## **SUPPLEMENTARY INFORMATION**

# **Microencapsulation of Indocyanine Green for potential applications in image-guided drug delivery**

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### **ESI (S1)**

#### **The mathematical formulation for the scaling law in Equation (1)**

Consider a co-flowing tri-axial liquid jet surrounded by the atmosphere air at the orifice exit. The tri-axial jet consists of an innermost liquid cylinder, an annular outer liquid and an annular focusing liquid.



**ESI Figure 1** Sketch of the compound jet at the orifice exit.

In this model, the quantities mainly include: (a) liquid flow rate  $Q_i$ ,  $Q_o$ ,  $Q_f$ , (b) jet radius  $R_i$ ,  $R_o$ ,  $R_f$  (corresponding diameter  $D_i$ ,  $D_o$ ,  $D_{\text{orig}}$ ); (c) liquid density  $\rho_i$ ,  $\rho_o$ ,  $\rho_f$ , (d) jet averaged velocity  $u_i$ ,  $u_o$ , *u*<sub>*f*</sub>; (e) interfacial tension  $σ<sub>io</sub>$ ,  $σ<sub>of</sub>$ , and surface tension  $σ<sub>fa</sub>$ ; (f) liquid pressure  $P<sub>i</sub>$ ,  $P<sub>o</sub>$ ,  $P<sub>f</sub>$ ; for the inner, the outer and the focusing liquids, respectively. The atmosphere pressure can be denoted by *Pa*.

The liquid flow rate can be expressed as,

$$
Q_i = \pi R_i^2 u_i
$$
,  $Q_o = \pi (R_o^2 - R_i^2) u_o$ ,  $Q_f = \pi (R_f^2 - R_o^2) u_f$ .

The liquid pressure and the atmosphere pressure can be written as,

$$
P_a = P_f + \frac{\sigma_{fa}}{R_f}, \quad P_f = P_o + \frac{\sigma_{of}}{R_o}, \quad P_o = P_i + \frac{\sigma_{io}}{R_i},
$$

and

$$
P_f = \frac{1}{2} \rho_f u_f^2, \ \ P_o = \frac{1}{2} \rho_o u_o^2, \ \ P_i = \frac{1}{2} \rho_i u_i^2.
$$

In this problem, the magnitudes of quantities can be estimated to be in the range of,  $[\sigma] \approx 10^{-2} \sim 10^{-3}$  N/m,  $[R] \approx 10 \sim 100$   $\mu$ m= $10^{-5} \sim 10^{-4}$  m, and  $[\sigma/R] \approx 10 \sim 10^{3}$  Pa. Therefore,

.

$$
P_f \gg \frac{\sigma_{fa}}{R_f}, \ P_o \gg \frac{\sigma_{of}}{R_o}, \ P_i \gg \frac{\sigma_{io}}{R_i}
$$

Then one can obtain,

$$
\frac{1}{2}\rho_f u_f^2 \sim \frac{1}{2}\rho_o u_o^2 \sim \frac{1}{2}\rho_i u_i^2
$$

As a result, the diameter of the outer liquid jet can be expressed as,

$$
D_o \sim \left(\frac{\sqrt{\rho_i Q_i} + \sqrt{\rho_o Q_o}}{\sqrt{\rho_f Q_f} + \sqrt{\rho_i Q_i} + \sqrt{\rho_o Q_o}}\right)^{1/2} D_{\text{orig}}.
$$
\n
$$
\tag{*}
$$

For different liquids with similar densities in LDCFF process, one can assume  $\rho_i \sim \rho_o \sim \rho_f$ ,

 $Q_f \gg Q_i$ ,  $Q_f \gg Q_o$ . The diameter of the outer liquid jet can be simplified to be,

$$
D_o \sim (\frac{Q_i + Q_o}{Q_f})^{1/2} D_{orif}
$$

For an axisymmetric breakup of a liquid jet, the diameter of the resultant droplets *D* can be estimated by classical linear instability theory, which results in the form of

$$
D \sim \alpha \left(\frac{Q_i + Q_o}{Q_f}\right)^{1/2} D_{\text{orig}} , \qquad \qquad (*)
$$

where  $\alpha$  is a coefficient that is closely related to the process parameters such as the liquid properties and the liquid flow rates in LDCFF.

#### **ESI (S2)**

### **Discussion of the fitting curves predicted by the scaling law in comparison with experimental observations**

Generally, the droplet diameter *D* increases as the inner and outer liquid flow rates increase, while it decreases as the focusing liquid flow rate increases. In this work, the scaling law of the droplet diameter is compared with experimental observations. We use a fixed fitting coefficient  $\alpha$ =1.9 to fit all the experimental data so that all the data points could be weighed equally. The tendency of the variation curves predicted by the scaling law is in accordance with the experimental results, but a little difference still exists. The discrepancy can be mainly contributed to two factors. On one hand, there are some assumptions and simplifications in the theoretical model, as described in ESI (S1). The model can be modified by considering the effect of interfacial tensions and the difference between the liquid densities. On the other hand, the value of  $\alpha$  is dependent on the process parameters such as the liquid properties and the liquid flow rates, which determine the jet diameter, the wavelength of perturbations on the jet interface as well as the behavior of the jet breakup. The fitting coefficient for the scaling law should be slightly changed for different group of process parameters in experiments.