## **Electronic supporting information**

# Investigation of droplet coalescence in nanoparticle suspensions by a microfluidic collision experiment

Qianqian Zhou, Yue Sun, Shiting Yi, Kai Wang\*, Guangsheng Luo

The State Key Lab of Chemical Engineering, Department of Chemical Engineering, Tsinghua University, Beijing 100084, China

#### Contact angles on nanoparticle tablets

To evaluate the wetting properties of nanoparticles, the contact angles of water phase on nanoparticle tablets were measured using the pendant droplet tensiometer (OCAH200, DataPhysics Instruments GmbH, Germany), which has an image analysis function for measuring contact angles from a sessile droplet on a plate. The tablets were made using a high-pressure molding machine commonly used for preparing KBr plates for infrared spectroscopy (IR) experiments. The operated pressure was 10 MPa at 25 °C. Fig. S1 shows some pictures of sessile droplets and the scanning electron micrographs of the tablet surfaces.

<sup>\*</sup>Corresponding author email: <u>kaiwang@tsinghua.edu.cn</u>



Fig. S1. Pictures of contact angle experiment.

(a)–(d) Sessile droplets on the particle tablets; (e) and (f) SEM images of the surface of 210 nm and 306 nm particle tablets. The scale bars show 1  $\mu$ m.

#### Surface elemental analysis

To investigate the reasons behind the different wetting properties of polystyrene (PS) nanoparticles, X-ray photoelectron spectroscopy (XPS) analysis was conducted for all the particles used in this study. The results (Table S1) show that the 158 nm particles contain 6.31 at % nitrogen on their surfaces. These nitrogen-containing hydrophilic groups may have originated from the initiator or surfactant used during the styrene polymerization process.

Particles	158 nm	179 nm	210 nm	306 nm
C (at %)	87.06	92.81	92.10	90.67
N (at %)	6.31	1.55	1.17	0.64
O (at %)	6.62	5.64	6.73	8.69

Table S1. Surface elemental analysis for nanoparticles.

### **Droplet velocity analysis**

A self-made MATLAB program was used to measure the relative velocities of droplets in the microchannel for calibrating the dimensionless droplet mobility coefficient  $\beta$  in the equation  $u = 2\beta Q/S = 2\beta u_{av}$ , where u is the droplet relative velocity; Q is the total flow rate; S is the section area of the entry channel of the collision chamber; and  $u_{av}$  is the average velocity of two phases. This program can recognize the centers of the droplets, as shown in Fig. S2a, and hence, the distance between the centers of two droplets was measured. Several consecutive frames were selected as the time intervals, and the center distance variations during those intervals were measured, so that the droplet relative velocity could be estimated. Using the measured droplet relative velocity and the total flow rate Q,  $\beta$  was calibrated. As shown in Fig. S2b, a control experiment with a blank continuous phase and an experiment with 210 nm particles at  $c_2$  concentration ( $3.22 \times 10^{10}$  mL<sup>-1</sup>) was used for the calibration. The measured velocities were organized along a straight line, from which  $\beta$  could be estimated as 1.57.



Fig. S2. Calibration of droplet mobility coefficient  $\beta$ 

(a) MATLAB program analysis. (b) Relationship between measured droplet velocities and two-phase average velocity  $u_{av} = Q/S$ . Control experiment (black squares) is quite in accordance with the experiment using 210 nm particles.  $\beta$  is determined by measuring the half slope of the fitting line.

#### Videos for images in Figs. 3 and 8

Video A: Fig. 3a, recorded at  $Q_W = 5 \mu L/min$ ,  $Q_{A1} = 10 \mu L/min$ ,  $Q_{A2} = 50 \mu L/min$ , 306 nm particles with  $c_1$  concentration.

Video B: Fig. 3b, recorded at  $Q_W = 5 \ \mu L/min$ ,  $Q_{A1} = 10 \ \mu L/min$ ,  $Q_{A2} = 50 \ \mu L/min$ ,

306 nm particles with  $c_1$  concentration.

Video C: Fig. 3b, recorded at  $Q_W = 8 \mu L/min$ ,  $Q_{A1} = 16 \mu L/min$ ,  $Q_{A2} = 25 \mu L/min$ , 306 nm particles with  $c_1$  concentration.

Video D: Fig. 3b, recorded at  $Q_W = 8 \mu L/min$ ,  $Q_{A1} = 16 \mu L/min$ ,  $Q_{A2} = 50 \mu L/min$ , 306 nm particles with  $c_1$  concentration.

Video E: Fig. 8a, recorded at  $Q_W = 5 \mu L/min$ ,  $Q_{A1} = 10 \mu L/min$ ,  $Q_{A2} = 10 \mu L/min$ , 179 nm particles with  $c_3$  concentration.

Video F: Fig. 8b, recorded at  $Q_W = 5 \mu L/min$ ,  $Q_{A1} = 10 \mu L/min$ ,  $Q_{A2} = 10 \mu L/min$ ,

179 nm particles with  $c_3$  concentration.