Supporting Information

Photothermally Caused Propylene Glycol-Water Binary Droplet Evaporation on a Hydrophobic Surface

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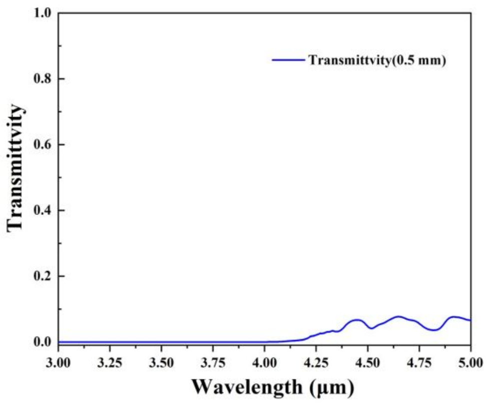
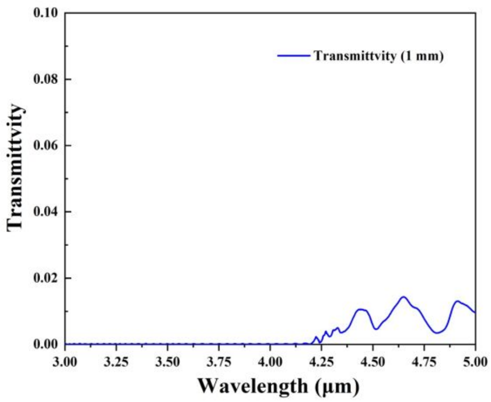
Droplet Interface Temperature Measurement by an Infrared Camera

For the measurement of the droplet interface temperature by an infrared camera, the determination of emissivity is quite important, which can be affected by the variations of the droplet concentration, droplet height, droplet temperature and the reflection of surrounding objects. In this work, the emissivity of the binary droplet was fixed at 0.95 for all cases, and the reasons are given as follows.

The optical properties are needed to determine the global emissivity. Thus the spectral transmittivity of propylene glycol was firstly measured by the Thermo Nicolet iS5 FT-IR spectrophotometer at a given liquid thickness of 0.5 mm in the spectral range of 2.5-25 μm. The result is given in **Figure S1a**. Here, only 3-5 μm spectral range was extracted to analyze because of the spectral range of infrared camera. As seen, for the propylene glycol with a thickness of 0.5 mm, the transmittivity in the 3- 4.1 μm range is negligible and the maximum monochromatic transmittivity was less than 8%. The global transmittivity τ is about 5% after integration on the 3-5 μm spectral range. The global emissivity of propylene glycol was then estimated to be about 0.95 at a thickness of 0.5 mm with the hypothesis of non-scattering liquid.[1] Therefore, propylene glycol is a semi-transparent in the given spectral range. With the increase of the thickness, the transmittivity decreases and the emissivity of propylene glycol increases. To verify this point, the monochromatic transmittivity of propylene glycol at the thicknesses of 1 mm in the 3-5 μm spectral range was also obtained, as shown in **Figure S1b**. The corresponding global transmittivity is less than 0.6%, indicating that the propylene glycol at this thickness (1 mm) can be regarded to be opaque in this spectral range of infrared camera.



(a)

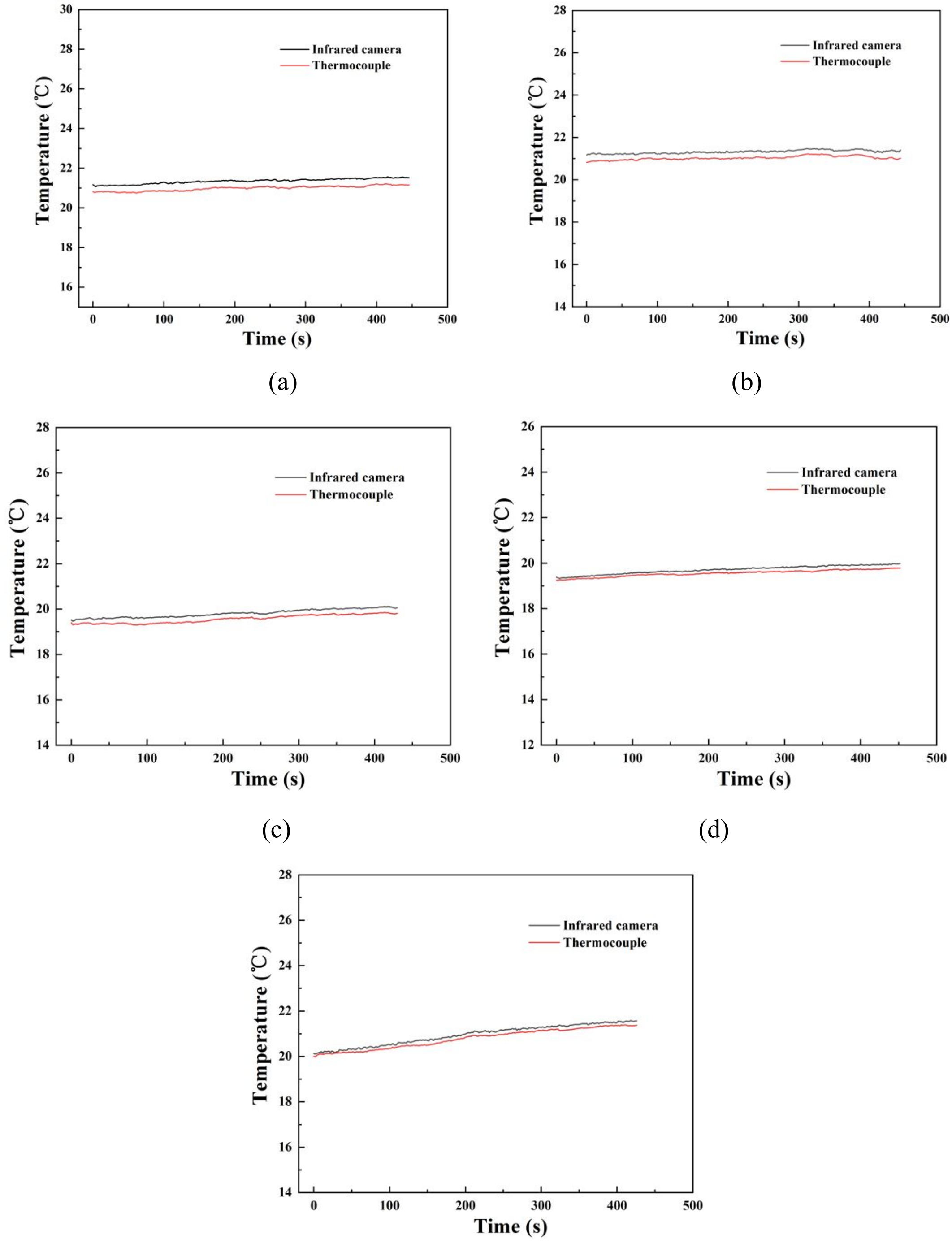


(b)

Figure S1. Spectral transmittivity of propylene glycol in the spectral range of 3-5 μm for liquid thickness of (a) 0.5 mm and (b) 1 mm at ambient temperature of ~25 ℃.

In this study, the concentrations can change with the laser heating time due to the evaporation. For this reason, the global emissivity may change with time. To determine the emissivity of the binary liquid with different concentrations, the temperatures of the liquid-air interface of a liquid bath with 20 mm in thickness were measured by using a type-K miniature thermocouple with a diameter of 0.1 mm (Kaipusen, WRNT-01), which were compared with the temperatures derived from the IR images. We first compared the infrared visualization result with that measured by a thermocouple in case of pure water at ~20 ℃, because the emissivity of deionized water is well-known to be 0.95. As shown in **Figure S2a,** when the emissivity was set to 0.95, the variation of the results by the infrared visualization and thermocouple displayed the same tendency and the infrared visualization result was just slightly 0.35 ℃ higher than that measured by the thermocouple, demonstrating the accuracy of the IR camera measurement (±2 ℃).

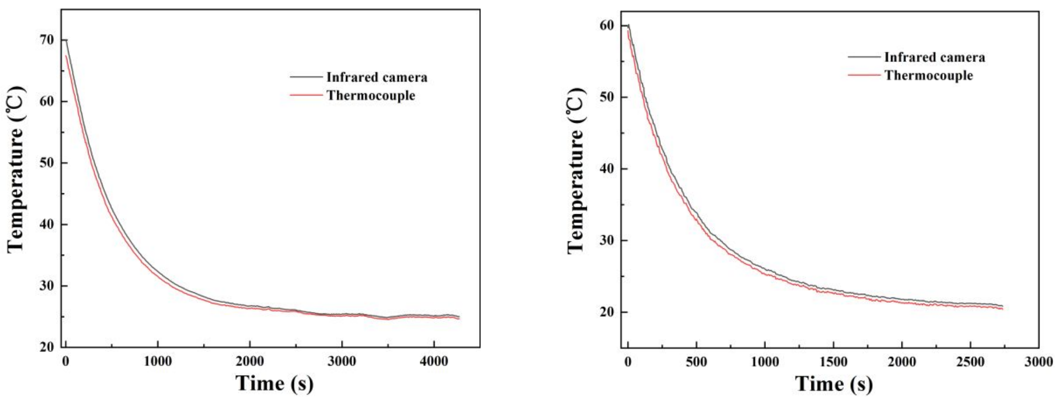
For the cases of the PG-water binary liquid (25%, 50% and 75%) and pure propylene glycol, the emissivity was still kept at 0.95. The comparison between the infrared visualization and thermocouple results are given in **Figure S2b-e**. As shown, the corresponding temperature differences were 0.29℃, 0.24℃, 0.15℃ and 0.16℃, respectively. These results implied that although the emissivity of the binary liquid changed with the PG content, the temperatures measured by an infrared camera were still highly consistent with those measured by the thermocouple. Therefore, the influence of the change of the emissivity with the composition on the temperature measurement by the IR camera is negligible when the emissivity was fixed at 0.95.



(e)

Figure S2. The liquid-air interface temperature of a 20-mm liquid bath measured by an infrared camera and a thermocouple with different propylene glycol contents: (a) pure water, (b) 25% propylene glycol, (c) 50% propylene glycol, (d) 75% propylene glycol and (e) pure propylene glycol.

Besides, we also measured the interface temperatures by an IR camera and a thermocouple during the cooling process of the liquid pool to investigate the influence of the interface temperature on the emissivity. Here, the emissivity was fixed at 0.95. The results are compared in **Figure S3a-b**. As shown, for the case of pure water and pure propylene glycol, the temperature difference between the infrared visualization and thermocouple results was quite small. Moreover, the cooling process of a binary liquid with 50% PG content was measured. As shown in **Figure S4**, despite the temperature and concentration changed with time, the temperatures measured by the IR camera and thermocouple were still quite similar. Therefore, the emissivity of 0.95 used for measuring the interface temperature during the PG-water binary droplet evaporation is acceptable with sufficient accuracy.



(a) (b)

Figure S3. The liquid-air interface temperature of a 20-mm liquid bath measured by an infrared camera and a thermocouple during the cooling process: (a) pure propylene glycol and (b) pure water.

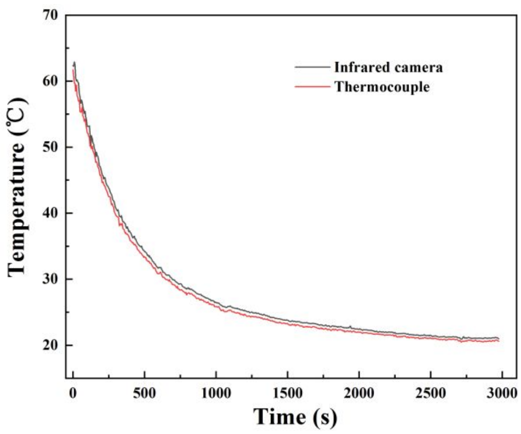
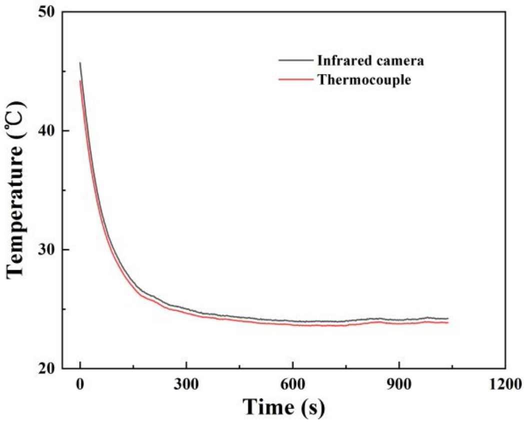
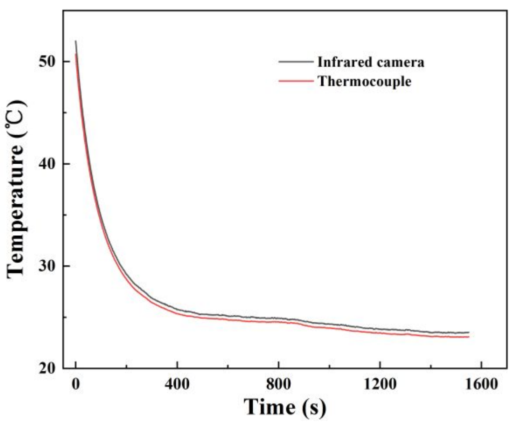


Figure S4. The liquid-air interface temperature of a 20-mm liquid bath of 50% propylene glycol measured by an infrared camera and a thermocouple during the cooling process.

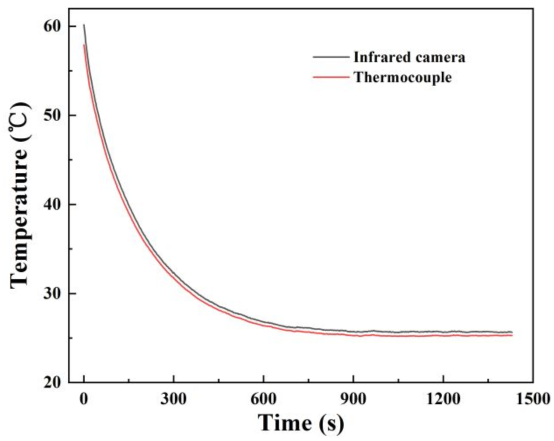
As mentioned above, we used the Thermo Nicolet iS5 FT-IR spectrophotometer to measure the spectral transmittivity of propylene glycol at a given liquid thickness in the spectral range of 2.5-25 μm. The results are given in **Figure S1a** and **S1b**. It was found that with the increase of the thickness of pure propylene glycol from 0.5 mm to 1 mm, the propylene glycol became more opaque and the emissivity increased. In this work, the emissivity was kept at 0.95 throughout the evaporation process. This is because when the liquid thickness was larger than 0.4 mm, the influence of the liquid thickness on the temperature measurement was negligible. Actually, we have also compared the infrared visualization results with the thermocouple measured temperatures for different liquid thicknesses (0.4 mm, 1 mm, 2 mm) during the cooling process of propylene glycol. As shown in **Figure S5a-c**, for all these three cases, the temperatures measured by the IR camera and thermocouple were also highly consistent. This fact indicated that although the emissivity was fixed at 0.95 in this work, the measured interface temperature by the IR camera was still satisfactory to reflect the interface temperature with sufficient accuracy.



(a)



(b)



(c)

Figure S5. The liquid-air interface temperature of a PG liquid bath measured by an infrared camera and a thermocouple during the cooling process under different thicknesses: (a) 0.4 mm, (b) 1 mm and (c) 2 mm.

In this work, the glass slide covered with transparent thin film of amorphous fluoropolymers layer was used as the hydrophobic surface. Therefore, the glass reflection was required to take into consideration because the glass reflection may influence the emissivity. Actually, the emissivity is the highest when the camera is normal to the surface, and decreases with the increase of the angle of incidence (refer to http://www.flirmedia.com/MMC/THG/Brochures/T820325/T820325_EN.pdf). In this work, an infrared camera was utilized to capture and analyze the evolution of the droplet interface temperature at an angle of incidence of 20° normal to the substrate. However, the glass slide covered with transparent thin film of amorphous fluoropolymers layer inevitably influenced the thermal image because of the reflection. When the IR camera was normal to the glass substrate, the glass reflection was the greatest, a shadow of IR camera appeared at the thermal image, which changed the measured temperature distribution. When the infrared camera was set at an angle of incidence of 20° normal to the substrate, this undesirable shadow disappeared. Therefore, in order to weaken the effect of glass reflection on the IR camera, an angle of 20° normal to the substrate was applied. Besides, to further understand the influence of reflection of the surrounding objects on the temperature readings of the thermal image under the experimental conditions, an opaque tape with a tiny circular hole was used to cover the hydrophobic surface to minimize the reflection, and the circular hole was used to deposit the droplet.[2] A uniform substrate heating was applied to the 6 μL PG droplet at the wall temperature of 36 ℃, and the emissivity was maintained at 0.95. Once the droplet temperature was stable, the average minimal temperature of the droplet interface was extracted. It was found that the difference of the average minimal temperature with or without the tape was less than 0.2 ℃. Moreover, the temperature of the evaporating droplet was much higher than the surrounding. These results indicated that the influence of the glass reflection could be negligible in this work.

In summary, fixing the emissivity at 0.95 in this study is acceptable with sufficient accuracy to measure the interface temperature by the infrared camera during the binary droplet evaporation process for all cases.

References

(1) Brutin, D.; Sobac, B.; Rigollet, F.; Le Niliot, C. Infrared Visualization of Thermal Motion inside a Sessile Drop Deposited onto a Heated Surface. *Exp. Therm. Fluid Sci.* **2011,** 35, 521-530.

(2) Katre, P.; Gurrala, P.; Balusamy, S.; Banerjee, S.; Sahu, K. C. Evaporation of Sessile Ethanol-Water Droplets on a Critically Inclined Heated Surface. *Int. J. Multiphase Flow* **2020,** 131, 103368.