Characterization of Surface Free Energy of Composite Electrodes for Lithium-Ion Batteries

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One critical step in lithium-ion battery manufacturing process is the electrolyte wetting step, which usually takes up to several days at elevated temperatures, posing a distinctive bottleneck in the manufacturing process especially for large-size cells. The electrode wettability depends on both the surface tension of electrolyte and the surface free energy of electrodes. This work reports a systematic study on the characterization of surface free energy of battery electrodes and the correlation between surface free energy and electrode processing. It shows that, when switching manufacturing from conventional solvent based processing to aqueous processing, the polar component of surface free energy increases 74.1% for NMC532 electrodes and 48.2% for A12 graphite electrodes. The results provide valuable guidance for electrode processing to optimize the electrolyte wetting process, helping lower processing costs and enable higher product quality and throughput.

Theory

Ideal surface.—The wettability of an ideal surface is quantified by the well-known Young’s equation:

\[ \gamma_{lv} \cos \theta = \gamma_{lv} - \gamma_{li} \]  

where \( \theta \) is the contact angle, i.e., the angle at which the liquid–vapor interface meets the solid-liquid interface, \( \gamma_{lv} \) is the solid/liquid interfacial free energy, \( \gamma_{li} \) is liquid/vapor interfacial tension, i.e., surface tension of the liquid, and \( \gamma_{lv} \) is the SFE of the solid. We here assume that the SFE of the surface is equal to the surface tension in equilibrium with the vapors of the probe liquid. In other words, the film pressure term responsible for molecular absorption onto the surface is assumed to be negligible.

A low contact angle usually indicates that wetting of the surface is favorable, and the fluid will spread over a large area of the surface, whereas a high contact angle generally means that wetting of the surface is unfavorable, so the fluid will reduce contact area with the surface by producing a compact liquid droplet. For water, a wettable surface is referred to as hydrophilic and a nonwettable surface hydrophobic. For nonwater liquids, the two terms lyophilic and lyophobic are used for low and high contact angle conditions, respectively.

It can be seen from Eq. 1 that the electrode wettability depends on both the surface tension of electrolyte and the SFE of the electrodes. SFE is a property resulted from the chemical structure and the orientation of the molecules at the surface boundary of the materials.

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The SFE value of a material can be used to predict whether or not the material is wettable by a certain liquid: solids with similar or larger SFE than the surface tension of a liquid are wettable by that liquid. Therefore, having higher SFE in electrodes than the surface tension of electrolyte is essential to optimize the electrolyte wetting process.

The characterization of SFE is not straightforward even for an ideal surface; in spite of decades of struggle, actual formulation of SFE and its determination are still problematic. Several semi-empirical analytical models have been developed to compute the SFE, e.g., the Zisman model, the Saito model, the geometric and harmonic mean approximation, the Berthelot model, the Fowkes model, acid-based approximation, and the OWRK model. All these methods are based on contact angle measurements and each method has its own limitations. The OWRK model originally proposed by Owens and Wendt is adopted in this study. It is a standard method for calculating the SFE of a solid from the contact angle with several liquids, which has been applied to calculate SFE for various surfaces, including polymeric surfaces, wood surfaces, molybdenum nitride coatings, niobium nitride coatings, diamond-like carbon coatings, and 2D heterostructures, such as graphene, molybdenum disulfide, and tungsten disulfide. The method is based on two fundamental assumptions.

The cohesion between the atoms and molecules that is responsible for the SFE of a solid can be categorized into dispersive interactions and polar interactions. Interactions caused by random fluctuations of the charge distribution in atoms and molecules are called dispersive interactions. Polar interactions comprise coulomb interactions between permanent dipoles and between permanent and induced dipoles such as hydrogen bonds. According to the OWRK model, the first fundamental assumption is that the SFE of a solid is the sum of the two independent contributions, where the superscript D and P represent the dispersive and polar components, respectively. The second fundamental assumption is that the Berthelot’s combining rule applies, i.e.,

$$\gamma_{sv} = \gamma_{sv}^{D} + \gamma_{sv}^{P}$$  \[2\]

where

$$W_{ld} = \sqrt{W_{ld}^{D}W_{ld}^{D} + W_{ld}^{P}W_{ld}^{P}}$$  \[3\]

where

$$W_{ld} = \gamma_{lv} + \gamma_{sv} - \gamma_{sv}^{*}$$ is the work of adhesion, and

$$W_{sv} = 2\gamma_{sv}^{*}$$

are the work of cohesion for the solid and liquid, respectively. Substituting the Young-Dupré equation for work of adhesion, i.e., $\gamma_{lv} = \gamma_{lv}(1 + \cos \theta)$, into Eq. 3 yields:

$$\frac{1}{2}\gamma_{lv}(1 + \cos \theta) = \sqrt{\gamma_{lv}^{D}\gamma_{lv}^{D} + \gamma_{lv}^{P}\gamma_{lv}^{P}}$$  \[4\]

Dividing Eq. 4 by $\sqrt{\gamma_{lv}^{D}}$ on both sides yields:

$$\sqrt{\gamma_{lv}^{D}} + \sqrt{\gamma_{lv}^{P}} \frac{\sqrt{\gamma_{lv}^{D}}}{\sqrt{\gamma_{lv}^{P}}} = \frac{\gamma_{lv}(1 + \cos \theta)}{2\sqrt{\gamma_{lv}^{D}}}$$  \[5\]

where

$$b + ax = y$$  \[6\]

where

$$b = \sqrt{\gamma_{sv}^{D}}, \quad a = \sqrt{\gamma_{sv}^{P}}, \quad x = \frac{\sqrt{\gamma_{sv}^{D}}}{\sqrt{\gamma_{sv}^{P}}}, \quad y = \frac{\gamma_{sv}(1 + \cos \theta)}{2\sqrt{\gamma_{sv}^{D}}}$$  \[7\]

Typically, by using two different liquids for one solid surface, a system of two linear equations can be established to solve for a and b, from which the SFE of the solid can be calculated as $\gamma_{sv} = a^{2} + b^{2}$. Therefore, at least two liquids with known dispersive and polar components are required to determine the SFE of a solid. The liquids used for such a purpose are often referred to as probe liquids. In practical applications, using more than two probe liquids is very common to obtain more reliable results. In this work, propylene carbonate, nicotine alcohol, and dimethyl sulfoxide were used as probe liquids, and their contact angles on various electrode samples were measured.

versus y were calculated as data points for each electrode sample, and the best-fit least-square regression lines were plotted.

While the OWRK model is a two-component model for SFE, it is also a two-component model for liquid surface tension. Both polar and dispersive components of the surface tension of each probe liquid must be determined. This is often achieved by using a standard reference surface. In this study, a standard polytetrafluoroethylene (PTFE) plate was used as a reference surface. Pure untreated PTFE has a surface energy of 18.0 mJ/m², and is assumed to be incapable of polar interactions, i.e., for PTFE, $\gamma_{sv} = \gamma_{sv}^{D} = 18.0$ mJ/m², and $\gamma_{sv}^{*} = 0$ mJ/m². Substituting these values into Eq. 5, followed by rearrangement yields:

$$\gamma_{sv}^{D} = \gamma_{sv}^{D}(1 + \cos \theta_{PTFE})^{2} \frac{1}{72}$$  \[8\]

where $\theta_{PTFE}$ is the contact angle measured between PTFE and the probe liquid. Therefore, simply by measuring $\theta_{PTFE}$, $\gamma_{sv}^{D}$ can be determined for any liquid for which the overall surface tension $\gamma_{sv}$ is known. The value of $\gamma_{sv}$ can then be determined by using $\gamma_{sv}^{D} = \gamma_{sv} - \gamma_{sv}^{D}$.

Non-ideal surface.—The static contact angle $\theta_{s}$ is the contact angle with which the contact area between liquid and solid is not changed during the measurement, in contrast to the dynamic contact angle which is produced in the course of wetting (advancing contact angle) or de-wetting (receding contact angle). Dynamic contact angles depend on the speed of the moving contact line. In the limiting case where the advancing or receding speed approaches zero, the static advancing contact angle ($\theta_{a}$) and the static receding contact angle ($\theta_{r}$) are defined. For an ideal surface, the static contact angle, the static advancing contact angle, and the static receding contact angle are all equal, and can be described by the Young’s equation.

However, real surfaces do not have perfect smoothness, rigidity, or chemical homogeneity. For example, the surface of the electrode samples deviates from ideality because it always has a certain degree of roughness, porosity, and heterogeneity. Such deviations from ideality result in contact-angle hysteresis, i.e., a non-zero difference between the advancing and receding contact angles:

$$\Delta \theta_{AR} = \theta_{a} - \theta_{r}$$  \[9\]

Contact-angle hysteresis occurs because many different thermodynamically stable contact angles can be found on a non-ideal solid. Deviations of solid surfaces from ideality result in an apparent contact angle $\theta^{\ast}$ differing from the intrinsic Young’s contact angle $\theta$. The Wenzel and Cassie–Baxter models are the two main models used to describe wetting behavior on rough surfaces. The two models apply when the drop size is sufficiently large compared with the surface roughness scale. The Wenzel model describes the homogeneous wetting regime, as shown in Fig. S1a and Fig. S1b in Supplementary Information, when the liquid completely penetrates into the grooves. The apparent contact angle $\theta^{\ast}$ is given by the following equation:

$$\cos \theta^{\ast} = r \cos \theta$$  \[10\]

where the roughness ratio $r$ is defined as the true surface area divided by the projected surface area, and $\theta$ is the Young’s contact angle as defined for an ideal surface.

With increasing surface roughness, vapor pockets may be trapped underneath the liquid yielding a composite interface. This heterogeneous wetting regime is usually described by the Cassie–Baxter model. For a two-component system, the apparent contact angle $\theta^{\ast}$ is given by:

$$\cos \theta^{\ast} = f_1 \cos \theta_1 + f_2 \cos \theta_2$$  \[11\]

where $f_1$ and $f_2$ are the area fractions of material 1 and 2, respectively. Note that $f_1 + f_2 = 1$. The two angles $\theta_1$ and $\theta_2$ are the Young’s contact angles of pure materials 1 and 2, respectively.

If the voids of a rough surface are filled with liquid, as shown in Fig. S1c in Supplementary Information, then $\theta_2 = \theta^{\ast}$ and Eq. 11 can
be written as:

\[
\cos \theta_a = f(\cos \theta - 1) + 1 \tag{12}
\]

where the subscript of \( f \) is dropped for simplicity. If air is assumed to be entrapped in the voids of a rough surface, as shown in Fig. S1d in Supplementary Information, then \( \theta_2 = 180^\circ \) and Eq. 11 can be written as:

\[
\cos \theta_2 = f(\cos \theta + 1) - 1 \tag{13}
\]

where again the subscript of \( f \) is dropped for simplicity. According to Bico et al., 31–35 two critical contact angles \( \theta_{1\alpha} \) and \( \theta_{2\alpha} \) can be defined as follows:

\[
\cos \theta_{1\alpha} = \frac{1 - f}{r - f} \tag{14a}
\]

\[
\cos \theta_{2\alpha} = \frac{f - 1}{r - f} \tag{14b}
\]

For rough surfaces, we generally have \( r > 1 \) and \( f < 1 \), so Eq. 14a always defines a critical contact angle between 0 and \( \pi/2 \), whereas Eq. 14b always defines a critical contact angle between \( \pi/2 \) and \( \pi \). Based on Eq. 10, Eq. 12, and Eq. 13, the relationship between \( \cos \theta^0 \) and \( \cos \theta \) is plotted in Fig. S1e in Supplementary Information. Two parameter values \( r = 1.9 \) and \( f = 0.4 \) are assumed to generate the plot.

When trying to assess the SFE value of a real surface, many researchers support the idea of using the advancing contact angle \( \theta_a \) as the apparent contact angle \( \theta^0 \) 31–35 while others use the static contact angle \( \theta_s \). 36 The receding contact angle \( \theta_r \) is used much less frequently. Attempts were also made to extract a single, unique value by using various averaging methods so that the averaged angle can represent the apparent contact angle. 37,38 For example, Andrieu et al. suggested that a cosine average of \( \theta_s \) and \( \theta_r \) was the most stable contact angle for SFE calculation. 38 Although their experimental data supported this method, all their experiments were performed on samples with very small hysteresis, i.e., \( \Delta \theta_{rk} \leq 10^\circ \). Due to the highly porous nature of the electrode surfaces, the hysteresis will be too large for any averaging method to be appropriate. 39 Thus, in this study, the advancing contact angle \( \theta_a \) will be used as the apparent contact angle \( \theta^0 \) for the SFE calculation.

**Experimental**

**Electrode fabrication.—**Materials used in electrode fabrication are listed in Table S1 in Supplementary Information. Four different types of electrodes were fabricated for this study: NMC532 and A12 graphite processed with NMP and deionized water, respectively, as solvent. As received NMC532 powder (particle size distribution of approx. 11 \( \mu \)m and Brunauer-Emmett-Teller surface area approximately 0.3 m\(^2\)/g), Denka CB (powder grade), CMC (molecular weight approximately 250,000 g/mol and degrees of substitution approximately 0.9), PVDF Latex, and S130 PVDF were used in preparing NMC 532 cathodes. The components of A12 electrodes include A12, C65 CB, 9300 PVDF, CMC, and SBR.

The electrode fabrication process follows the standard procedure which has been reported previously. 40 Electrode slurry was prepared by a planetary mixer (Ross Laboratory Mixers & Blenders, Hauppauge, NY, USA) and electrodes were coated with a pilot-scale slot-die coater (Frontier Industrial Technology, Towanda, PA, USA). The mass loading of the NMC532 and A12 electrodes is 12.5 mg/cm\(^2\) and 6.5 mg/cm\(^2\), respectively. Table 1 lists the detailed information for the four types of electrodes used in this study.

The electrodes are then calendered by a 2-roller mill (International Rolling Mill, Pawtucket, RI, USA) with roller diameter of 152 mm and roller width of 330 mm at 80 \(^\circ\) C to different extents. The roller speed was set to 3 mm/min. Various gaps were set for final thickness. Thickness measurements of the fabricated electrodes were carried out with a digital micrometer (Marathon Watch Company, Richmond Hill, Ontario, Canada) with resolution of 1 \( \mu \)m. The thickness of the electrodes was measured at 24 positions spread over the width and length of the electrode area. The bulk porosity of the electrode samples was calculated based on the measured thickness of the electrodes following the procedure provided by Haselrieder et al. 41 Details of the bulk porosity calculation are provided in Supplementary Information.

**Surface tension measurement for probe liquids.—**The overall surface tension \( \gamma_{pl} \), of the probe liquids: propylene carbonate (Sigma-Aldrich, St. Louis, MO, USA), nicotinyl alcohol (Fisher Scientific, Hampton, NH, USA), and dimethyl sulfoxide (Fisher Scientific, Hampton, NH, USA), was measured using a BP100 bubble pressure tensiometer (Kruess, Hamburg, Germany). The immersion of the capillary is controlled by software, so the measuring procedure is entirely automatic. An air flow from the capillary produces bubbles in the sample, and a pressure sensor determines the maximum pressure during bubble formation, from which the surface tension is automatically calculated by software.

The static contact angle between the PTFE surface and the probe liquid (\( \theta_{PTFE} \)) was measured by following the procedure described in Static Contact Angle Measurement section. Once \( \theta_{PTFE} \) and \( \gamma_{pl} \) were known, \( \gamma_{pl}^D \) was determined by using Eq. 8. The value of \( \gamma_{pl}^D \) was then calculated by using \( \gamma_{pl}^D = \gamma_{pl} - \gamma_{pl}^D \). The results were verified by five independent experiments.

**Static contact angle measurement.—**The static contact angle was measured by using a contact angle goniometer. A small droplet, approximately 1 \( \mu \)L to 5 \( \mu \)L, was gently deposited by using a 500 \( \mu \)L threaded plunger syringe (Hamilton, Reno, NV, USA), which was positioned above the sample surface. Once the sessile drop reached its equilibrium, a PL-A662 microscopy camera (PixeLINK, Gloucester, ON, Canada) captured the image from the side view. Image processing was then performed using public domain ImageJ software. 42 If the droplet spreads over a substrate when gravity is absent, then the equilibrium shape is a spherical cap because this shape minimizes the free energy of the system. In the presence of gravity, the departure from the spherical cap shape increases with increasing Bond number, a dimensionless number representing the effect of gravity relative to the surface tension. 35 Since the Bond numbers for the droplets used in this study are typically in the range of 0.1 to 0.8, spherical cap configuration was always assumed in the drop shape analysis for determining the static contact angle. Each contact angle measurement was repeated at least five times to ensure excellent repeatability of the result.

**Advancing contact angle measurement.—**Advancing contact angles for various electrode surfaces were measured by using volume-changing method, i.e., \( \theta_a \) was measured by monitoring the advancing contact angle as the drop volume on the surface was dynamically changed. Before the measurements, the electrode samples were kept in a vacuum oven at 80 \(^\circ\) C for 14 hours to remove residual moisture. During the measurements, a small droplet was first formed and placed on the surface. A needle connected to a 500 \( \mu \)L threaded plunger syringe (Hamilton, Reno, NV, USA) was then brought close to the surface, and by dispensing more liquid from the syringe, the volume of the droplet was gradually increased. The whole process was recorded by a PL-A662 microscopy camera (PixeLINK, Gloucester, ON, Canada). Image processing was then performed using public domain ImageJ software. 42 Again, each measurement was repeated at least five times to ensure repeatability of the result.

**Characterization of surface elemental composition.—**To make sure that calendering does not change the surface composition of the electrode, X-ray photoelectron spectroscopy (XPS) analyses were carried out with a PHI VersaProbe Scanning XPS System (Physical Electronics, Chanhassen, MN, USA) using a focused monochromatized Al K\( \alpha \) radiation (h\( \nu = 1486.6 \) eV) operating at 50 W. Survey spectra were acquired at 117.40 eV pass energy with energy step size of 1 eV, whereas region spectra were acquired at 23.5 eV pass energy.
Table I. Four types of electrodes fabricated for this study.

<table>
<thead>
<tr>
<th>Electrode</th>
<th>Solvent</th>
<th>Electrode Composition</th>
<th>Loading</th>
</tr>
</thead>
<tbody>
<tr>
<td>NMC532-NMP</td>
<td>NMP</td>
<td>NMC532/Denka CB/5130 PVDF in 90/5/5 wt%</td>
<td>12.5 mg/cm²</td>
</tr>
<tr>
<td>NMC532-Water</td>
<td>Water</td>
<td>NMC532/Denka CB/CMC/PVDF in 90/5/14 wt%</td>
<td>12.5 mg/cm²</td>
</tr>
<tr>
<td>A12-NMP</td>
<td>NMP</td>
<td>A12/C65 CB/9300 PVDF in 92/2/6 wt%</td>
<td>6.5 mg/cm²</td>
</tr>
<tr>
<td>A12-Water</td>
<td>Water</td>
<td>A12/C65 CB/CMC/SBR in 92/2/24 wt%</td>
<td>6.5 mg/cm²</td>
</tr>
</tbody>
</table>

at step size of 0.075 eV. Prior to XPS measurements, the samples were sputter-cleaned with argon ions for 3 minutes at 2 kV and then another 3 minutes at 3 kV. The pressure in the analysis chamber was approximately 5 × 10⁻⁷ Pa. The analyzed area of the samples was 200 × 200 μm², and XPS measurements were conducted at three different spots on each electrode.

Surface texture characterization.—In order to effectively quantify the surface roughness and porosity of electrode surfaces, all the surfaces were characterized by using four different instruments, i.e., scanning electron microscope (SEM), atomic force microscope (AFM), optical profilometer (OP), and stylus profilometer (SP). The experimental details are shown as follows.

Surface characterization by scanning electron microscope.—SEM was performed on a SUPRA 550 field emission SEM (Carl Zeiss, Oberkochen, Germany). Since the electrode was already carbon-coated, there was no need for further sputter coating to improve the conductivity of the material. The distance of the electron source was set to 6 mm. An electron beam of 20 kV intensity was applied. For each surface, totally five 435 μm × 284 μm SEM images were taken at different spots.

Surface characterization by atomic force microscope.—AFM topography was measured using a CombiScope 1000 AFM (AIST-NT, Novato, CA, USA) in tapping mode. The tapping mode results in less damage to the sample than contact mode and is more accurate than non-contact mode when moisture is present on a sample. Tap300-G silicon probes (Budget Sensors, Sofia, Bulgaria) with a spring constant of 40 N/m, a resonance frequency of 300 kHz, and a probe radius of 10 nm were employed. True surface area was automatically calculated by software. All AFM measurements were performed in a nitrogen-filled glove box. The acquisition time for a typical 25 μm × 25 μm area was approximately 2 hours. For each surface, totally six 25 μm × 25 μm areas were scanned at different spots. Each measurement contains 500 × 300 data points, and the corresponding sampling resolution is 83.3 nm in both directions.

Surface characterization by optical profilometer.—OP measurement was done by Wyko NT1100 optical profiling system (Veeco Instruments, Plainview, NY, USA), which uses the interference patterns of light to scan through a range of heights and create a 3D profile of a desired surface without physically touching it. It can scan height changes of up to 100 μm with nanometer height resolution. The software Wyko Vision 32 associated with the instrument was used to create 2D profiles and 3D images of the surfaces as well as calculate true surface area. The magnification of the objective lens used in the measurement is ×10. The scan size is fixed by the magnification of the optical system, but a stitching procedure can be employed to obtain results for larger areas, in which a number of partially overlapping measurements are combined into one surface profile. The acquisition time for a typical 1 mm × 1 mm area was approximately 5 minutes. For each surface, totally five 1 mm × 1 mm areas were scanned at different spots. Each measurement contains 1214 × 1214 data points, and the corresponding sampling resolution is approximately 823.5 nm in both directions.

Surface characterization by contact profilometer.—SP measurement of surface profile was done by using a Dektak 8 stylus profilometer (Veeco Instruments, Plainview, NY, USA). This profilometer is very accurate with vertical resolution better than 1 nm. Two stylies with tip radius of 12.5 μm and 700 nm were used to scan across the sample surface, respectively. Vertical movements of the stylus were recorded, which revealed topography details of the surface. Measurements were done with the needle pressure value of 3 mg, as higher measuring pressure causes plastic deformation and even damage on the sample surface. The acquisition time for a typical 2-mm-long line was approximately 2 minutes. For each surface, totally ten 2-mm-long lines in different directions were scanned. The sampling resolution for the two stylies with tip radius of 12.5 μm and 700 nm was set to be 200 nm and 100 nm, respectively, in all directions.

Results

Fabricated electrode samples.—In order to investigate the relationship between degree of calendering and electrode wettability, the electrodes were calendered to different extents. Table II lists the detailed information including the bulk porosity for each electrode sample used in this study.

Surface tension of probe liquids.—The results for surface tension of the probe liquids, i.e., propylene carbonate (PL1), nicotinyl alcohol (PL2), and dimethyl sulfoxide (PL3), are summarized in Table S2 in Supplementary Information. Our results are close to, but different from, the data reported in the existing literature.

The slight deviation could be caused by variations in liquid quality and ambient temperature, pressure, and humidity, etc. The comparison shows that directly using the data reported from the existing literature could potentially lead to inaccuracies in SFE calculation. Fig. S2 in Supplementary Information shows the representative images of the static contact angles between the probe liquids and the PTFE surface.

Advancing contact angles on electrode surfaces.—The values of advancing contact angle θa measured for the three probe liquids, which are used as the values of apparent contact angle θ of the electrode, the advancing contact angles between the probe liquids and the electrode surfaces generally increase with decreasing values of bulk porosity of the electrodes (with only a few exceptions), as predicted by Eq. 10 and Eq. 12. In Eq. 10, for the same type of electrode with different values of bulk porosity, cos θ is a fixed positive value.

Table II. Electrode samples fabricated for this study.

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Electrode</th>
<th>Fabrication Process</th>
<th>Bulk Porosity</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>NMC532-NMP</td>
<td>Uncalendered</td>
<td>55.5% ± 2.5%</td>
</tr>
<tr>
<td>2</td>
<td>NMC532-Water</td>
<td>Uncalendered</td>
<td>55.1% ± 1.9%</td>
</tr>
<tr>
<td>3</td>
<td>A12-NMP</td>
<td>Uncalendered</td>
<td>55.9% ± 2.8%</td>
</tr>
<tr>
<td>4</td>
<td>A12-Water</td>
<td>Uncalendered</td>
<td>55.8% ± 1.3%</td>
</tr>
<tr>
<td>5</td>
<td>NMC532-NMP</td>
<td>Calendered</td>
<td>53.0% ± 1.8%</td>
</tr>
<tr>
<td>6</td>
<td>NMC532-NMP</td>
<td>Calendered</td>
<td>39.9% ± 2.9%</td>
</tr>
<tr>
<td>7</td>
<td>NMC532-NMP</td>
<td>Calendered</td>
<td>42.0% ± 2.0%</td>
</tr>
<tr>
<td>8</td>
<td>NMC532-NMP</td>
<td>Calendered</td>
<td>30.0% ± 2.5%</td>
</tr>
<tr>
<td>9</td>
<td>NMC532-Water</td>
<td>Calendered</td>
<td>46.4% ± 2.1%</td>
</tr>
<tr>
<td>10</td>
<td>A12-NMP</td>
<td>Calendered</td>
<td>44.2% ± 2.2%</td>
</tr>
<tr>
<td>11</td>
<td>A12-Water</td>
<td>Calendered</td>
<td>28.0% ± 1.8%</td>
</tr>
</tbody>
</table>
and thus when $\cos \theta^*$ is positive, and $\theta^*$ increases with decreasing values of $r$. Since $r$ increases with increasing bulk porosity, $\theta^*$ then increases with decreasing bulk porosity. Similarly, in Eq. 12, for the same type of electrode with different values of bulk porosity, $\cos \theta - 1$ is a fixed negative value, and thus when $\cos \theta$ is positive, and $\theta^*$ increases with increasing values of $f$. Since $f$ decreases with increasing bulk porosity, $\theta^*$ still increases with decreasing bulk porosity.

Surface elemental composition of electrode samples.—XPS measurements were carried out to study the chemical composition of each electrode surface. As we expected, the results indicate that calendaring does not change surface composition of electrode samples. For example, Sample 1, 5, 6, 7, and 8 are all NMC532-NMP, in which Sample 1 is not calendered but all the others are calendered to different degrees, and the XPS results demonstrate that they have almost the same surface composition, as shown in Fig. S3 in Supplementary Information.

Surface texture of electrode samples.—Representative SEM images and OP measurements for Sample 1 to Sample 8 are shown in Fig. 1 and Fig. 2, respectively. In Supplementary Information, representative AFM images are shown in Fig. S4, and representative SP roughness measurements carried out using styli with tip radius of 12.5 $\mu$m and 700 nm are shown in Fig. S5a and Fig. S5b, respectively. It can be seen that the sample surfaces are quite irregular, significantly deviating from ideal surfaces because of the presence of roughness and porosity. It also shows that calendaring process could significantly change the electrode surface texture.

### Table III. The values of advancing contact angle $\theta_A$ measured for probe liquids on each electrode surface.

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Electrode</th>
<th>$\theta_A$ (°) PL1</th>
<th>$\theta_A$ (°) PL2</th>
<th>$\theta_A$ (°) PL3</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>NMC532-NMP</td>
<td>11.19 ± 2.79</td>
<td>19.46 ± 1.52</td>
<td>14.88 ± 0.92</td>
</tr>
<tr>
<td>2</td>
<td>NMC532-Water</td>
<td>13.47 ± 2.87</td>
<td>20.88 ± 1.64</td>
<td>16.44 ± 0.94</td>
</tr>
<tr>
<td>3</td>
<td>A12-NMP</td>
<td>20.65 ± 2.34</td>
<td>30.32 ± 1.39</td>
<td>22.07 ± 0.58</td>
</tr>
<tr>
<td>4</td>
<td>A12-Water</td>
<td>14.99 ± 1.62</td>
<td>23.02 ± 1.59</td>
<td>16.73 ± 0.61</td>
</tr>
<tr>
<td>5</td>
<td>NMC532-NMP</td>
<td>10.60 ± 1.65</td>
<td>18.92 ± 1.40</td>
<td>13.92 ± 0.09</td>
</tr>
<tr>
<td>6</td>
<td>NMC532-NMP</td>
<td>19.08 ± 2.22</td>
<td>26.65 ± 1.11</td>
<td>21.57 ± 1.93</td>
</tr>
<tr>
<td>7</td>
<td>NMC532-NMP</td>
<td>18.29 ± 2.70</td>
<td>25.53 ± 1.29</td>
<td>19.35 ± 2.56</td>
</tr>
<tr>
<td>8</td>
<td>NMC532-NMP</td>
<td>24.25 ± 1.36</td>
<td>33.65 ± 1.49</td>
<td>25.80 ± 0.43</td>
</tr>
<tr>
<td>9</td>
<td>NMC532-Water</td>
<td>14.90 ± 0.93</td>
<td>21.78 ± 1.49</td>
<td>17.92 ± 1.60</td>
</tr>
<tr>
<td>10</td>
<td>A12-NMP</td>
<td>30.37 ± 1.44</td>
<td>35.68 ± 1.36</td>
<td>31.46 ± 1.21</td>
</tr>
<tr>
<td>11</td>
<td>A12-Water</td>
<td>31.48 ± 1.94</td>
<td>41.20 ± 0.93</td>
<td>34.80 ± 1.39</td>
</tr>
</tbody>
</table>

Surface roughness ratio.—One of the surface texture parameters of interest in contact angle calculations is the roughness ratio, $r$, defined as the true surface area divided by the projected surface area. It is used as an input parameter in the Wenzel model, as shown in Eq. 10. However, for irregularly rough surfaces such as the electrode surfaces used in this study, it is difficult to reliably assess the value of $r$.

When the sample surfaces are characterized using both AFM and OP, the value of $r$ is automatically calculated by software. The measurement of roughness using SP, however, is done by scanning of lines. Although the linear roughness ratio can be obtained by software, a method for estimating $r$ based on linear measurements needs to be developed. It is not an easy task to develop a general method for this purpose, and therefore simplistic models of regular roughness are often assumed in order to obtain a correlation between $r$ and the linear measurements. The simplistic model proposed by Meiron et al. demonstrated that if the linear roughness ratio is similar in both $x$ and $y$ directions, it may serve as a good approximation for the surface roughness ratio $r$. For the electrode surfaces used in this study, we verified that the linear roughness ratios obtained in different directions were very close, and their average value was therefore used as the surface roughness ratio $r$.

The values of the surface roughness ratio $r$ obtained from AFM, OP, and SP are listed in Table IV, respectively. Note that the SP results obtained by using two styli with tip radius of 12.5 $\mu$m and 700 nm are both provided in Table IV. It shows that the $r$ values of each electrode surface differ significantly depending on the measurement techniques used. The $r$ values obtained by OP and SP with two different tips show approximately the same trend: the $r$ values for Sample 1 and Sample 5 are higher than the others; the $r$ values for Sample 8 and Sample 11 are among the lowest; and the $r$ values for the other samples are somewhere in between. This trend makes sense to us considering the fact that Sample 1 and Sample 5 are both NMC532-NMP electrodes, one uncalendered and the other only slightly calendered, whereas Sample 8 and Sample 11 are both heavily calendered, having the lowest values of bulk porosity.

The $r$ values obtained by AFM do not follow this trend, and therefore are regarded as the least reliable. Remember that the maximum area reasonably measurable by AFM is about $25 \mu$m × 25 $\mu$m, which takes 2 hours to assess. The $r$ values obtained by AFM are thus local roughness ratios, highly depending on the sampling area. Local morphology and compositions vary significantly depending on the distribution of active materials, conductive carbon black, and pores in the binder. It can be concluded that, for irregularly rough surfaces, despite the high resolution, the small area allowed by AFM measurement restricts the representativeness of the roughness ratios and thus hinders accurate roughness analysis.
Although the $r$ values obtained by OP and SP with two different tips show approximately the same trend, the values still differ significantly. The $r$ values obtained by SP with tip radius of 700 nm is much higher than those obtained with tip radius of 12.5 μm. Since the tip of the stylus is spherical, the stylus cannot trace the surface profile properly if the widths of the pores or grooves are narrower than the radius of the stylus tip. The large tip makes the stylus to glide over finer details, filtering out minute roughness to provide a better view of the overall shape, while the smaller tip provides more accurate roughness details, thus much larger values of $r$. It can be concluded that, the $r$ values obtained by SP are very sensitive to tip radius, since the ability of the stylus to reproduce the original surface feature depends on the stylus size, and thus, to precisely measure delicate shapes and roughness of electrode surfaces, the radius of the stylus tip must be as small as possible. Based on the AFM measurement, it can be seen that the electrode surfaces contain submicron pores and structures. Even using the tip with radius of 700 nm, the submicron features are expected not to be collected. If SP is required to perform the roughness measurement, a much smaller tip is desirable to generate more accurate results. Moreover, SP measurements only produce linear roughness ratios, which is a different parameter from surface roughness ratios, although they are often assumed to be equal based on simplistic models of regular roughness. For surface roughness analysis, the surface is best represented as an “area with height deviation rather than a line with height deviation”, and therefore, the $r$ values obtained by SP are not adopted in this study.

Therefore, the optical profiler is the most suitable instrument for the roughness characterization of electrode surfaces due to its built-in capability to detect large range of vertical variation and to accommodate a large imaging area. The other advantages of the optical profiler include its non-contacting nature and fast data acquisition. In many previous investigations, the optical profiler has proven to be a sufficiently accurate method for the roughness analysis of electrode surfaces, however, a previous study indicated that objective lens with magnification of 10 or lower would not be sufficient for the samples with submicron roughness, because the submicron details would be smoothed out. Thus, we performed OP measurement with objective lens of higher magnification and AFM characterization on a typical sample, and the results are very similar to the $r$ value obtained from OP measurement with objective lens with magnification of 10. Therefore, the $r$ values obtained by OP using the objective lens with magnification of 10 were adopted in this study.

Surface solid-area fraction.—Another surface texture parameter of interest in contact angle calculations is the solid-area fraction, $f$, which is used as an input parameter in the Cassie-Baxter model, as shown in Eq. 12 and Eq. 13. It has always been a very challenging task to accurately estimate the value of $f$. In most studies, the value of $f$ was obtained by measuring the geometric dimensions of the nano-/micro-structures on the surface. For irregularly rough surfaces such as the electrode surfaces used in this study, direct measurement of the geometric dimensions of the surface structures would be prohibitive.

### Table IV. The surface roughness ratio of each electrode surface obtained by AFM, OP, and SP with two different tips, respectively.

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Electrode</th>
<th>$r$ (AFM)</th>
<th>$r$ (OP)</th>
<th>$r$ (SP: 12.5 μm)</th>
<th>$r$ (SP: 700 nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>NMC532-NMP</td>
<td>1.60 ± 0.19</td>
<td>2.47 ± 0.03</td>
<td>1.05 ± 0.01</td>
<td>1.23 ± 0.01</td>
</tr>
<tr>
<td>2</td>
<td>NMC532-Water</td>
<td>1.53 ± 0.27</td>
<td>1.73 ± 0.01</td>
<td>1.03 ± 0.01</td>
<td>1.16 ± 0.01</td>
</tr>
<tr>
<td>3</td>
<td>A12-NMP</td>
<td>1.44 ± 0.14</td>
<td>1.86 ± 0.01</td>
<td>1.03 ± 0.01</td>
<td>1.13 ± 0.01</td>
</tr>
<tr>
<td>4</td>
<td>A12-Water</td>
<td>1.42 ± 0.12</td>
<td>1.80 ± 0.01</td>
<td>1.02 ± 0.01</td>
<td>1.15 ± 0.01</td>
</tr>
<tr>
<td>5</td>
<td>NMC532-NMP</td>
<td>1.39 ± 0.15</td>
<td>2.49 ± 0.07</td>
<td>1.06 ± 0.01</td>
<td>1.23 ± 0.01</td>
</tr>
<tr>
<td>6</td>
<td>NMC532-NMP</td>
<td>1.28 ± 0.12</td>
<td>1.80 ± 0.01</td>
<td>1.01 ± 0.01</td>
<td>1.12 ± 0.01</td>
</tr>
<tr>
<td>7</td>
<td>NMC532-NMP</td>
<td>1.31 ± 0.09</td>
<td>1.90 ± 0.04</td>
<td>1.01 ± 0.01</td>
<td>1.13 ± 0.01</td>
</tr>
<tr>
<td>8</td>
<td>NMC532-NMP</td>
<td>1.25 ± 0.06</td>
<td>1.20 ± 0.02</td>
<td>1.00 ± 0.01</td>
<td>1.05 ± 0.01</td>
</tr>
<tr>
<td>9</td>
<td>NMC532-Water</td>
<td>1.28 ± 0.10</td>
<td>1.71 ± 0.02</td>
<td>1.03 ± 0.01</td>
<td>1.11 ± 0.01</td>
</tr>
<tr>
<td>10</td>
<td>A12-NMP</td>
<td>1.27 ± 0.09</td>
<td>1.72 ± 0.01</td>
<td>1.03 ± 0.01</td>
<td>1.12 ± 0.01</td>
</tr>
<tr>
<td>11</td>
<td>A12-Water</td>
<td>1.24 ± 0.06</td>
<td>1.47 ± 0.01</td>
<td>1.01 ± 0.01</td>
<td>1.08 ± 0.01</td>
</tr>
</tbody>
</table>
Moreover, there exists little evidence about whether this geometric assessment of roughness truly represented \( f \). In some studies, researchers tried to obtain the value of \( f \) by direct observation of the true solid fraction that a liquid is actually in contact with. For example, Kwon et al.\textsuperscript{24} observed liquid droplets on a hydrophobic fabric surface using an environmental scanning microscope, but the resolution of the microscope was not high enough to observe the interface between the liquid and the fabric surface. Even worse, the liquid droplets on the surface were not stably positioned, and they fell off the surface during the observation.

In this study, an innovative method to obtain the value of \( f \) is proposed as shown below. Assume that there are two different liquids: Liquid A and Liquid B, and there are two different surfaces made of the same material but with different surface textures: Surface \( \alpha \) and Surface \( \beta \). For example, Sample 1 and Sample 5 are both NMC532-NMP, so they have the same surface composition as confirmed by XPS measurements, but they have different surface textures, since Sample 5 was calendared, but Sample 1 was not calendared. Assuming that the wetting state of the two liquids on such surfaces falls in the Cassie-Baxter regime, the relationship between the apparent contact angle \( \theta^* \) and the intrinsic Young’s contact angle \( \theta \) can be expressed by Eq. 12 or Eq. 13. Here Eq. 12 is used for illustration. The derivation is very similar if Eq. 13 is used.

Denote the solid-area fraction of Surface \( \alpha \) and Surface \( \beta \) as \( f_\alpha \) and \( f_\beta \), respectively. Since the two surfaces are made of the same material, the intrinsic Young’s contact angle of Liquid A on the two surfaces are the same, which is denoted as \( \theta_\alpha \), and the intrinsic Young’s contact angle of Liquid B on the two surfaces is denoted as \( \theta_\beta \). Since the two surfaces have different textures, the apparent contact angles of Liquid A on Surface \( \alpha \) and Surface \( \beta \) are different, which are denoted as \( \theta^*\alpha \) and \( \theta^*\beta \), respectively. Similarly, the apparent contact angles of Liquid B on Surface \( \alpha \) and Surface \( \beta \) are denoted as \( \theta^*\alpha \) and \( \theta^*\beta \), respectively. Then we have the following nonlinear system of four equations:

\[
\begin{align*}
\cos \theta^*\alpha - [f_\alpha \cos \theta_\alpha - 1 + 1] &= 0 \\
\cos \theta^*\beta - [f_\alpha \cos \theta_\beta - 1 + 1] &= 0 \\
\cos \theta^*\alpha - [f_\beta \cos \theta_\alpha - 1 + 1] &= 0 \\
\cos \theta^*\beta - [f_\beta \cos \theta_\beta - 1 + 1] &= 0
\end{align*}
\]

[15]

Since the apparent contact angles \( \theta^*\alpha \), \( \theta^*\beta \), \( \theta^*\alpha \), and \( \theta^*\beta \) can be obtained from experimental measurements, we can solve the nonlinear system of four equations with four unknowns \( f_\alpha \), \( f_\beta \), \( \theta_\alpha \), and \( \theta_\beta \). This process will not only provide the solution for the solid-area fractions \( f_\alpha \) and \( f_\beta \), but also the actual contact angles \( \theta_\alpha \) and \( \theta_\beta \).

Note that this derivation assumes that the wetting state of the two liquids on both surfaces falls in the lyophilic Cassie-Baxter regime. Once the solid-area fractions \( f_\alpha \) and \( f_\beta \) are obtained, Eq. 14a and Eq. 14b can be used to calculate the two critical contact angles. By comparing the critical contact angles with the obtained actual contact angles, we can check if this assumption is true. If the wetting state does not fall in this regime, the results are then not valid.

The 33 apparent contact angles listed in Table III are all less than 90°, so the wetting states fall in the lyophilic regime, either Cassie-Baxter or Wenzel. Assuming that all of them are in the lyophilic Cassie-Baxter regime, based on the 33 apparent contact angles, we can generate 33 nonlinear equations just like Eq. 15. Since we tested 4 types of electrodes with 11 different surface textures using 3 probe liquids, we have 12 intrinsic contact angles and 11 different values of solid-area fraction as unknowns. For this overdetermined system with 23 unknowns and 33 equations, we obtain the solution by minimizing the Frobenius norm of the residual.

The results show that there are four cases that do not fall in the lyophilic Cassie-Baxter regime: nicotinyl alcohol on Sample 3, Sample 4, Sample 10, and Sample 11. We then solve the remaining 29 equations following the same procedure, and this time the results show that all the 29 cases fall in the lyophilic Cassie-Baxter regime.

At this stage, we have obtained the values of 8 intrinsic contact angles and 11 solid-area fractions. The MATLAB codes are provided in the Supplementary Information. The intrinsic contact angles for the four remaining cases are then solved by using the lyophilic Wenzel model, i.e., Eq. 10. Substituting the \( r \) values obtained from OP measurements and the apparent contact angles listed in Table II, we can calculate the intrinsic contact angles for the four cases. Table V lists the values of solid-area fraction, roughness ratio, and two critical contact angles of each electrode surface. Table VI lists the values of actual contact angles for each case.

Note that when we solve the system of 29 equations with 23 unknowns, such a system represents four independent systems for four different types of electrodes with one significantly more overdetermined than the others, since for NMC532-NMP-type electrodes, we have five different surface textures, but for the other types of electrodes, we have only two different surface textures. To demonstrate that two surface textures are sufficient to accurately predict the intrinsic contact angles, the results obtained for NMC532-NMP type of electrodes based on two surface textures and those obtained based on five surface textures are compared. The results and the associated MATLAB codes are provided in the Supplementary Information.

### Surface free energy of electrode samples

Substituting the polar and dispersive components of the surface tension of the three probe liquids, as listed in Table S2 in Supplementary Information, and the intrinsic Young’s contact angles, as listed in Table VI, into Eq. 7, we then use the OWRK model to compute the polar and dispersive components of the SFE of the electrode samples. The two parameters \( x \) versus \( y \), as defined in Eq. 7, are plotted as data points for each electrode sample, and then the best-fit least-square regression line is drawn. The slope of the line gives the polar component and the vertical intercept generates the dispersive component of the solid SFE, respectively. Fig. 3 shows the SFE plots of all the electrode samples fitted using the OWRK model. The corresponding SFE values and the convergence factors are listed in Table VII. Note that all the convergence factors are close to 1, showing that the OWRK model is suitable to calculate the SFE of the electrode samples with a high degree of accuracy. It shows that, when switching manufacturing from conventional solvent based processing to aqueous processing, the polar component of SFE increases; it increases 74% from NMC532-NMP to NMC532-Water, and 48.2% from A12-NMP to A12-Water. It is important to point out that the solvent, which is evaporated during the drying process, has negligible influence on the final SFE of the electrode. The SFE change

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Electrode</th>
<th>PL1 ( \theta ) (°)</th>
<th>PL2 ( \theta ) (°)</th>
<th>PL3 ( \theta ) (°)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1, 5–8</td>
<td>NMC532-NMP</td>
<td>28.66</td>
<td>44.92</td>
<td>34.07</td>
</tr>
<tr>
<td>2, 9</td>
<td>NMC532-Water</td>
<td>23.03</td>
<td>35.94</td>
<td>28.18</td>
</tr>
<tr>
<td>3, 10</td>
<td>A12-NMP</td>
<td>49.69</td>
<td>62.02</td>
<td>53.32</td>
</tr>
<tr>
<td>4, 11</td>
<td>A12-Water</td>
<td>46.32</td>
<td>59.15</td>
<td>52.03</td>
</tr>
</tbody>
</table>

Table VI. The values of intrinsic Young’s contact angles for probe liquids on each electrode surface.
we observed when switching manufacturing to aqueous processing is attributed to the different binders as listed in Table I.

The improved polar component of the SFE of the water-based electrodes is believed to be beneficial to electrolyte wetting, as electrolyte wetting not only depends on the overall SFE of the electrode but also its individual components. In the best scenario of electrolyte wetting, the dispersive and polar components of the surface tension of electrolyte should be identical to the corresponding component of the SFE of the electrode. In general, however, the often-used electrolytes have a higher polar component of the SFE than the electrodes. Thus, the increased polar component of the water-processed electrodes would lead to enhanced electrolyte wetting. In addition, the slight increase in the overall SFE of the water-processed electrodes improves their wettability as well.

For the purpose of comparison, we also substituted the apparent contact angles, as listed in Table III, into Eq. 7, to compute the apparent SFE of the electrode samples. The apparent SFE values and the convergence factors are listed in Table S3 in Supplementary Information. It shows that the apparent SFE is significantly higher than the corresponding actual SFE, indicating that using the apparent contact angles to calculate SFE could greatly overestimate SFE. It also shows that, for the same type of electrode, the apparent SFE values decrease with increasing degree of calendaring, which indicates lower wettability of electrolyte on denser electrodes. It can be concluded that the electrolyte wetting process becomes even more critical and challenging when electrodes are highly compressed, e.g., bulk porosity less than 20%, for higher volumetric energy density.

### Concluding Remarks

In this study, we report a systematic study on the characterization of the SFE of composite electrodes for lithium-ion batteries. The OWRK model is adopted to calculate SFE of electrode surfaces. Propylene carbonate, nicotinyl alcohol, and dimethyl sulfoxide are used as probe liquids, and their contact angles on various electrode samples are measured. Roughness and porosity are characterized for each electrode surface, and the results are analyzed using the Wenzel model and the Cassie–Baxter model. The effect of calendering on electrode SFE is investigated in detail. In addition, the SFE of electrodes obtained via solvent-based and water-based processing are compared. It shows that, when switching manufacturing from conventional solvent based processing to aqueous processing, the polar component of SFE increases; it increases 74.1% from NMC532-NMP to NMC532-Water, and 48.2% from A12-NMP to A12-Water. The change in SFE we observed when switching manufacturing to aqueous processing is attributed to the different binders. The results provide valuable

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Electrode</th>
<th>$\gamma_{sv}$ (mN/m)</th>
<th>$\gamma_{sv}^D$ (mN/m)</th>
<th>$\gamma_{sv}^P$ (mN/m)</th>
<th>Convergence Factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>1, 5–8</td>
<td>NMC532-NMP</td>
<td>38.43</td>
<td>28.83</td>
<td>9.59</td>
<td>0.95</td>
</tr>
<tr>
<td>2, 9</td>
<td>NMC532-Water</td>
<td>40.56</td>
<td>23.86</td>
<td>16.70</td>
<td>0.89</td>
</tr>
<tr>
<td>3, 10</td>
<td>A12-NMP</td>
<td>30.10</td>
<td>24.80</td>
<td>5.31</td>
<td>0.99</td>
</tr>
<tr>
<td>4, 11</td>
<td>A12-Water</td>
<td>30.33</td>
<td>22.46</td>
<td>7.87</td>
<td>1.00</td>
</tr>
</tbody>
</table>
guidance for our goal to optimize the electrolyte wetting process, helping lower costs and enable higher product quality. In addition, the characterization method presented in this study can be used to systematically investigate the effect of other manufacturing factors on electrode wettability, such as slurry formulation, mixing sequences, and drying protocols, etc.

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