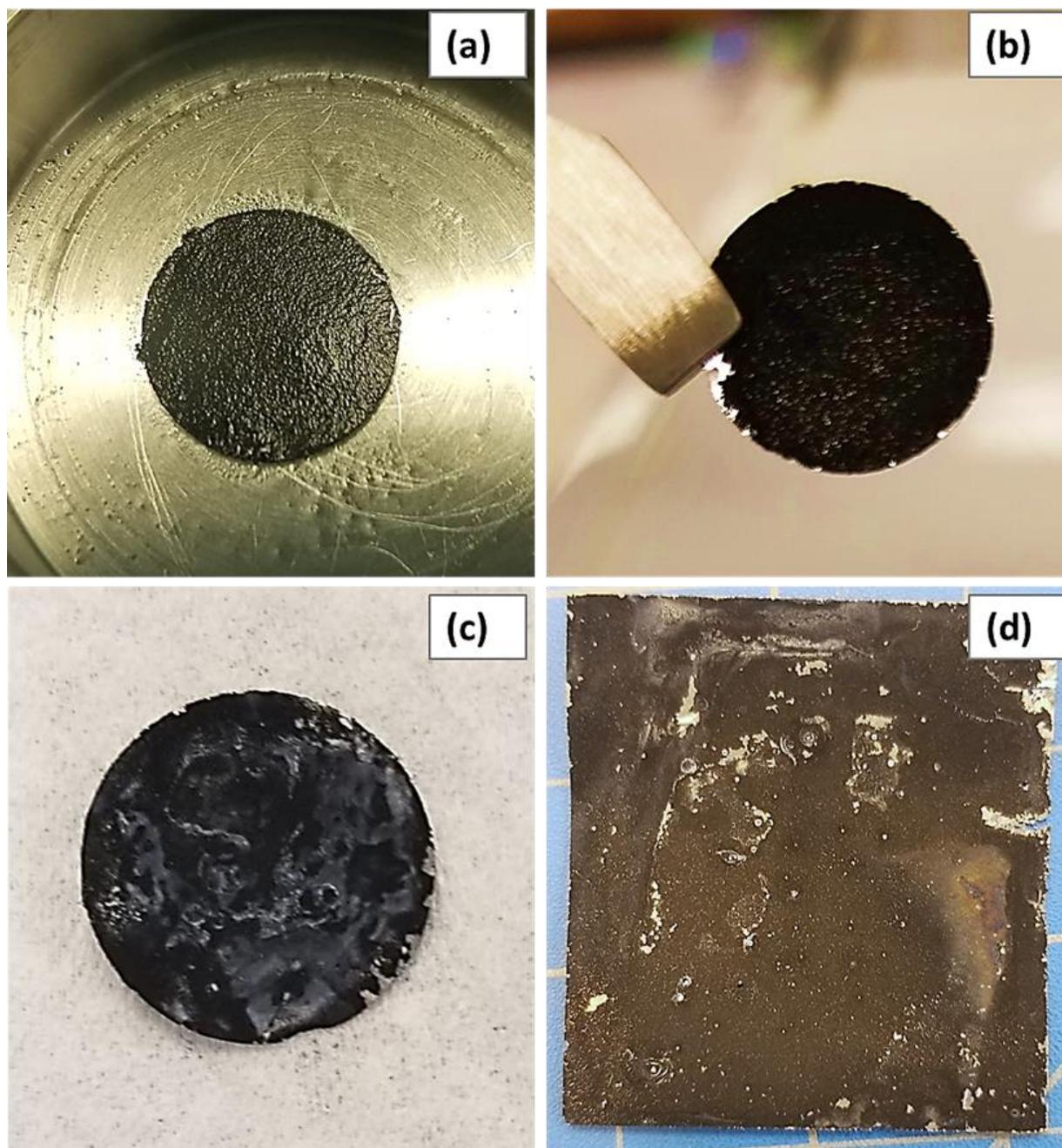


Figure S3: Images of representative cycled NMC811 cathodes from the different cycling conditions. (a) 4.0V+LT, (b) 4.3V+LT, (c) 4.0V+HT, (d) 4.3V+HT. Cathode cycled under LT conditions show little to no changes in their surfaces. Heavy presence of electrolyte-salt decomposition is present in both HT cathodes, with additional burn out regions for NMC cycled at 4.3V+HT.

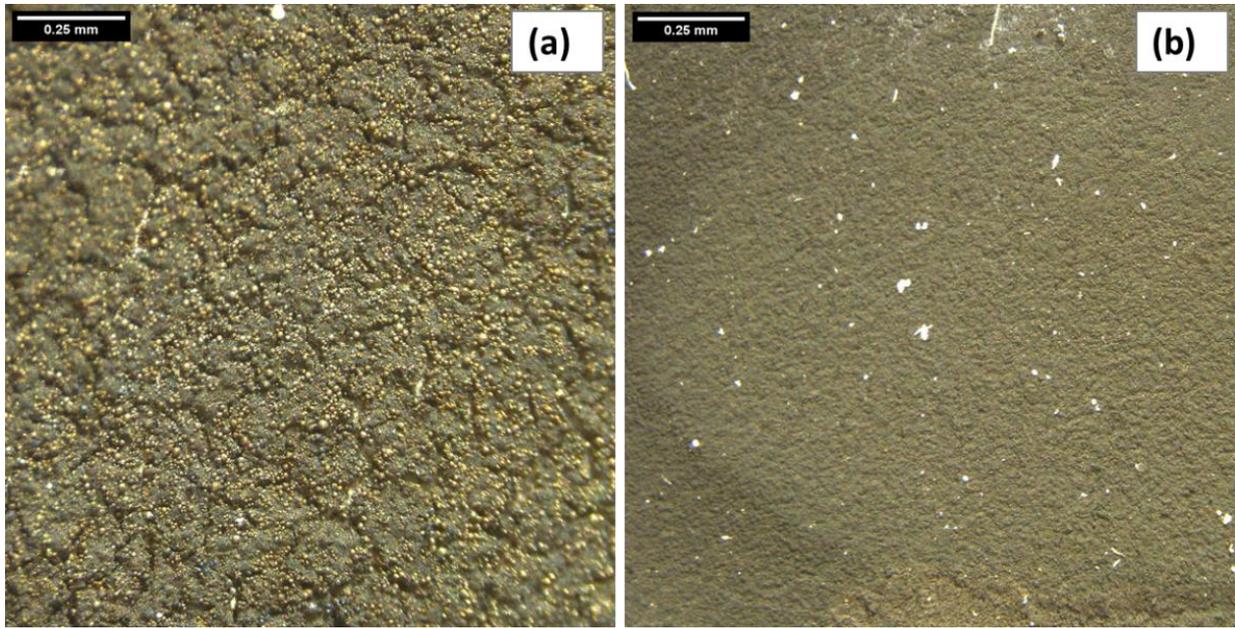


Figure S4: Images taken at 4x magnification for (a) 4.0V+LT and (b) 4.3V+LT, showing slight changes in their surfaces.

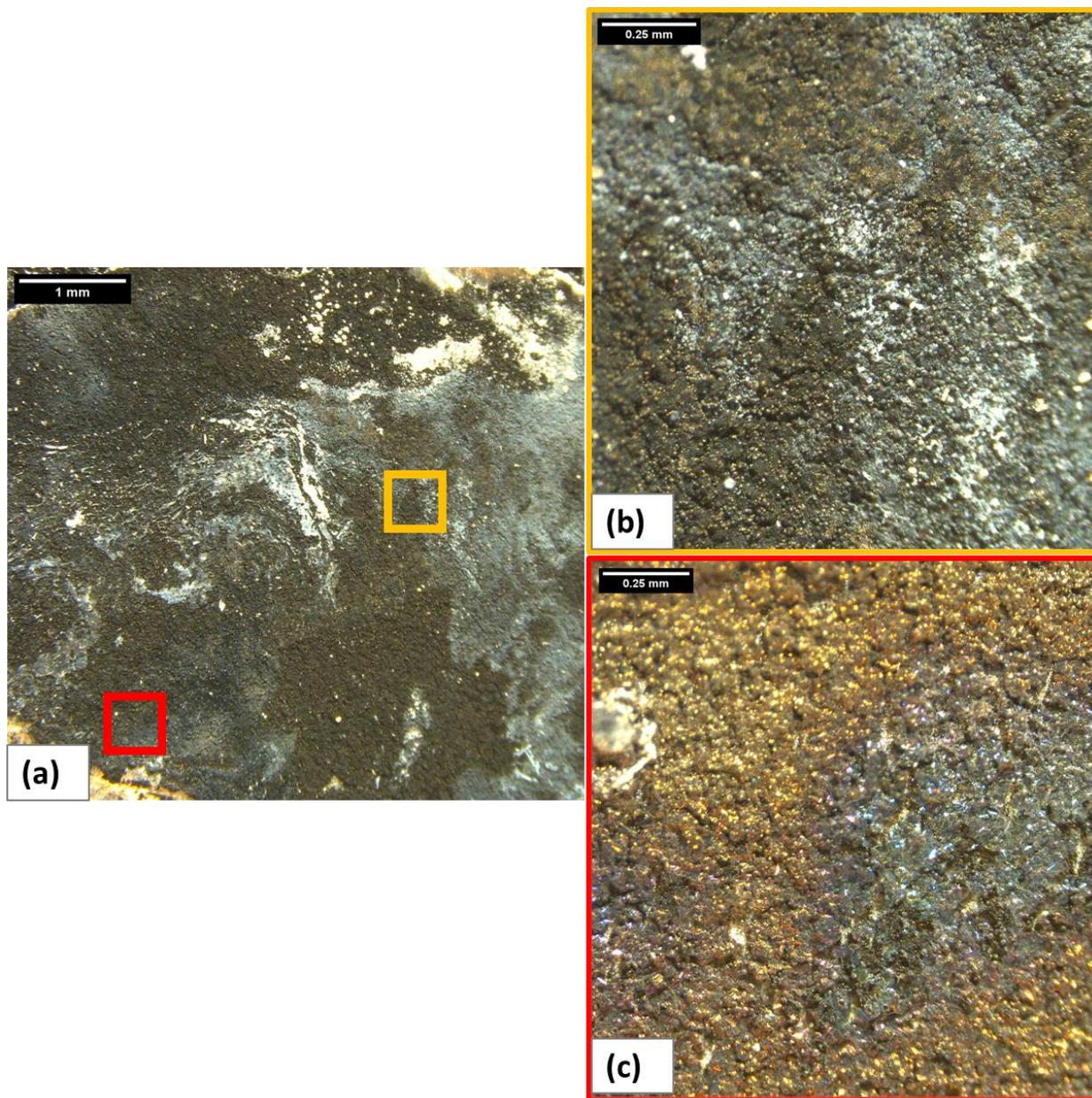


Figure S5: Images from a 4.3V+HT cycled cathode at 1x and 4x magnification levels. (a) Shows the x1 magnification of the general view of the highly degraded surface with two subsections of interest enclosed in orange and red squares. (a) Heavy electrolyte-salt formation regions and (b) decolored sections of the electrode, both features contributing to the extreme decay of the battery under this condition.

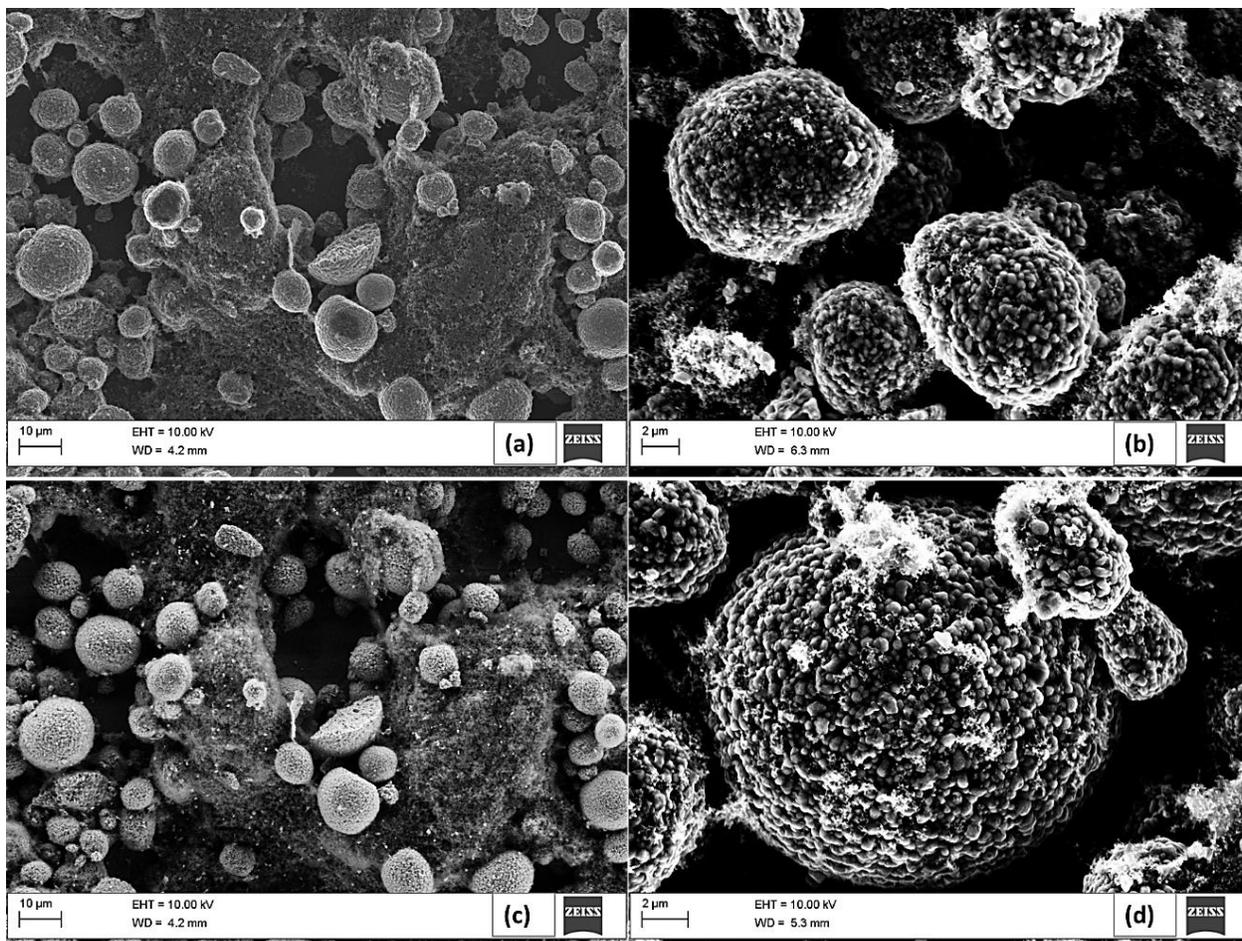


Figure S6: SEM images from NMC811 pristine microstructure at 10kV. Lower magnification images are shown with (a) SED and (c) BS. Highest magnification images with SED are shown for (b) groups of particles and (d) single particles. Spherical secondary particles, supporting phase, and principal granular particles are observable.