Comment on "Large Slip of Aqueous Liquid Flow over a Nanoengineered Superhydrophobic Surface"

In a recent Letter [1], Choi and Kim reported slip lengths of a few tens of microns for water on nanoengineered superhydrophobic surfaces, on the basis of rheometry (cone-and-plate) measurements. We show that the experimental uncertainty in the experiment of Ref. [1], expressed in term of slip lengths, lies in the range 20–100 μm, which is precisely the order of magnitude of the reported slip lengths. Moreover, we point out a systematic bias expected on the superhydrophobic surfaces. We thus infer that it is not possible to draw out any conclusion concerning the existence of huge slip lengths in the system studied by Choi and Kim.

Choi and Kim performed torque measurements using a commercial rheometer (AR2000, TA Instruments) with a cone-and-plate geometry. The slip length δ is deduced from the correction of the torque M with respect to a reference no-slip value M_0: $\delta = \frac{2}{3}\theta_0 R(1 - M/M_0)$. $M_0 = \frac{2\pi}{3}\mu\Omega R^3/\theta_0$ is the prediction in the absence of slippage; R is the cone radius ($\simeq$3 cm) and θ_0 its opening angle (2°); μ is the viscosity of the liquid; Ω is the rotational velocity. Putting numbers in this expression shows that the reported 20 μm slip length for water corresponds to a 3% correction to the (small) reference torque $M_0 \sim 5$ μNm. The authors, however, claim a 3 μm uncertainty on the slip length, which corresponds to an overall 0.5% error bar on the *relative deviation* of the torque $(M_0 - M)/M_0$. Such a precision is not attainable in the present experiment. To illustrate this uncertainty issue, we have performed benchmark experiments using an AR2000 rheometer with a smooth, stainless steel cone-and-plate geometry (with the same radius and cone angle), very close to that of Ref. [1]. This rheometer was calibrated using a reference Newtonian silicon oil (BR0050CPS, $\mu = 48.4$ mPa s at 25 °C), which yielded the expected value to within 0.5%. Then, turning to distilled deionized water at 25 °C, we performed torque measurements similar to [1] and measured the torque standard deviation $\Delta M/M$ with this liquid, yielding $\Delta M/M \simeq 1.4\%$ at $\dot{\gamma} = 150$ s^{-1}, up to 3.5% at $\dot{\gamma} = 50$ s^{-1}. This is far above the claimed 0.5% uncertainty. If the uncertainties on the filling volume and on the gap size are included, the global uncertainty on the measured torque is at least $\Delta M/M \simeq 3\%$. Finally, using the expression $\delta(M)$ and adding a similar uncertainty on the reference viscosity μ in M_0 leads to $\Delta\delta/\delta \simeq 100\%$–200%, so that $\Delta\delta \sim 20$–40 μm for water and $\Delta\delta \sim 50$–100 μm for glycerin. The reported effect are therefore *within uncertainty* and the experiment of Ref. [1] should be considered as inconclusive.

Another source of difficulties in the interpretation of the experiment in Ref [1] is the role of secondary flows. The relevant Reynolds number is $\mathrm{Re} = \rho\Omega R^2\theta_0^2/\mu \gtrsim 2$, and inertial effects should lead to an increase in the torque up to 2% ($M - M_0 = 6 \times 10^{-4}\mathrm{Re}^2$ [2]). In view of the claimed resolution, this correction should be measurable in the experiment of Ref. [1] and interpreted as an apparent negative slip length up to ~ -15 μm for the smooth hydrophilic surfaces. This effect is not detected in [1], which furthermore confirms the weakness in the interpretation of the measurements. At this stage, it is worth pointing out a *systematic bias* on the superhydrophobic surfaces. Indeed, for the same liquid volume filling the gap in the cone and plate, the meniscus at the edges makes the radius R slightly smaller on the superhydrophobic surface (with very large contact angle), with respect to the other surfaces with smaller contact angles. The variation in R is predicted to be of the order of a fraction of the gap at the border $\Delta R \approx -\alpha\theta_0 R$ (with $\alpha \lesssim 1$). Assuming no slip at the surfaces, the resulting decrease of the solid-liquid area leads to a reduction of torque on the superhydrophobic surfaces, $\Delta M_{\mathrm{cap}}/M_0 \approx -3\alpha\theta_0$, of the order of a few percents. The misinterpretation of this effect using the equation for $\delta(M)$ thus erroneously predicts a slippage effect with a slip length $\delta_{\mathrm{eff}} \approx 2\alpha\theta_0^2 R$, of the order of a few tens of micrometers. See also note [3].

In summary, the experimental uncertainty that we estimate is comparable to the amplitude of the effect the authors have observed. Moreover, a systematic bias could be wrongly interpreted in terms of very large slippage on superhydrophobic surfaces. The experiments of Ref. [1] are therefore inconclusive.

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[1] C.-H. Choi and C. Kim, Phys. Rev. Lett. **96**, 066001 (2006).
[2] K. Walters, *Rheometry* (Chapman Hall, London, 1975).
[3] To fix the ideas, a retreat of ~0.2 mm in the average position of the edge meniscus could be misinterpreted as a slip length increase of ~14 μm. This underlines the importance of an effect which was erroneously overlooked in Ref. [1].

