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Contact angle measurement using a Hele-Shaw cell: A proof-of-concept study

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ABSTRACT

Contact angle is an important property to quantify the wettability of a solid surface with a liquid, which characterizes interactions of the solid-liquid pair. Generally, to measure contact angle, special instruments such as a goniometer are necessary, but they are not readily available in certain research settings. In this study, an alternative method to measure contact angle based on a Hele-Shaw cell, microscopy imaging, and image processing is suggested. In this method, a liquid drop is injected into a transparent Hele-Shaw cell, the meniscus of the drop is captured in the top or bottom view using a brightfield microscope, and the contact angle is measured from the captured image based on a geometrical model of the drop in the Hele-Shaw cell. The proposed method was tested using two different liquids (deionized water and glycerol) and two different solid surfaces (polydimethylsiloxane [PDMS] and commercial water-repellent-coated glass) with various gap heights of Hele-Shaw cells and validated in comparison with a goniometer. Our results show that the proposed method could measure contact angles reasonably well. Although the concept of the proposed quick-and-dirty contact angle measurement method was proven in this study, there is still room to improve the accuracy and reliability of the proposed method.

1. Introduction

When a liquid drop is placed on a solid surface in a gas, the contact angle (CA) of the drop is an important parameter to quantify the surface wettability of the solid base, which can be seen as an interaction between a fluid and a solid structure [1,2]. Various methods have been developed and employed to measure CA [3–5]. Direct CA measurement methods include the sessile drop method and the captive bubble method in which CA can be measured directly from the profile of a drop or bubble on a solid surface at its three-phase contact line. Indirect measurement methods include the tilting plate method, the reflection method, the Wilhelmy-gravitational method, the capillary rise method, and the capillary bridge method. In these methods, physical quantities other than CA are measured, such as tilting angles in the tilting plate method and downward forces on a plate in the Wilhelmy-gravitational method, and CA is found from the measured quantities based on various theoretical models.

In general, goniometers are widely used for direct CA measurements. In this method, a side view image of a drop on a solid surface is captured using a camera, and the profile of the drop is found by image processing. Then, CA is measured directly between the tangential line of the drop profile at the three-phase contact point and the solid surface. Despite its popularity, the goniometer method has some limitations. First, CA measurement can be affected by uncertainty in determining the tangential line and the solid surface. Second, goniometers are not readily available in various research settings due to its front-end investment.

In this article, we propose an easy-to-adopt method to measure CA and provide preliminary results for proof-of-concept. In the proposed method, a liquid drop of interest is sandwiched between two parallel surfaces of a transparent solid of interest with a small gap (i.e., the Hele-Shaw cell), and a top or bottom view image of the meniscus of the drop is captured. Since the meniscus appears dark in the image, its width can be determined by image processing, which is converted to CA via a simple geometrical model. Since this method is based on the fact that the curved meniscus of a drop appears dark under collimated illumination because light is refracted while passing through the meniscus, the method is similar to CA measurement methods based on top view imaging [6–10]. The proof-of-concept study of the proposed method was conducted by measuring the CA of deionized (DI) water on polydimethylsiloxane (PDMS) surfaces in air using the proposed method and

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then by comparing the measured CA values with the reference value measured by a goniometer. Then two more cases (glycerol on PDMS and DI water on water-repellent-coated glass) were tested to broaden the application scope of the method.

2. Experimental procedure

A Hele-Shaw cell device consisted of two PDMS-coated glass slides ($75 \times 25 \times 1$ mm^3) and spacers and was fabricated as follows. PDMS (Sylgard 184, Dow Corning) was prepared with the standard weight ratio of 10 (base) :1 (curing agent), and degassed PDMS was spin-coated on a clean glass slide using a spin coater (WS-650MZ-23NPPB, Laurell; 1000 rpm for 1 min). Then, PDMS was cured at 60°C overnight in an oven. The thickness of the obtained PDMS layer was measured as 80 μm [11]. Two PDMS-coated glass slides were stacked with the PDMS side facing each other while No. 1.5 glass coverslips were placed at both ends of the glass slides as spacers to control the gap height of the Hele-Shaw cell ($H = 0.17$, 0.36, 0.57, 0.74 and 1 mm). The entire Hele-Shaw cell device was clamped tightly by using magnets placed on top of the device and below the microscope stage.

As shown in Fig. 1, the Hele-Shaw cell device was placed on the stage of an inverted brightfield microscope (IX81, Olympus) and a drop of DI water was injected through a needle (outer diameter: 0.025 inch, New England Small Tube Corporation) using a syringe, tubing and a syringe pump (Fusion 200, Chemyx; volume flow rate = 0.001 mL/min). Thus, a DI water drop grew in the Hele-Shaw cell, and once the drop grew big enough for imaging, injection was stopped. After a short period, the drop stopped growing and remained stationary. The meniscus of the drop farthest from the needle tip was chosen for the region of interest (ROI; the red dashed box in Fig. 1). The drop meniscus in the ROI was imaged with 4× and 10× lenses and a digital camera (PL-B959U, Pixelink; pixel sizes: 1.1 μm/pixel and 0.44 μm/pixel, respectively) while the outer boundary of the meniscus was focused (the focal plane in Fig. 1). The outer diameter of the needle was larger than the gap height in some experiments, but the gap height at the ROI was not affected significantly due to the distance between the needle tip and the ROI.

Fig. 2(a) shows an example image ($H = 0.36$ mm, 4×), and the dark ring in the image is the meniscus of the drop. The width of the meniscus (w) was defined to be the shortest distance between the inner and outer boundaries of the dark ring and measured by image processing using MATLAB (MathWorks). First, the two boundaries were identified based on contrast between the background and the dark ring. The captured image was converted to a binary image as shown in Fig. 2(b) by using MALTAB function 'imbinarize' with the 'adaptive' method and the 'sensitivity' factor of 0.92. Then, the outlines of the meniscus were traced by using 'bwtraceboundary' as shown in Fig. 2(c). Second, distances for all pairs between points on the inner boundary and those on the outer boundary were calculated, and the minimum distance for each point on the inner boundary was found as the local width of the meniscus. Last, the found minimum distances were averaged to determine the averaged meniscus width (w). Fig. 2 shows that our method could identify the inner and outer boundary of the meniscus well, and w was found to be 37.9 ± 1.9 μm.

Fig. 3 shows a geometrical model to convert H and w to CA (θ_C). This model assumes that the meridional profile of the drop in the Hele-Shaw cell is circular, and that w is the horizontal distance between the three-phase contact point (A), and the outermost point (or equator) of the meniscus (B). Here, θ_C equals $90^\circ + \angle DAC$, and $\angle DAC$ is found as follows. Since ΔACB is an isosceles triangle, $\angle BAC = \angle ABC$, which is named as θ. Since AD and BC are parallel, $\angle DAB = \angle ABC$. Thus, $\angle DAC = 2\theta$. Here, $\theta = \tan^{-1}[w/(H/2)]$ from ΔABD, and thus $\theta_C = 90^\circ + 2\tan^{-1}(2w/H)$. With this model, the CA of Fig. 2 was found to be $\theta_C = 114^\circ$.

3. Result and discussion

Table 1 summarizes measured w values and calculated θ_C values of DI water drops between PDMS surfaces with various H values and the magnification ratios of microscopy imaging. Repeated measurements with a certain H value and a certain magnification ratio showed that θ_C values were similar, and the ratio of standard deviation (σ) to average (m) was also low. For example, the largest σ was 5° for the 10× and $H = 0.36$ mm case, and the case's m was 111°. Thus, the maximum σ/m ratio was 4.5%. Therefore, the proposed method could measure CA with good repeatability. Fig. 4 shows that differences in the average θ_C values between 4× and 10× imaging were smaller than 5°, which is comparable to the uncertainty of other CA measurement methods (±2°) [3–5, 12]. Thus, the magnification ratio of microscopy imaging did not affect the CA measurements significantly.

As a reference to validate the proposed method, the CA of DI water drops on PDMS was measured to be $106 \pm 2^\circ$ ($N = 8$) by using a goniometer (Theta Goniometer, Attension). As shown in Fig. 4, θ_C was larger than the reference value when H was small, and as H increased over 0.57 mm, θ_C approached closer to the reference value. Overall, the proposed method could measure θ_C less than 10° different from the reference value, and it depended on H.

For further validation of the proposed method, the method was tried with different liquid and substrate: glycerol (86–89%, Sigma-Aldrich) on PDMS-coated glass (Fig. 5), and DI water on glass coated with water repellent (Rain-X original, Illinois Tool Works; Fig. 6). For the water

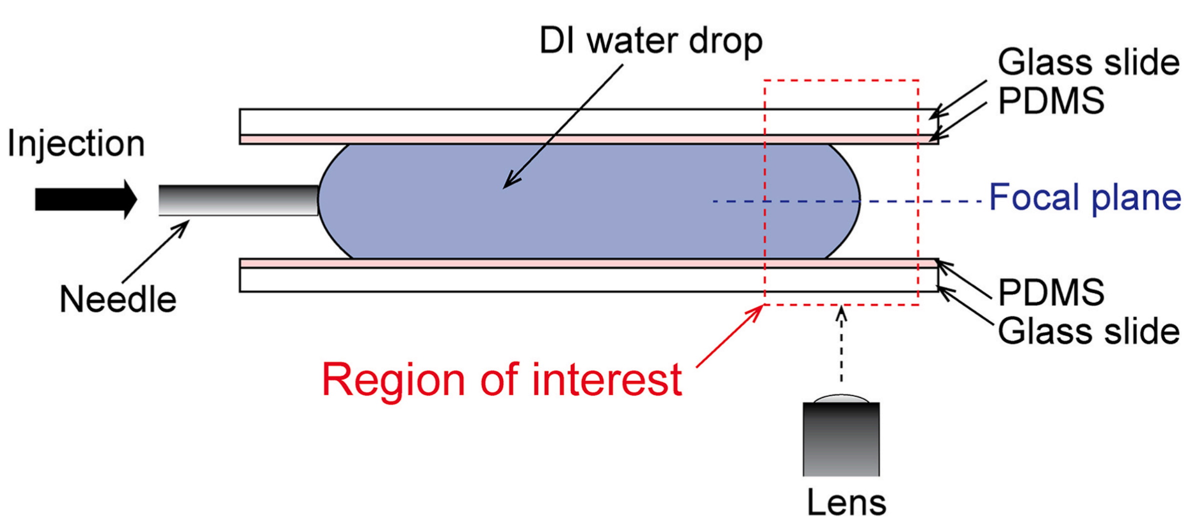

Fig. 1. Schematic of the experimental setup for a DI water drop between PDMS surfaces. Spacers and magnets are omitted for clarity.

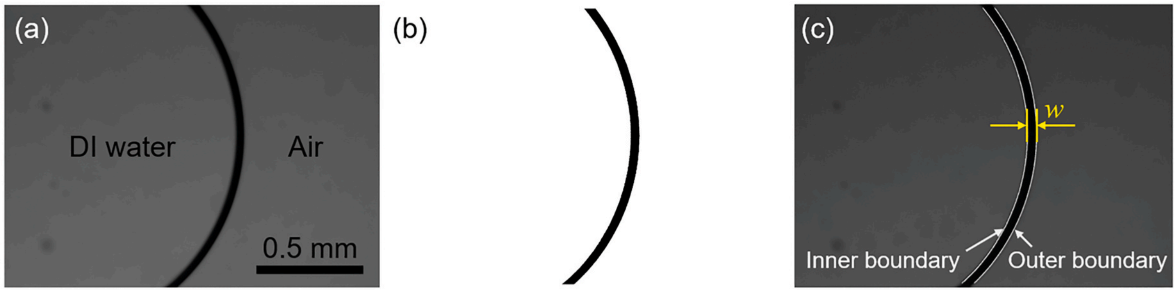

Fig. 2. An example image of a DI water drop ($H = 0.36$ mm, 4×) between PDMS surfaces. (a) Original image. (b) Binary image converted by MATLAB. (c) Identified inner and outer boundaries agree well with the original image. The width (w) of the meniscus was measured to be 37.9 μm, and the contact angle (CA) of the drop was calculated to be $\theta_C = 114°$.

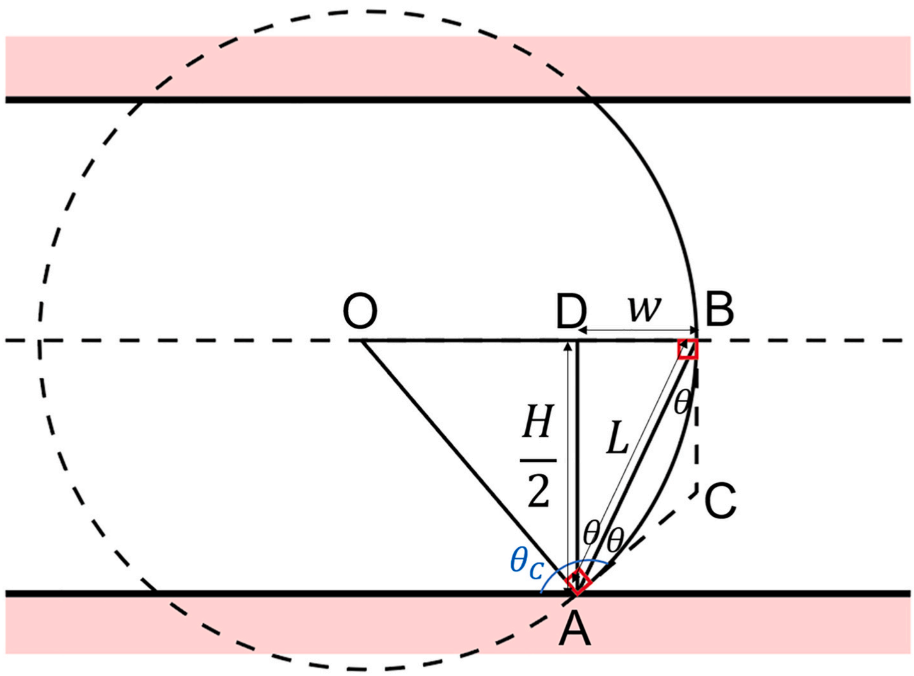

Fig. 3. The geometrical model for the meniscus profile of the drop in the Hele-Shaw cell.

Table 1
Summary of w and θ_C under different experimental conditions (DI water on PDMS).

H (mm)	4× lens		10× lens	
	w (μm)	θ_C (°)	w (μm)	θ_C (°)
0.17	18.5	115	20.7	117
	20.5	117	23.2	121
	23.2	121	23.2	121
	21.9	119	21.3	118
	23.5	121	19.1	115
	22.1	119		
	19.1	115		
0.36	37.9	114	35.6	112
	44.4	118	39.3	115
	32.9	111	24.2	105
0.57	47.4	109	33.2	103
0.74	72.8	112	61.0	109
1	86.9	110	87.6	110
	90.0	110	77.2	108

repellent-coated glass, glass slides were completely wetted by the water repellent, dried at room temperature for about 1 h, rinsed in water, and then dried using compressed air. Considering the dependence on H found in Fig. 4, the measurement was conducted with $H = 0.17$ and 1 mm. In the glycerol-on-PDMS case, θ_C was measured to be $109 \pm 2°$ ($N = 6$) with $H = 0.17$ mm, and $105 \pm 1°$ ($N = 10$) with $H = 1$ mm, while the reference value measured with the goniometer was $104 \pm 0.1°$ ($N = 3$). In the water-on-water-repellent case, θ_C was measured to be $110 \pm 1°$ ($N = 3$) with $H = 0.17$ mm, and $101 \pm 1°$ ($N = 6$) with $H = 1$ mm, while the reference value measured with the goniometer was $102 \pm 1°$ ($N = 3$). Both cases showed that the θ_C values measured with the proposed method decreased with H approaching the reference value.

The proposed method requires H to be small enough. With large H, the effect of the gravitational force on the meniscus becomes more significant, and thus the meniscus shape becomes asymmetric with respect to the mid-plane of the Hele-Shaw cell. This assumption of symmetric meniscus is valid when the characteristic dimension of the used Hele-Shaw cell (i.e., H) is much smaller than the capillary length ($[\gamma/\rho g]^{1/2}$) [13]. Here, γ is the surface tension coefficient between water and air (0.073 N/m), ρ the density of DI water (998 kg/m^3), and g the acceleration of gravity (9.8 m/s^2) [14]. Since the capillary length (= 2.7 mm) was much greater than H in this study, the gravitational force appeared negligible compared to the surface tension force, which supports the assumption that the meniscus of the water drop was symmetric in this study.

Even when the gravitational force is negligible, the meridional

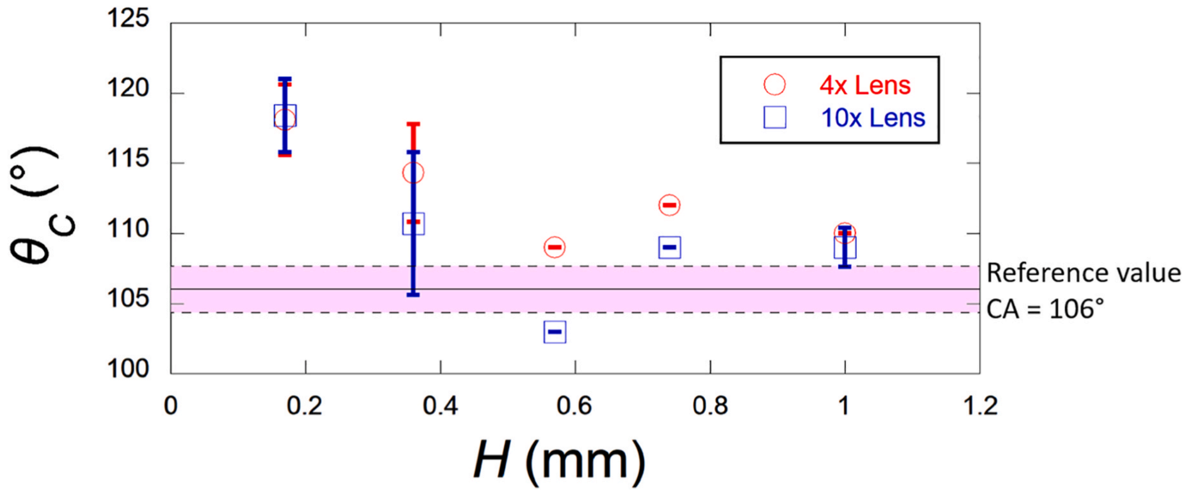

Fig. 4. Comparison of θ_C between the proposed method and the goniometer method (DI water on PDMS). Error bars and dashed lines show standard deviation values.

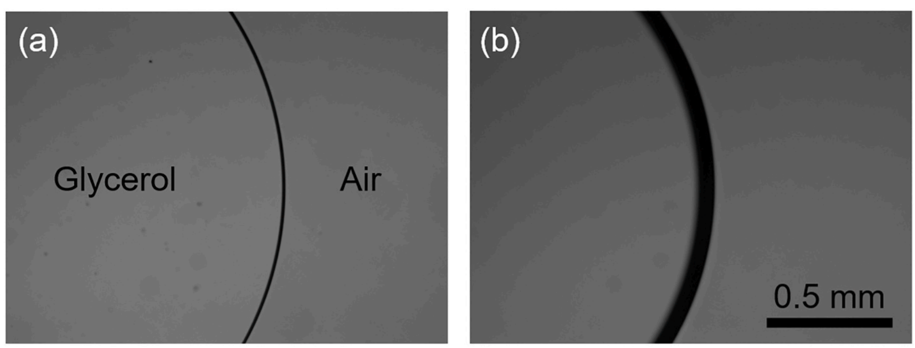

Fig. 5. Contact angle measurement of a glycerol drop on PDMS. (a) $H = 0.17$ mm, $w = 14.2 \pm 1.1$ μm, and $\theta_C = 109 \pm 2°$ ($N = 6$). (b) $H = 1$ mm, $w = 66.3 \pm 4.4$ μm, and $\theta_C = 105 \pm 1°$ ($N = 10$). Goniometer measurement: $\theta_C = 104 \pm 0.1°$ ($N = 3$).

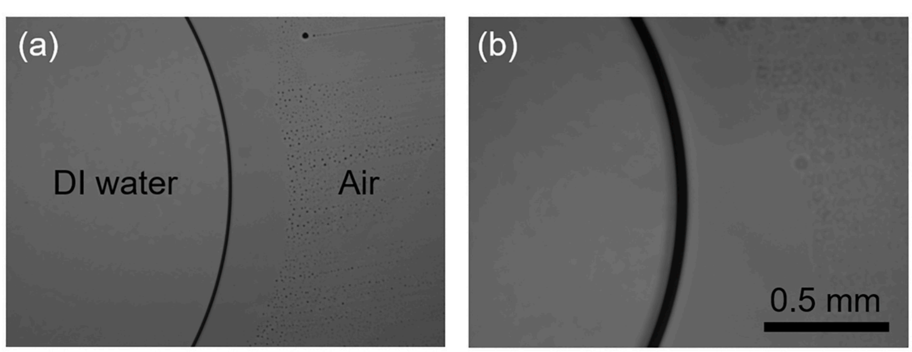

Fig. 6. Contact angle measurement of a DI water drop on water-repellant-coated glass. (a) $H = 0.17$ mm, $w = 15 \pm 1$ μm, and $\theta_C = 110 \pm 1°$ ($N = 3$). (b) $H = 1$ mm, $w = 48.2 \pm 2.7$ μm, and $\theta_C = 101 \pm 1°$ ($N = 6$). Goniometer measurement: $\theta_C = 102 \pm 1°$ ($N = 3$).

profile of the drop bridging two parallel planes is not circular. Since the exact meridional profile of the meniscus can be obtained by numerically computing the analytic equation of the profile [15], the profile is often assumed to be circular [16–19], which is also employed for the proposed method (Fig. 3). Hence, the accuracy of the proposed method depends on how the meniscus profile is approximated, and it is meaningful to discuss how well the circular approximation represents the exact profile of the meniscus. According to Megias-Alguacil and Gauckler [18], the difference in the radius of curvature between the circular approximation and the exact profile depends on θ_C and H. In particular, the difference decreases with increasing H for a constant θ_C value [18], which suggests that the θ_C dependence on H of the proposed method (Fig. 4) may be due to the circular approximation.

A major advantage of the proposed method is that it does not require special instruments. As long as a drop can be created between two parallel surfaces with a narrow gap (i.e., the Hele-Shaw cell) and its meniscus can be imaged in the top or bottom view, the method can enable dirty-and-quick measurements of contact angle. The proposed method has several limitations. First, tested solid surfaces must be transparent and smooth. Poor optical quality of a tested solid surface is expected to lower the quality of microscopy imaging and following image processing. For instance, when a water drop formed between

PDMS surfaces with roughness was imaged, its meniscus width could not be determined due to poor image quality. Second, the method requires the three-phase contact line and outermost part (or equator) of the drop to be imaged clearly in one image for reliable detection of the inner and outer boundaries of the meniscus image (Fig. 2). Third, the method seems to work better for larger CA cases (i.e., non-wetting cases). When the method was tried with small CA cases (i.e., wetting cases) such as mineral oil on PDMS, CA values were overestimated compared to reference values measured by the goniometer. Ray optics modeling of light trajectories passing through the meniscus is thought to be needed for successful application of the proposed method for wetting cases [9]. Last, the evaporation of a drop should be prevented. It was observed that when evaporation occurred during too slow growth of a drop or a prolonged experiment, the drop shrank gradually over time and the meniscus of the drop moved slowly, which resulted in non-circular or irregular three-phase contact lines.

4. Conclusion

In this study, a quick-and-dirty method for contact angle measurement was proposed, and its concept was proven in comparison with a conventional goniometer. In this method, a liquid drop was created in a Hele-Shaw cell of the known gap height, and a top or bottom view image of the meniscus of the drop was captured using a microscope. Then, the meniscus width of the drop was measured from the image using image processing. The contact angle of the drop was calculated from the meniscus width and the gap height of the Hele-Shaw cell with the meridional profile of the meniscus assumed to be circular. The proposed method was tested with two liquids (DI water and glycerol) and two solid surfaces (PDMS and water-repellent-coated glass) for a proof of concept. For tested gap heights, the proposed method resulted in higher contact angle values than the reference contact angle value measured with a goniometer, and the difference decreased with the gap height. More work is necessary in future to improve the accuracy of the method and to enable reliable applications of the method for lower contact angle cases.

Credit author statement

Haipeng Zhang: Methodology, Software, Validation, Formal analysis, Investigation, Writing - Original Draft, Writing - Review & Editing, Visualization **Jacob Gottberg:** Conceptualization, Methodology, Software **Sangjin Ryu:** Conceptualization, Methodology, Formal analysis, Resources, Writing - Original Draft, Writing - Review & Editing, Visualization, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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