Electronic Supplementary Information (ESI)

Robust fabrication of ultra-soft tunable PDMS microcapsules as a biomimetic model for red blood cells

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S1 Structure design of the Teflon connectors

The Teflon connectors referred to in Fig. 2(a) of the main text are fabricated according to the design reported by Levenstein *et al.*¹, as shown in Fig. S1. A hole of diameter 1 mm was drilled through the centre of a Teflon cylinder to create an axial through-channel and enable the insertion of the injection/collection capillary. Another inlet was provided by drilling an access channel (diameter 1.5 mm) halfway along the length of the cylinder to supply the middle (outer) phase. The axial channel was also enlarged to a diameter of 1.5 mm in this region. A circular recess (diameter 2.0 mm) was also drilled at one end of the cylinder through the centre of its face to enable the insertion of the wider outer capillary.

S2 Normal and abnormal flow behaviours in the device

Video 1. Generation of double emulsions under normal conditions.

Video 2. Mixing of inner and outer phases due to the misalignment of injection and collection capillaries.

Video 3. Interface rupture due to the non-hydrophobic treatment of the injection capillary.

Video 4. Irregular interface duo to the non-hydrophilic treatment of the outer and collection capillaries.



Fig. S1 Structure of the Teflon connectors. All dimensions are given in mm.

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S3 Design of planar porous media

The planar porous media used for capsule filtering and as biomimetic models of porous tissues (e.g., placenta), respectively, were made of PDMS cured in a micromilled mould. The channels were designed by positioning cylindrical pillars (diameter $d_{\text{pillar}} = 715 \ \mu\text{m}$) in a 40 × 19.8 mm Hele-Shaw cell, in an hexagonal ordered array (Fig. S2 (a)) and a randomly disordered array (Fig. S2 (b)). Both channels have a depth of 250 μm .



Fig. S2 Top views of planar porous media as biomimetic analogues for placental micro-structures: (a) hexagonal ordered porous medium and (b) capsule suspension flow in a disordered porous medium (Haematocrit \approx 50%). Flow from left to right.

S4 Measurement of capsule size and membrane thick-

ness

The inner and outer diameters of the capsules were measured with close-up images (Fig. S3) by averaging at least 10 measurements of the diameter of enclosing circles (blue and red for bulk capsule and inner core, respectively).



Fig. S3 Close-up image of spherical capsules after fabrication. The blue and red circles are the contours of the bulk capsule and inner core, respectively. The scale bar is 100 $\,\mu m.$

S5 Measurement of Young's modulus

We employ a commonly-used compression method that has been described by Willshaw² to measure the Young's modulus of cured PDMS with the mixing ratio of PDMS base to the crosslinker ranging from 10:1 to 40:1. Liquid PDMS is moulded into cylindrical samples of 2.0 cm in diameter and 3.0 cm in height, followed by a uniaxial compression test with an Instron 5569 machine (Instron, High Wycombe, UK), where the load-displacement relationship is recorded, as shown in Fig. S4 (a). The measured load-displacement data is then converted into the nominal stress σ (Pa) and strain ε (mm/mm) according to

and

$$\sigma = \frac{L}{A_0} \tag{S1}$$

$$\varepsilon = \frac{\Delta h}{h_0},\tag{S2}$$

where L (N) is the force exerted on the top surface of the test cylinder, A_0 is the initial area of the top surface, Δh is the displacement of the top surface of the cylinder and h_0 is its initial height. We apply a thin lubricating layer of Vaseline to the top and bottom surfaces of the test cylinders to ensure they are deformed uniaxially. The Young's modulus is then obtained by fitting the experimental stress-strain data with the theoretical models of elastic materials. Here, we consider three common models: Hookean model^{2,4}, neo-Hookean model^{2,3} and the two-term Mooney–Rivlin model^{2,4}. By assuming the material to be isotropic, homogeneous and incompressible under uniaxial deformations, a relationship between the axial stress and onedimensional strain is derived from the basic equations. Then, the nominal stress-strain relationships for these three models are expressed as follows:

Hookean model:

$$\sigma = E\varepsilon, \qquad (S3)$$

neo-Hookean model:

$$\sigma = 2C_1 \left(1 - \varepsilon - \frac{1}{(1 - \varepsilon)^2} \right), \tag{S4}$$

and Mooney–Rivlin model:

$$\sigma = \left(2C_1 + 4C_2\left((1-\varepsilon)^2 + \frac{2}{1-\varepsilon} - 3\right)\right)\left(1-\varepsilon - \frac{1}{(1-\varepsilon)^2}\right),$$
(S5)

where *E* (Pa) is the Young's modulus, and *C*₁ (Pa) and *C*₂ (Pa) are the parameters obtained by fitting the experimental data with corresponding equations. For small deformations ($\varepsilon \ll 1$), the neo-Hookean eqn (S4) and Mooney–Rivlin eqn (S5) models reduce to

$$\sigma = 6C_1 \varepsilon. \tag{S6}$$

Then, the Young's modulus modulus is calculated as

$$E = 6C_1. \tag{S7}$$

Table S1 The Young's modulus E obtained by fitting Hookean, neo-Hookean and Mooney–Rivlin models to the experimental stress-strain data (unit: kPa). The error comes from the standard deviation of results performed at different compression rates (from 0.01 mm/s to 1.00 mm/s).

Mixing ratio	Hookean	Neo-Hookean	Mooney–Rivlin
10:1	$1633.8{\pm}32$	$1453.8{\pm}24$	$1405.8{\pm}20$
20:1	$534.1 {\pm} 3.5$	470.7±3.8	430.3±9.8
30:1	$148.1{\pm}1.2$	$133.9{\pm}1.3$	$124.5 {\pm} 2.7$
40:1	$50.3{\pm}0.4$	$45.5{\pm}0.3$	$41.6{\pm}0.7$

Fig. S4 (b) shows an example of the fittings of the equations eqn (S3) (red), eqn (S4) (blue) and eqn (S5) (green) to the experimental stress-strain data (black crosses). In this case, PDMS and its crosslinker are mixed in 40:1, the compression rate is 0.01 mm/s. The results show that Hookean model gives a poor fitting to the experimental data, which indicates that the generally accepted linear elasticity of PDMS under uniaxial compression is not suitable in our experiments. The two-term Mooney–Rivlin model eqn (S5) provides the best prediction of the elastic behaviour of PDMS across the whole range of strain considered in this experiment. Table S1 lists all the values of Young's modulus approximated by the three models, where the results predicted by Mooney–Rivlin model are considered for the capsule characterisation in this study.



Fig. S4 (a) Schematic illustration of the compression test on a cylindrical sample (see the text for more details). (b) Fitting of three elasticity models, Hookean (eqn (S3), red), neo-Hookean (eqn (S4), blue) and Mooney–Rivlin (eqn (S5), green) models, to the stress-strain data (crosses). In this case, PDMS and corsslinker is mixed in 40:1, and the compression is performed at a rate of 0.01 mm/s.

References

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