**XPS information**

This experiment uses X-ray photoelectron spectrometer (ThermoFischer, ESCALAB 250Xi; Axis Ultra DLD Kratos AXIS SUPRA; PHI-5000 versaprobe) for testing. The source gun type is Al Kα X-ray source (monochromatic, hν = 1486.6 eV) and the vacuum degree of the analysis room is 8×10-10 Pa. The working voltage is 12.5 kV and the filament current is 16 mA. The signal accumulation is performed for 10 cycles. The full spectrum and narrow spectrum of the Passing-Energy are 50 eV and 20 eV, respectively. The step size is 0.05 eV, and the dwell time is 40-50 ms. And we use C1s = 284.80 eV as energy standard to perform charge correction. Besides, the form of the sample is powder and the geometry structure of the sample is core-shell Si@SiOx particles.

**Table 1**

Fitting parameters obtained from XPSPEAK41 software.

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| Samples | Background | ValenceState | Position | FWHM | Area | Intensity | Fitting Degree |
| Low BE | High BE | Style |
| 550 °C | 100 | 108 | Shirley | Si+  |  | high |
| Si2+ |
| Si3+ | 102.7 | 1.31 | 12289.3 | 9543.8 |
| Si4+ | 103.5 | 1.65 | 33733.1 | 19956.8 |
| 1150 °C | 100 | 108 | Shirley | Si+  |  | high |
| Si2+ |
| Si3+ | 102.7 | 1.31 | 5239.5 | 4535.4 |
| Si4+ | 103.5 | 1.65 | 45355.0 | 26693.7 |

The wide spectrum of all elements, and the Si 2p spectra of the 550 °C and 1150 °C samples are very important. Therefore, these XPS data have been uploaded to the supplementary material. Among them, the valence of Si in the oxide coatings is distributed within the range of 100-108 eV, so the Si 2p spectra of the 550 °C and 1150 °C samples are fitted with four peaks in this region. The fitting software used in the analysis is XPSPEAK41. To compare the XPS difference between the low-crystallinity sample (550 °C) and the high-crystallinity sample (1150 °C), we import the Si 2p data of the 550 °C and 1150 °C sample into the software. First, the background is set in the range of 100-108 eV and the background style is set to shirley. Next, add fitting peaks in the region according to the peak position of Si+, Si2+, Si3+ and Si4+, respectively. Then, fix the peak position of each peak and adjust the FWHM (full width at half maximum) and the area to improve the fitting degree between the two total peak. Finally, based on the high fitting degree between the original total peak and the fitting total peak, we determine the fitting parameters. The peak position, FWHM, area and intensity are the main parameters to determine lineshapes and lineswidths. In addition, the fitting degree is a manifestation of the error, and the errors in fitting parameters are very small due to the high fitting degree. To make these information more accessible, the specific parameters from the XPSPEAK41 are shown in the table 1. The peak representing the Si3+ is significantly weakened and the peak representing the Si4+ is significantly enhanced after dry oxidation at 1150 °C. The transformation from Si3+ (SiO1.5) into Si4+ (SiO2.0) demonstrates the formation of oxygen rich Si compounds. This is consistent with the result in the paper.