## Pile-up/sink-in area calculation

To calculate the pile-up/sink-in area from the *in situ* observations, the following Equation was used (see FIG. S1(b) for the case of pile-up):

$A\_{p,s}=3\frac{de}{2}$ (1),

where *e* is the projected pile-up height as shown in FIG. S1. The length *e* is then determined using the following Equation:

$e=b\sin(θ)$ (2),

where *b* is shown is Figure 1(a). The length *b* is then calculated from the length *a* by the following relation:

$b=\frac{a}{\cos(\left(θ+γ\right))}$ (3).

Combining Equations 12-14, Equation 9 (from the paper) is directly obtained.

To determine geometrically *d*, lines are plotted along the tip edges in order to detect the contact borders. To determine a, lines are plotted tangent to the pile-up (or sink-in), the intersection giving directly *a*.



*FIG. S1: Schematic figure showing the contact area measurement from in situ indentation observations with (a) showing the angles and dimensions involved for the case of pile-up and (b) illustrating these dimensions on a plan-view secondary electron micrograph.*

## Error in the calculation of the projected contact area

During SEM imaging of the imprint, some errors can affect the precision of the measurement. The first error in the true projected contact area calculation is due to uncertainty in the included tip angle and the inclination angle of the indentation instrument. The angular deviation of the included tip angle and machine inclination is typically +/- 0.3°. Using this as a worst case scenario, a deviation of 0.3° would result in an error in *A*c measurement of 0.5%. Consequently, this error is almost negligible compared to the variation observed during testing.

A more significant error arises from the imprint lengths measurement. When measuring the lengths *a* and *d* from the peak-load image, variation can reach 2 to 4 pixels, which corresponds to an actual length of 100 to 200 nm at this magnification. To check how this error might affect the measured contact area calculation, the lengths *d* and *a* were replaced in Equations 8 & 9 (from the paper) by *d+Δd* and *a+Δd,* respectively, with *Δd* = 200 nm. In Equation 9 (from the paper), we suggested that *a*= 0.011*d*, which represents the smallest ratio we obtained in this study. This is also the case where the error in the contact area calculation is the most critical in our study. The results are plotted in FIG. S2, where the error in *A*c, *A*t and *A*ps are plotted as a function of the length *d*. For every contact area measurement, the error is more significant as the length d decreases. The most important error comes from the pile-up, or sink-in measurement, which is around 40% for high *d* values. But as shown in Figure 8, this error has less effect in the *A*c calculation. In our case the length *d* is around 42 µm, which means that the total error is around 10%. This figure underscores the most important errors comes from the pile-up measurement. However, it is important to note that this is the worst case encountered in our study. If the pile-up has larger dimensions, the error in the contact area calculation will be less important.

Finally, the last source error could come from the calibration of the SEM, itself. Usually, the SEM calibration is performed once a year, minimizing greatly this error.



*FIG. S2: Error in the calculation of the projected contact Ac as a function of the length d. The error in At calculation is very small in comparison to the pile-up or sink-in area Aps.*