Electronic Supporting Information (ESI) accompanying the paper:

"Independent responsive behaviour and communication in hydrogel objects"

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Experimental methods

Materials

Alginic acid sodium salt (referred to as sodium alginate in this paper), calcium chloride hexahydrate (98%), urease from Canavalia ensiformis (Jack bean) type III (100K units), urea (99.0 - 100.5%), oil blue N (96%) and oil red O were purchased from Sigma Aldrich. Ethylenediaminetetraacetic acid (99%) was purchased from Avocado Research Chemicals Ltd. and hydrochloric acid (37%) was purchased from VWR international. Vegetable oil was purchased from Costcutters Ltd. Bromothymol blue, ACS reagent, was purchased from Fisher Scientific. Oleoresin paprika oil 80,000 NS was purchased from Kalsec.

Standard wall borosilicate glass capillaries (GC100-10, OD 1.0 mm, ID 0.58 mm and GC200-7.5, OD 2.0 mm, ID 1.16 mm) were purchased from Harvard Apparatus. Evo-Stik Two Part epoxy resin was applied to seal the capillaries where necessary. Solutions were introduced to the microfluidic device through the Clear Tygon® tubing (0.8 mm ID, ColePalmer) attached to syringes driven by positive displacement syringe pumps (Harvard Apparatus, PHD 2000 series). Weller Dispensing Needles KDS3012P (GA 30, ID 0.15 mm) were used to connect the tubing to the microfluidic device.

Composite gel synthesis

A glass capillary with an inner and outer diameter of 1.16 mm and 2.00 mm, respectively, forms the outer casing of the device and contains the flow of the aqueous alginate phase. Within this, two capillaries with inner and outer diameters of 0.58 mm and 1.00 mm, respectively, were tapered to 110 μm with a laser puller to allow flow focusing of the inner oil phase. These tapered capillaries were aligned to create a flow-focused junction where droplets of oil are formed. The output capillary of the device is angled into a bath of 0.1 mol dm^{-3} aqueous calcium chloride solution.

A vegetable oil phase containing 2 mg mL^{-1} of either oil red O, oil blue N or paprika oil was pumped through a 0.1 g L^{-1} solution of sodium alginate of the desired urease concentration to form droplets of low size dispersity. This oil droplet containing flow of alginate solution was collected in a continuous phase of 0.1 mol dm^{-3} calcium chloride hexahydrate, where upon an alginate-droplet cable was formed. Typical flow rates were 0.005 mL min^{-1} for the oil phase and 0.15 mL min^{-1} for the aqueous alginate phase. This procedure generates calcium cross-linked sodium alginate hydrogel fibres in which the enzyme urease is entrapped, where it is locally confined but its activity is retained. 300 μm diameter oil droplets are embedded along the length of the fibre, their positions fixed until cross-linking in the gel matrix is disrupted. Fibres of 41 droplets in length are used for the fibre studies in all cases. These are cut from a freshly synthesised length of fibre, of which the enzyme-sodium alginate solution was prepared that day.

The cable was washed in distilled water and deposited into 20 mL of 0.1 mol dm^{-3} ethylenediaminetetraacetic acid (EDTA) containing 2 mL of 1 mol dm^{-3} urea, corrected to a pH of 3.5 using 1 mol dm^{-3} hydrochloric acid solution. Fibres are stable in this solution over a matter of days, eventually disintegrating after this time (a considerably longer timescale than studied here).

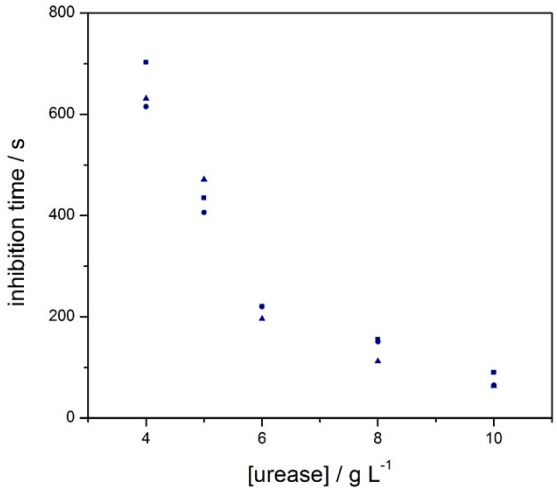
Responsive behaviour was filmed on a Nikon D5100 camera with AF-S DX Micro NIKKOR 40mm f/2.8G Lens immediately following the addition of the fibre to the urea/EDTA solution.

Local pH changes were observed by placing a fibre embedded with 10 g L^{-1} of urease into 20 mL of saturated bromothymol blue solution containing 2 mL of 1 mol dm^{-3} urea.

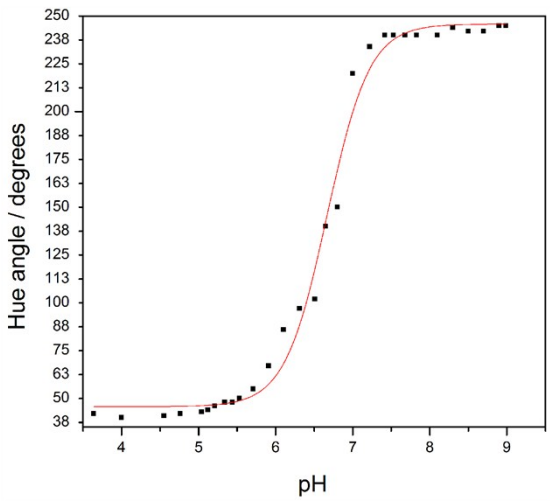
Gel beads were formed by dropping 0.5 g L^{-1} solutions of sodium alginate containing 2 mg mL^{-1} bromothymol blue and either 0.008 g L^{-1} or 10 g L^{-1} of urease into a 0.1 mol dm^{-3} calcium chloride hexahydrate solution. The 0.008 g L^{-1} urease beads were washed and placed into 20 mL of distilled water containing 2 mL of 1 mol dm^{-3} urea. The 10 g L^{-1} urease beads were washed then placed into a 1 mol dm^{-3} urea solution. They were introduced to the 0.008 g L^{-1} urease beads when required.

The gel beads were analyzed using ImageJ to collect an RGB histogram of their total area (excluding white reflection spots), from which the HSV value and its constituent hue angle is calculated.[1]

Extended Figures



Extended Figure 1 | Inhibition time at varying concentrations of urease. As the concentration of urease increases, the inhibition time prior to material disassembly (defined here as the time taken for the first oil droplet to be released) is reduced. This results in a shorter dormancy period.



Extended Figure 2 | pH calibration of bromothymol blue indicator. The pH of a bromothymol blue (BMB) solution containing 4 g L^{-1} of urease and 0.09 mol dm^{-3} of urea is measured. As the pH increases, a change from yellow to blue is observed. By taking an RGB histogram and converting to HSV, the change in hue angle, and by extension, colour, of the solution is tracked over the course of the experiment. A Boltzmann curve is fitted, where y = 245.84826 + (45.71295-245.84826)/(1 + exp((x-6.66741)/0.27339)).

1. Hanbury, A. *Constructing cylindrical coordinate colour spaces*. *Pattern Recognition Letters* **29,** (2008).