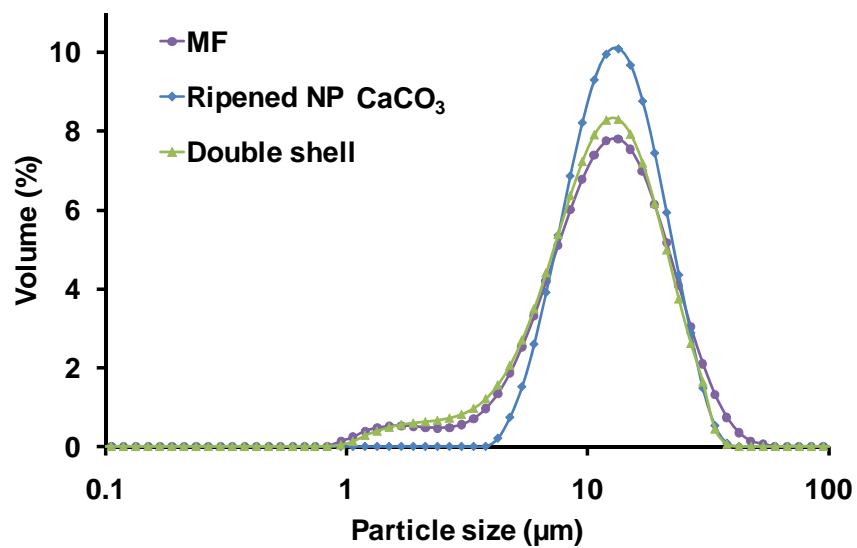


Supplementary Information:

AI. The core oil is a typical perfume blend of various components, which have a relatively low solubility in water and is used in consumer products. Calcium carbonate nanoparticles (average diameter 80 nm, Omya, UK), CaCl_2 and Na_2CO_3 (sigma-Aldrich, UK) were used for the formation of the inorganic shell material. MF precondensate (70 % wt (aq), formaldehyde to melamine molar ratio 0.2, British Industril Plastics Ltd, Birmingham, UK), and poly (acryl amide-acrylic acid, sodium salt) (Polyersciences, Inc., US) were used as the organic shell material. The MF, CaCO_3 and double shell microcapsules were characterized by ESEM (JEOL, 1200EX, Jeol Ltd, UK), TEM (FEI/Philips XL30 ESEM-FEG, Philips, UK), gas chromatography (GC, trace 2000 series) and the micromanipulation technique.¹⁰

A2. Size distribution curves (measured by laser diffraction technique) of MF, Ripened NP CaCO_3 , and double shell composite microcapsules.



A3. Leakage Measurement

Microcapsules (MF, Ripened NP CaCO_3 , and double shell composite) were filtered from the original aqueous dispersion, and re-dispersed in water (50 ml). To the resulting aqueous dispersion, hexane was added (30 ml), which was stirred for 10 min. A hexane aliquot (1 μl) was removed and analysed by GC to determine the amount of oil. Further aliquots (1 μl) were removed at various time intervals between 1 to 24 hours prior to which the dispersion was stirred for 10 minutes.

A4. Transmission Electron Microscopy (TEM)

Transmission Electron Microscopy (JEOL 1200EX, Jeol Ltd, UK) was used to examine the structure and thickness of the microcapsule wall. Microcapsules were embedded in LR white hard grade acrylic resin, and the ultra thin sections were obtained by using an ultracut microtome apparatus (Reichert-Jung), see below. Operating voltage was 80 eV.⁹

Sampling by Ultra-microtome

The microcapsule dispersions (2 ml) were centrifuged at a speed of 500 rpm for 3 min, and the supernatant liquid was separated. Gluteraldehyde (2 ml) was added and the microcapsules were re-dispersed by gently shaking the vial manually. The resulting microcapsule dispersions were stored for 1 hour to harden the outer shell of the microcapsules, and centrifuged for 3 min at 500 rpm. The supernatant liquid (excess gluteraldehyde) was separated, and a solution of ethanol and water was added (1:1, 2 ml), and shaken by hand. The resulting mixture was centrifuged again at 500 rpm for 15 min, and the supernatant ethanol/water mixture was separated. The ethanol/water process was repeated four times, but with ethanol/water 70 %, 90 %, 96 % (v/v), and finally the microcapsules were suspended in absolute ethanol. The resulting microcapsules dispersion was centrifuged again at 500 rpm for 15 min, and the supernatant liquid (excess absolute ethanol) was decanted. To the microcapsule slurry was added absolute ethanol and LR white resin (1:1, 2 ml), and the microcapsules were re-dispersed by gently shaking the vial manually. The resulting mixture was put on a rotator at a speed of 4 rpm. After ~3 min the microcapsules were displaced to the bottom of the vial, and rotating continued for a total of 3 hours. The supernatant ethanol/resin solution was separated. 100 % of LR white resin (2 ml) was added to the resulting microcapsule slurry, before the

mixture was rotated again at 4 rpm for 12 hours, and the microcapsules were separated from the supernatant. Two beam capsules (15×5 mm) were filled to the top with the air free LR white resin (2 ml). The separated microcapsules were added to each resin filled beam capsule. The microcapsules settled on the bottom of the beam capsules. The mixture was heated ($60\text{ }^{\circ}\text{C}$) under vacuum for 30 min, then left to cure for 48 hours at $60\text{ }^{\circ}\text{C}$, affording a polymerized resin-microcapsule block. The resulting block was secured into a REICHERT-JUNG ultramicrotome apparatus and the microcapsule end was trimmed to sections of thickness 90-150 nm. The gold coloured sections were placed on carbon coated grids (G2500C, $2\text{ mm} \times 1\text{ mm}$ slot, copper, 3.05 mm) ready to be examined by TEM. The wall thicknesses of microcapsules were measured from the analysis of their TEM images of the ultra microtome sections.⁹