**Supporting information for Lab on a Chip**

# Microfluidic Preparation of Flexible Micro-Grippers with Precise Delivery Function

## Fabrication of the microfluidic device

A T-junction and a cross junction were fabricated on a 20 mm*20 mm*3 mm and a 60 mm*20 mm*3 mm polymethyl methacrylate (PMMA) chip respectively using an Φ1.5mm end mill. The junction was approximately 1.5 mm wide * 1.5 mm depth. The gas phase fluid was introduced through the circular glass capillary with 0.7 mm inner-diameter and 1.00 mm outer-diameter, which was tapered to about 10 μm using a micropipette puller (P-97, SUTTER Co. Ltd., USA). A circular glass capillary with 1.05 mm inner-diameter and 1.50 mm outer-diameter was tapered to about 100 μm for the injection of the aqueous phase fluid, namely the middle phase fluid. The thinner tapered capillary was inserted into the wider capillary consisting a tube in tube top end, so the aqueous phase fluid was injected from the T-junction and flow through the annulus space. The top end was inserted into a third capillary for the oil phase at the cross junction area and coaxality was guaranteed by matching the outer diameter of the capillary with the inner dimension of the channel. The third capillary wasn't tapered. Three PTEE pipes were inserted into sides of the chip channel for carrying middle and outer phase fluids, and a PTEE pipe was cover the thinnest capillary for carrying inner phase fluid. Three micro-syringe pumps (PHD Ultra, Harvard Apparatus Co., Ltd) were used to pump the three-phase fluids into the micro-fluidic device respectively. The complete microfluidic device is shown in Figure S1.

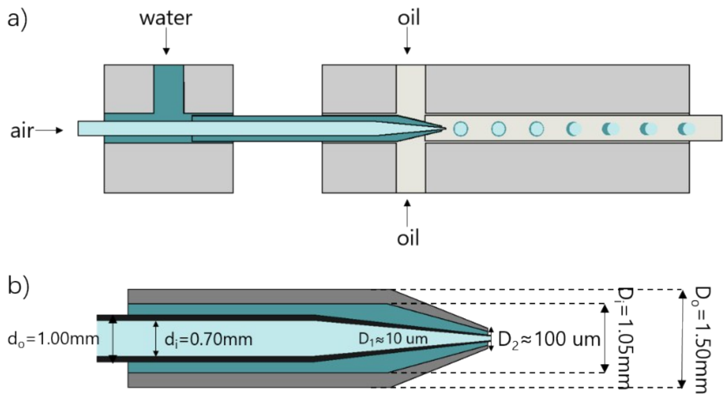


Fig.S1 Microfluidic device composing of two coaxial micro capillaries. a) The inner flow is air, the middle flow is aqueous solution with $Fe_3O_4$ nanoparticles and the outer flow is the mixture of soybean oil and n-tetradecane. b) The diameters of two sizes of capillaries and the size of tapered end.

## Preparation of continuous phase and dispersion phase

The aqueous solution contains chemicals of N-Isopropyl acrylamide monomer (NIPAM, TCI, 98%, 11.32 wt%), N,N -methylenebisacrylamide (MBA) crosslinker (J&K Chemicals,98%, 0.77 wt%) , ammonium persulfate (APS, Sigma Aldrich, 98%, 0.6 wt%) , Sodium lauryl sulfonate surfactant (SDS, Sigma Aldrich, analytically pure, 0.1 wt%) and fluoro-surfactant (Shanghai jianbang industrial co. LTD, 0.1wt%). All the chemicals were dissolved by deionized water by mixing for half an hour. Monodispersed $\mathbf{Fe_3O_4}$ Magnetite Microspheres (Aladdin, 25 wt%) was added 0.04μl per 10ml aqueous solution. Thus the middle phase was obtained.

The oil phase contains soybean oil (58.8% v/v), n-tetradecane (Aladdin, 98%, 41.2% v/v) and benzoin dimethyl ether (BDK, Aladdin, 99%, 1 wt%). The outer phase needed to be mixed by a heating mixer, due to the high viscosity of 15.4 mPa·s and low solution rate of BDK. In order to obtain a suitable viscosity of the oil phase mixture, we measured the viscosity of soybean oil, n-tetradecane and five oil phase mixtures respectively. The viscosity data is shown in Table S1.

Table S1 The viscosity data of oil phase (T=24℃)

| Soybean oil （**ml**） | **n-tetradecane （ml）** | **viscosity (mPa·s)** |
|---|---|---|
| 8.0 | 0 | 44.3 |
| 6.5 | 1.5 | 28.0 |
| 5.5 | 2.5 | 19.4 |
| 5.0 | 3.5 | 15.4 |
| 5.0 | 4.0 | 14.4 |
| **4.5** | **4.0** | **13.2** |
| 0 | 8.0 | 2.5 |

During the experiment, because the glass capillary is hydrophilic, which might influence the Janus emulsions' monodispersity. We need to change the hydrophilism to hydrophobicity by covering the inwall of the capillary a layer of hydrophobic molecule. The modified solution is 95 wt% ethanol solution dissolving 0.5 wt% acetic acid and 2 wt% n-Dodecyltrimethoxysilane (J&K Chemicals,95%). The 1.5 mm-inner-diameter capillaries are dipped in the modified solution for more than 4 hours and stoved at 90℃. Then we get the hydrophobic capillaries.

### Analysis of the emulsions

The surface tension (gas-liquid) and the interfacial tension (water-oil) was measured by an interfacial tension meter (OCAH200, Data Physics Instruments GmbH, Germany) using the pendent drop technique. The forming process of G/W/O emulsions was recorded by a high-speed camera (Model 8964, DANTEC Dynamics, USA), which was connected to a microscope (Olympus ,Japan). The average droplet diameter and bubble size were determined by analyzing the recorded videos and the picture of emulsions or gels in culture dish captured by a high-speed camera.

**Janus structure prediction code**

```
clear;
clc;
format long
digits(8)
O=[4.961,29.844,25.308];%[Interfacial tension between: Tetradecane/Water, Air/Water, Air/Tetradecane ]
q=4;%[volume ratio of Water to Air]
a=acos(0.5*(O(1)^2+O(3)^2-O(2)^2)/(O(1)*O(3)));%Tetradecane/Water 与 Air/Tetradecane 夹角
b=acos(0.5*(O(1)^2+O(2)^2-O(3)^2)/(O(1)*O(2)));%Air/Water 与 Tetradecane/Water 夹角
c=acos(0.5*(O(2)^2+O(3)^2-O(1)^2)/(O(3)*O(2)));%Air/Tetradecane 与 Air/Water 夹角
J1=a*180/3.14;
J2=b*180/3.14;
J3=c*180/3.14;
x0=[2,1.5,pi/4,pi/4,pi/4];%Initial Value
opt=optimset('MaxFunEvals',10000,'MaxIter',10000);
[x1,fv1,ef1,out1]=fsolve(@Janus,x0,opt,q,b,c);
if ef1==1
%h=cos(x1(4))-x1(1)*cos(x1(3));
h=sin(x1(3)-x1(4))/sin(x1(3));
[X,Y,Z]=sphere(30);

X1=X;
Y1=Y;
Z1=Z;
h0=cos(x1(4));
surf(X1,Y1,Z1,'facealpha',1,'linestyle','-');
%alpha(0.3);
hold on
X2=x1(1)*X;
Y2=x1(1)*Y;
Z2=x1(1)*Z+h;
surf(X2,Y2,Z2,'facealpha',1,'linestyle','-');
%alpha(0.3);
hold off
h1=sin(x1(4)-x1(5))/sin(x1(5));
axis equal,axis off
view(20,10)
q
```

```
angle=[J1 J2 J3]
R=[x1(1) 1 x1(2)]
h=[h 0 -h1]
w=abs(cos(acos((1+(-h1)^2-(x1(2))^2)/(2*abs(-h1)))))
colormap([0 0.7 0;1 0 0])
caxis([w-5 w+5])
else
x0=[1,1.5,pi/4,pi/4,-pi/4];
[x2,fv2,ef2,out2]=fsolve(@Janus,x0,opt,q,b,c);
h=sin(x2(3)-x2(4))/sin(x2(3));
[X,Y,Z]=sphere(30);
X1=X;
Y1=Y;
Z1=Z;
h0=cos(x2(4));
surf(X1,Y1,Z1,'facealpha',1,'linestyle','-');
%alpha(0.3);
hold on
X2=x2(1)*X;
Y2=x2(1)*Y;
Z2=x2(1)*Z+h;
surf(X2,Y2,Z2,'facealpha',1,'linestyle','-')
%alpha(0.3);
h1=sin(x2(4)-x2(5))/sin(x2(5));
hold off
axis equal,axis off
view(20,10)
q
angle=[J1 J2 J3]
R=[x2(1) 1 x2(2)]
h=[h 0 -h1]
w=abs(cos(acos((1+(-h1)^2-(x2(2))^2)/(2*abs(-h1)))))
colormap([1 0 0;0 0.7 0])
caxis([w-5 w+5])
end
```