Methods and preliminary measurement results of liquid Li wettability

G. Z. Zuo,[1,a)] J. S. Hu,[1] J. Ren,[1] Z. Sun,[1] Q. X. Yang,[1] J. G. Li,[1] L. E. Zakharov,[2] and D. K. Mansfield[2]
[1]*Institute of Plasma Physics, Chinese Academy of Sciences, Hefei 230031, China*
[2]*Princeton Plasma Physics Laboratory, MS-27 P.O. Box 451, Princeton, New Jersey 08543, USA*

(Received 11 September 2013; accepted 19 January 2014; published online 12 February 2014)

A test of lithium wettability was performed in high vacuum (< 3 × 10^{-4} Pa). High magnification images of Li droplets on stainless steel substrates were produced and processed using the MATLAB® program to obtain clear image edge points. In contrast to the more standard "θ/2" or polynomial fitting methods, ellipse fitting of the complete Li droplet shape resulted in reliable contact angle measurements over a wide range of contact angles. Using the ellipse fitting method, it was observed that the contact angle of a liquid Li droplet on a stainless steel substrate gradually decreased with increasing substrate temperature. The critical wetting temperature of liquid Li on stainless steel was observed to be about 290 °C. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4865118]

I. INTRODUCTION

Lithium (Li), especially flowing liquid Li, when employed on Plasma Facing Components (PFCs), has the potential to make significant improvements to plasma performance in fusion research devises.[1–4] In 2012, two types of flowing liquid Li limiters (FLLL) were employed in the HT-7 fusion device and encouraging results were obtained. During the course of that work, it was also found that the wettability of Li onto PFCs should be studied and possibly improved.[5]

The contact angle, i.e., the angle between the tangent of liquid at the contact point of a 3-phase (solid/liquid/gas) droplet and the baseline of a solid substrate, has normally been used to characterize wettability.[6,7] When the contact angle is higher/lower than 90°, the solid surface is said to be hydrophobic/ hydrophilic, and liquid wettability to the solid surface is characterized as bad/good. Therefore, a contact angle of 90° represents a critical state of wetting for a liquid droplet.

Several approaches to measuring contact angle have been reported in the literature. The most widely used techniques are the "θ/2" method as well as various curve fitting methods.[8,9] The "θ/2" method assumes that the droplet shape is part of a circle. By measuring the height of the droplet and the length between the two contact points, the contact angle can be estimated in an extremely simple manner. As compared to the "θ/2" method, ellipse[10] and polynomial fittings[6,11] are two more sophisticated and accurate methods to determine contact angle. In this work, comparisons of data processing and analysis for the "θ/2", ellipse and polynomial fitting methods are discussed in detail. This comparison was done in an effort to determine the "best" method to measure a wide range of Li contact angles—thus allowing Li wettability to stainless steel (SS) to be accurately characterized.

a)Electronic mail: zuoguizh@ipp.ac.cn

II. EXPERIMENTAL SET-UP

The experimental set-up used to measure liquid Li contact angles on a SS surface is shown in Fig. 1. It includes a large vacuum vessel containing a sample table with attached heater and thermocouple. A movable sample plate (150 × 80 × 3 mm) could be translated across the sample table—while maintaining good thermal contact—using a positioning system consisting of a bellows and a push-pull rod. An injection system was used for the controlled deposition of liquid Li droplets onto the sample plate. This system consisted of a liquid Li reservoir connected to delivery pipes with either 10 mm or 4 mm inside diameter.

Digital images of droplets were taken with a high resolution camera (5184 × 3456 pixels) under conditions of bright lighting. Reproducible positioning of the droplet in front of the camera was assured by mechanical means and a background plate positioned behind the sample plate ensured that high optical contrast images could be attained.

Before each measurement, the vacuum vessel was evacuated to < 3 × 10^{-4} Pa and the measured vessel leak rate was certified to be < 1 × 10^{-9} Pam^3/s. At the beginning of a measurement, the sample plate was inserted to a position directly under the outlet of the Li delivery pipe. Liquid Li droplets with volumes in the range 30−200 μL were injected onto the sample plate. This deposition took place when the temperature of the Li reservoir and the delivery pipe remained above 450 °C while substrate temperature was held at ∼180 °C—the melting temperature of Li. Following deposition of a droplet, the sample plate was pulled back into the center of the vacuum vessel and positioned directly and accurately in front of the camera. After allowing the Li droplet to cool on the substrate for ∼ 3−5 min, wetting tests began by gradually increasing the substrate temperature from ∼ 180 °C to ∼ 400 °C. As the substrate temperature changed, images of the droplet were taken about every 5–10 °C. Finally, these images were analyzed using the MATLAB® program.

Using MATLAB®, images underwent a grayscale transformation, binarization processing, and edge extraction by Prewitt filtering. With good optical contrast between the

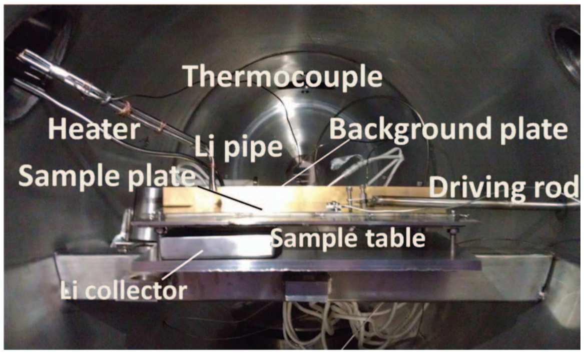

FIG. 1. Test equipment of liquid Li wettability.

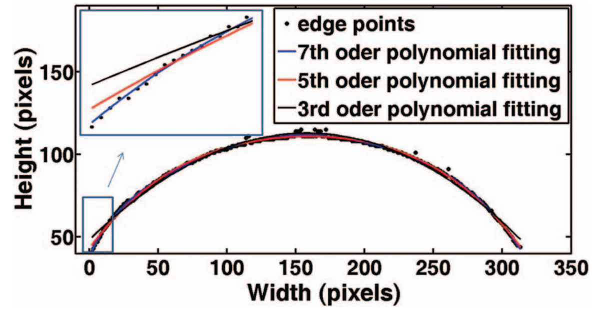

FIG. 3. 3rd, 5th, and 7th order polynomial fitting curves of edge points.

droplet edge and the background, the gray values of the first two background line arrays were averaged and divided by 255 to obtain a gray threshold. This threshold was used to get a binary image. At that point a Prewitt filter, used to detect vertical and horizontal edges, which were calculated by using difference between corresponding pixel intensities of an image, was used to detect edges in the image.

III. DATA PROCESSING AND ANALYSIS

A. Calculating the contact angle

1. Small contact angle (<90°)

a. "θ/2" method Figure 2 shows a raw high-magnification image of a liquid Li droplet with contact angle $< 90°$. The x and y coordinates of three points (i.e., left and right contact points and the top point of the liquid Li droplet) are indicated in the image. Using the "$\theta/2$" method, the contact angle can be determined as follows:

$$\theta = 2\arctan(\mathrm{h}/\mathrm{r})180/\pi, \quad (1)$$

where h and r are the height and half-width of the droplet, respectively. Thus $\mathrm{h} = \mathrm{Y_T} - (\mathrm{Y_L} + \mathrm{Y_R})/2$ and $\mathrm{r} = (\mathrm{X_R} - \mathrm{X_L})/2$.

Applying the measured coordinates to Eq. (1), the contact angle of the droplet was calculated to be $\theta = 49.1°$.

b. Curve fitting methods As shown in Fig. 3, the black quasi-circle represents the edge points after grayscale transformation, binarization, and Prewitt processing of the image shown in Fig. 2. The black, red, and blue curves are the 3rd, 5th, and 7th order polynomial fitting curves of the complete Li droplet shape. Obviously, the 7th order polynomial fitting appears to be optimal, and fits almost all edge points, as shown in the magnified curve of the left contact point.

As seen in Fig. 4, the red and blue curves represent the ellipse and 7th order polynomial fittings of the complete droplet shape. Both techniques appear to generate curves that fit the droplet image reasonably well. By enlarging the image of the left contact point, however, it was noted that the contact angle calculated by the ellipse fitting of the complete droplet shape had larger deviations in the region near the contact point as compared to the 7th order polynomial fitting.

The equations of the 7th order polynomial and ellipse fitting curves y(x) were readily generated by the MATLAB® program. Contact angles (θ) for both the left and right points of contact were calculated by the following equation:

$$\theta = \arctan\left(\frac{\mathrm{dy}}{\mathrm{dx}}\right)180/\pi, \quad (2)$$

where y(x) is the curve used to approximate the edge points of the Li droplet. According to the polynomial/ellipse fitting equations, $\left(\frac{dy}{dx}\right)_{\mathrm{left}} = 1.55/1.23$ and $\left(\frac{dy}{dx}\right)_{\mathrm{right}} = -1.22/-1.33$. The average contact angle of the liquid Li droplet calculated by 7th polynomial and ellipse fitting methods were 53.9° and 52.0°, respectively. So the contact angles calculated by the 7th polynomial and ellipse fitting of complete droplet shape differed by less than 2° as listed in Table I. This indicated that 7th polynomial fitting method was slightly

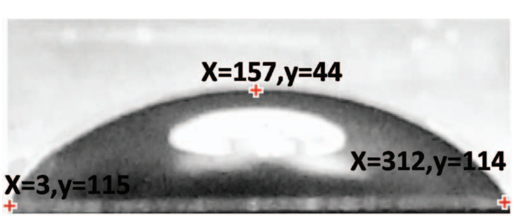

FIG. 2. Image of a liquid Li droplet including the coordinates of three points.

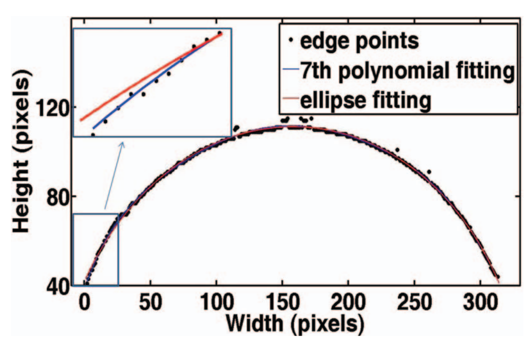

FIG. 4. Comparison curves generated by 7th order polynomial and ellipse fitting.

TABLE I. Comparison of calculation methods and angles.

Method	"$\theta/2$"	7th polyfit of entire shape	Ellipse fitting of entire shape
Contact angle	49.1°	53.9°	52.0°

more accurate than the ellipse method although both fitting methods were suitable to measure contact angles <90°.

2. Large contact angle (>90°)

a. "θ/2" method Figure 5 shows the image of a liquid Li droplet with contact angle >90°. Using the "$\theta/2$" method, the droplet contact angle was determined to be $\theta = 104.7°$.

b. Curve fitting methods In Fig. 6, red and blue lines represent the ellipse and 7th order polynomial fitting curves of Fig. 5 edge points, respectively. It can be seen that the 7th polynomial fitting of the complete droplet shape does not reproduce the entire edge region.

This illustrates a point about polynomial fittings in general. When the contact angle approaches 90°, the slope of the tangent near the contact point approaches infinity, so clearly 7th order polynomial fitting of the complete droplet shape can only fit droplet shapes with contact angle ≪ 90°—a severe restriction. In order to address this problem for contact angles > 90°, the region near the contact point was fitted instead with a 2nd order polynomial. This quadratic fitting is shown in the upper left corner of Fig. 6 and reproduces the droplet shape reasonably well. However, polynomial fitting of edge points is generally extremely sensitive to the resolution of image at the contact point, the number of edge points, and the order of the polynomial. Hence, under some conditions, these fittings can produce unacceptably large errors and are thus not considered viable options for data processing over a wide range of contact angles.

Conversely, the ellipse fitting shown in Fig. 6 appears to reproduce the complete droplet shape extremely well even though the contact angle > 90°.

Finally, the average contact angle calculated by ellipse fitting method was 112.5°, as listed in Table II.

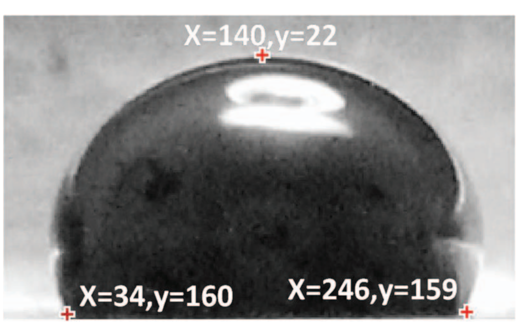

FIG. 5. Image of liquid Li droplet with contact angle >90°.

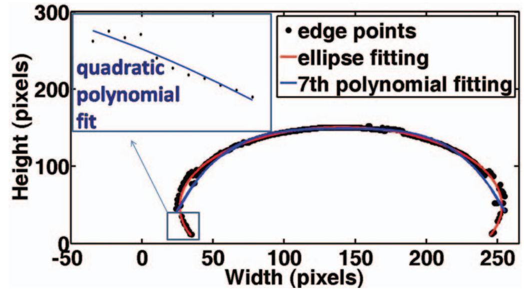

FIG. 6. Comparison of the two fitting curves.

B. Comparison the contact angle determined by the "θ/2" and ellipse methods

As discussed above, compared with polynomial fitting, ellipse fitting of the complete Li droplet shape can be used over a wider range to get reliable contact angle measurements. Hence in this section, the "$\theta/2$" method is compared against ellipse fitting method only.

The general equation of an ellipse is:

$$\frac{x^2}{a^2} + \frac{y^2}{b^2} = 1, \tag{3}$$

where a is the major axis and b the minor axis of the ellipse. Here, a ≥ b, and x = acosβ while y = bsinβ. The tangency slopes k_1 and k_2 of an ellipse and a circle, respectively, can be expressed as

$$k_1 = \tan\alpha = -\frac{b^2}{a^2} \cdot \frac{x}{y} = \frac{-b\cos\beta}{a\sin\beta}, \tag{4}$$

$$k_2 = \tan(\theta/2) = \frac{b-y}{-x} = \frac{b-b\sin\beta}{-a\cos\beta}, \tag{5}$$

where α and θ are the contact angles calculated using ellipse and circle fittings ("$\theta/2$" method), respectively. The difference in contact angles ($\alpha-\theta$) is plotted against α in for different values of b/a in Fig. 7. It can be seen that the difference is small at low values of α but reaches a maximum at about $\alpha = 130°$. Also the maximum of contact angle difference increases strongly with decreasing values of b/a. These trends indicate that for a fixed b value (i.e., fixed height of Li droplet), larger volume Li droplets (larger a) would have larger contact angle differences ($\alpha-\theta$). Hence, when associated with large-scale Li droplets and at large contact angles, measurements of contact angles determined from the "$\theta/2$" method can be expected to be less accurate than those determined by ellipse fittings.

TABLE II. Comparison of calculated method and contact angle.

Method	"$\theta/2$"	2nd polyfit of edge points	Ellipse fitting of entire shape
Contact angle	104.7°	113.0°	112.5°

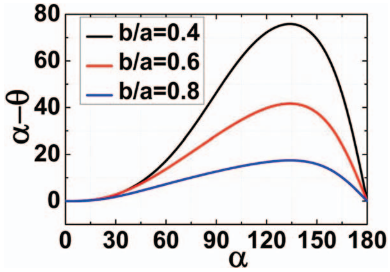

FIG. 7. Comparison of angle differences with different b/a values.

Therefore, wettability data described below were analyzed using ellipse fitting because the range of measured contact angles and the range of droplet sizes were both large.

C. Preliminary test results of Li wettability

Both a large and a small Li droplet were successively placed on SS substrate after 2 days of baking the vacuum vessel with a temperature ∼ 300 °C. Both droplets were analyzed contemporaneously. As shown in Fig. 8, below ∼ 250 °C, the contact angle, calculated using the ellipse fittings, exhibited little change with increasing SS substrate temperature. Above ∼ 250 °C, the contact angle gradually decreased with increasing temperature. The critical wetting temperature of liquid Li droplets was found to be ∼ 290 °C. It is also clear that the initial contact angle of the large Li droplet was smaller than that of small droplet due to the larger effect of gravity on a large droplet. As the contact angles decreased, the droplet heights decreased and the effect of the gravity also decreased. The difference in contact angles between large and small Li droplets therefore decreased gradually with substrate temperature. Hence, to obtain more accurate contact angle data, it appears that smaller scale Li droplets should be selected.

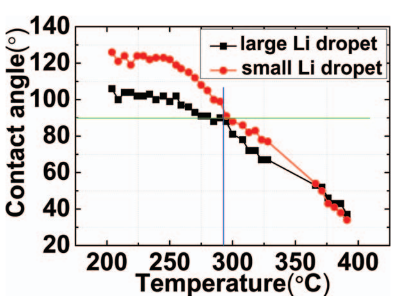

FIG. 8. Test results of contact angles with large and small scale liquid Li droplets using the ellipse fitting method.

IV. CONCLUSIONS

Both of "$\theta/2$" method and the curve fitting methods are based on the image acquisition and processing technology, so the large test error might result from the size of Li droplet, the image resolution, optical contrast between the droplet edge and the background, data processing methods, and so on. For liquid Li droplets with volumes of 30−200 μL, the contact angle calculated by "$\theta/2$" method produced large errors, especially at large contact angle. Using 7th order polynomial fitting of the complete droplet shape produced a satisfactory curve of Li droplet profiles only for contact angle $\ll 90^\circ$. Using 2nd order fitting in the neighborhood of the contact point yielded a reasonable result for angles $> 90^\circ$. By comparison, however, ellipse fittings of the complete droplet shape could produce accurate measurements over a wide range of contact angles. Using the ellipse fitting method, it was found that the contact angle of liquid Li droplets gradually decreased with increasing SS substrate temperature. The measured critical wetting temperature of liquid Li on SS was ∼290 °C. Hence, as guidance to the FLLL experiment mentioned above, it appears that before injecting liquid Li onto a SS limiter, the temperature of limiter should be higher than 290 °C.

ACKNOWLEDGMENTS

This research was funded by the National Magnetic confinement Fusion Science Program under Contract Nos. 2013GB114004 and 2011GB10700, the National Nature Science Foundation of China under Contract Nos. 11075185 and 11021565, and the JSPS-NRF-NSFC A3 Foresight Program in the field of Plasma Physics (NSFC No. 11261140328).

[1]R. Majeski, R. Doerner, T. Gray, R. Kaita, R. Maingi, D. Mansfield, J. Spaleta, V. Soukhanovskii, J. Timberlake, and L. Zakharov, Phys. Rev. Lett. **97**, 075002 (2006).
[2]G. S. Xu, B. N. Wan, J. G. Li, X. Z. Gong, J. S. Hu, J. F. Shan, H. Li, D. K. Mansfield, and D. A. Humphreys, V. Naulin, for EAST Team and International Collaborators, Nucl. Fusion **51**, 072001 (2011).
[3]G. Z. Zuo, J. S. Hu, S. Zhen, J. G. Li, D. K. Mansfield, B. Cao, J. H. Wu, and L. E. Zakharov, Plasma Phys. Controlled Fusion **54**, 015014 (2012).
[4]J. P. Allain, M. Nieto, M. D. Coventry, R. Stubbers, and D. N. Ruzic, Fusion Eng. Design **72**, 93 (2004).
[5]J. Ren, J. S. Hua, and G. Z. Zuo, Phys. Scr. "First Results of Flowing Liquid Lithium Limiter in HT-7" (unpublished).
[6]A. Bateni, S. S. Susnar, A. Amirfazli, and A. W. Neumann, Colloids Surf., A **219**, 215 (2003).
[7]Jose Bico, Uwe Thiele, and D. Q. re, Colloids Surf., A **206**, 41 (2002).
[8]E. Mooy, P. Chang, Z. Policova, S. Treppo, D. Kwok, D. R. Mack, P. M. Sherman, and A. W. Neumann, Colloids Surf. **58**, 215 (1991).
[9]R. Y. Tsay, S. C. Yan, and R. S. Y. Lin, Rev. Sci. Instrum. **66**, 5065 (1995).
[10]Z. N. Xu, IEEE Trans. Power Delivery **28**, 491 (2013).
[11]O. I. del Rıo, D. Y. Kwok, R. Wu, J. M. Alvarez, and A. W. Neumann, Colloids Surf., A **143**, 197 (1998).