Dear Referee

We would like to thank you for your valuable and constructive comments to greatly improve the manuscript *micromachines-606855*. The manuscript has been carefully revised according to your suggestions.

The changes that refer to your comments are highlighted in green in the attached modified version. The content of manuscript is written in italics, the answers of authors are written in standard style.

**Comment 1:** The starting equation, Equation (1), is WRONG. Surface forces account as a boundary condition and cannot be mixed up with the equation describing the bulk of liquid (i.e. Navier-Stokes equation). I dug in the reference paper cited by the authors to support their claim (ref 20). In the present paper, no transition zone is defined, to equivalence check is done, and even worse the pressure force is not even correctly written.

**Answer:**

Thank you for your constructive comments to greatly improve the model of droplet squeezing-and-deforming. Based on your comments, we analyzed and improved the model.

After analysis, we found that the continuum surface tension (CSF) model proposed by Brackbill et al. uses mainly in computational fluid dynamic method. But here the CSF is not suitable. Thus, followed your suggestion, we accounted surface tension force as a boundary condition to analyze the contribution of surface tension force and viscous force in the squeezing process. Moreover, the corresponding pressure distribution function has also been corrected.

Modified in the Eq. (1)–(6) of section 2.2.
**Comment 2:** L 107: you write that the motion of the droplet should be a low speed flow and make the association with the mass conservation equation. Which was already evoked L 103 under the approximation of incompressibility.

**Answer:**

Thanks to the referee for careful reading and pointing out the problem that the condition of simplifying the model is reused. The revised sentences are as follows: “(1) The liquid-bridge is axisymmetric and incompressible low-speed flow (\( \frac{\partial p}{\partial \theta} = 0 \), \( \rho \vec{u} \cdot \vec{u} + \rho \vec{u} \cdot \nabla \vec{u} = 0 \));” Modified in the L107~L108 of section 2.2.

**Comment 3:** The definition of the surface \( S_m \) is WRONG. You do not take into account the inclination toward the z-direction.

**Answer:**

With the improved model, it is no longer necessary to calculate the surface area \( S_m \), thus it has been deleted.

**Comment 4:** L170 The reader does not understand what is meant by contact force. Is it the force at rest? at minimal position? Nothing is given to reproduce the experiment.

**Answer:**

According to the suggestion of referee, we redefined the contact force and explain the variation of contact force to allow others to reproduce the experiment.

Modified in the P2 L90~L93 L95~L96 of section 2.1. “In squeezing stage, the stamp with initial droplet is positioned above a substrate. Then it slowly moves down to contact and squeeze the substrate, during which the contact force \( F_C \) is generated by the squeezing effect on the substrate. Thus the \( F_C \) is defined as the force of the droplet acting on the substrate and measured with a force sensor.” …… “Once the contact force \( F_C \) reaches a defined threshold (critical contact force \( F_{CC} \)), the stretching stage is triggered and the stamp is lifted up until the liquid-bridge is pulled off.”

In addition, in the experiment process, the effect of the contact force is further explained, taking the critical contact force of 1 mN as an example.

Modified in the P1 L170~L172 of section 3.2. “(d) The initial adhesive droplet is squeezed under the increasingly contact force \( F_C \), during which a liquid-bridge is formed and deformed; (e) Until \( F_C \) reaches a defined value of 1 mN (\( F_{CC} \)), the
Comment 5: L181 Equation 8 does not make sense, what is \( n \)? the pixel position? Authors tell about the physical scale \( x \) but no link is given with the volume computation.

Answer:

According to the recommendations of referee, the calculation formula is re-expressed, and the calculating process is elaborated based on the formula.

The calculation process is as follows: Firstly, initial and transferred droplets in the image can be divided into \( n \) micro-cylinders with the height of one unit pixel \( h_p \) from top to bottom by MATLAB. Secondly, the unit pixel \( h_p \) is equivalent to 1.9 \( \mu \)m via spatial calibration. Thirdly, \( x_i \) \((i=1, 2, \ldots n)\) is defined as the radius of the \( i \)-th micro-cylinder. Since the first micro-cylinder \((i=1)\) is in contact with the substrate, the contact area is \( S_{w-z} = \pi x_i^2 \). At last, the sum of all micro-cylinder volumes is the volume of the initial or transferred droplet \( V = \sum_{i=1}^{n} \pi x_i^2 h_p \).

Modified in the P2 L186~L189 of section 3.3. “Their volumes \( V \) and contact areas \( S_a-s \) could be obtained by image characteristic extraction and calculation with Eq. 7.

\[
V = \sum_{i=1}^{n} \pi x_i^2 h_p \quad (i=1, 2, 3\ldots); \quad S_{w-z} = \pi x_i^2
\] (7)

where the initial and transferred droplets in the image can be divided into \( n \) micro-cylinders with the height of one unit pixel \( h_p \) from top to bottom. The unit pixel \( h_p \) is equivalent to 1.9 \( \mu \)m via spatial calibration. \( x_i \) is the radius of the \( i \)-th micro-cylinder. The first micro-cylinder \((i=1)\) is in contact with the substrate, and so on.”

Comment 6: contact angles \( \alpha_i \) and \( \beta_i \) are not defined except in figure 6.

Answer:

In addition to the variation of contact angles \( \alpha_i \) and \( \beta_i \) as depicted in Figure 6b, we firstly defined the contact angles \( \alpha_i \) and \( \beta_i \) in the squeezing-and-deforming model of initial droplet (section 2.2). Next, the static contact angles \( \alpha_0 \) and \( \beta_0 \) were given in
The relevant sentences are corrected in the L109–L110 of section 2.2. “(2) The wettability of stamp is similar to that of substrate, thus the contact angle $\alpha_i$ on the needle-stamp is approximately equal to $\beta_i$ on the substrate, and the meniscus of liquid-bridge is assumed to be the concave surface of spherical cap, shown in Figure 2;”

### Table 1. Properties of adhesive.

<table>
<thead>
<tr>
<th>Surface Tension (mN/m)</th>
<th>Contact Angle $\alpha_0$ on Steel ($^\circ$)</th>
<th>Contact Angle $\beta_0$ on Silicon Wafer ($^\circ$)</th>
<th>Viscosity $\mu$ (Pa·s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>36.96</td>
<td>73.54</td>
<td>70.65</td>
<td>93.93</td>
</tr>
</tbody>
</table>

**Comment 7:** We do not know if Figure 7 is obtained by numerical computation of physical measurement.

**Answer:**

According to the improved model, the graphs in Figure 7 are solved again by MATLAB and the solving process is as follows: Firstly, as the contact force $F_C$ increases from 0 to 5 mN, the slit pitch $h_i$ is defined as a variable that decreases from 42.0 to 12.3 $\mu$m. Secondly, the parameters that are used to solve the graphs are given as shown in Table 1, Table 2 and Figure 6b. These parameters are assigned and defined as the MATLAB program is written. Thirdly, since the $F_C$ consists of viscous force $F_v$ and surface tension force $F_s$, the equations for $F_v$ and $F_s$ are listed and solved respectively. Finally, the graphs are obtained by the plot $(h_i, F_v)$ and plot $(h_i, F_s)$ functions.

$$
F_v = -\frac{6\pi\mu U}{h_i^3} \left[ \frac{r_i^4}{4} - \frac{r_i^4}{2} - r_i \left( \frac{h_i}{2\cos\alpha_i} - \frac{h_i\tan\alpha_i}{2} \right) \right]
$$

$$
F_s = -\pi r_i^2 \left( \frac{1}{r_i} + \frac{1}{h_i / 2\cos\alpha_i} \right)
$$

**Comment 8:** L187 authors told about picoliter droplet but reports values in the order of 70 picoliters in figure 6.

**Answer:**

The focus of this manuscript are sub-nanoliter level. The purpose of Figure 6a is
to show that the measuring error of transferred droplet instead of the minimum amount of transfer. The measuring difference of one picoliter is obtained by comparison of two methods. That is, the transferred volumes (a set of five droplets) measured by optical imaging are verified with a confocal laser scanning microscope (CLSM, VK-X200, Keyence, Japan). It demonstrated that the measured method with optical imaging is considered to be accurate and reliable for sub-nanoliter droplets.