Supporting Information

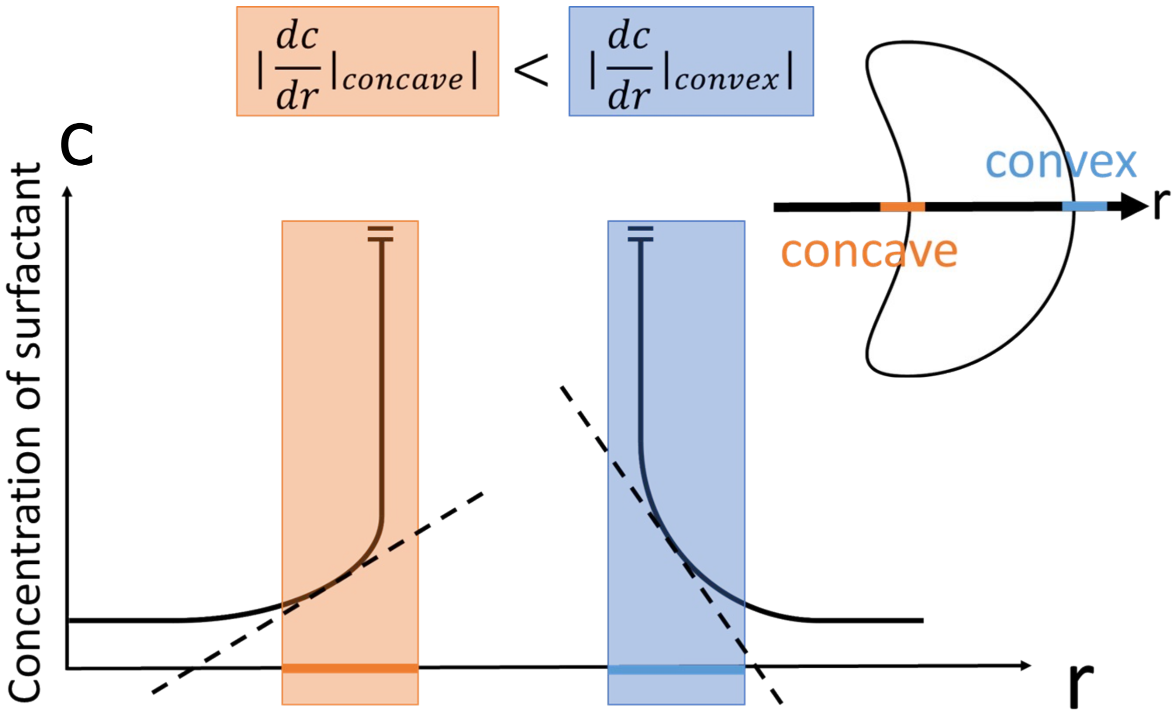

Fig. S1 Schematic representation of the concentration of oleic acid (surfactant) in a boomerang shape. By observing the concentration of oleic acid in r direction, the concentration value is different between the convex region and the concave region as shown in the graph, due to the different curvatures of the droplet surface. The absolute value of the concentration gradients is higher at the convex region than that at the concave region in the boomerang shape. This difference in the concentration leads to the difference in water-oil interface tension.

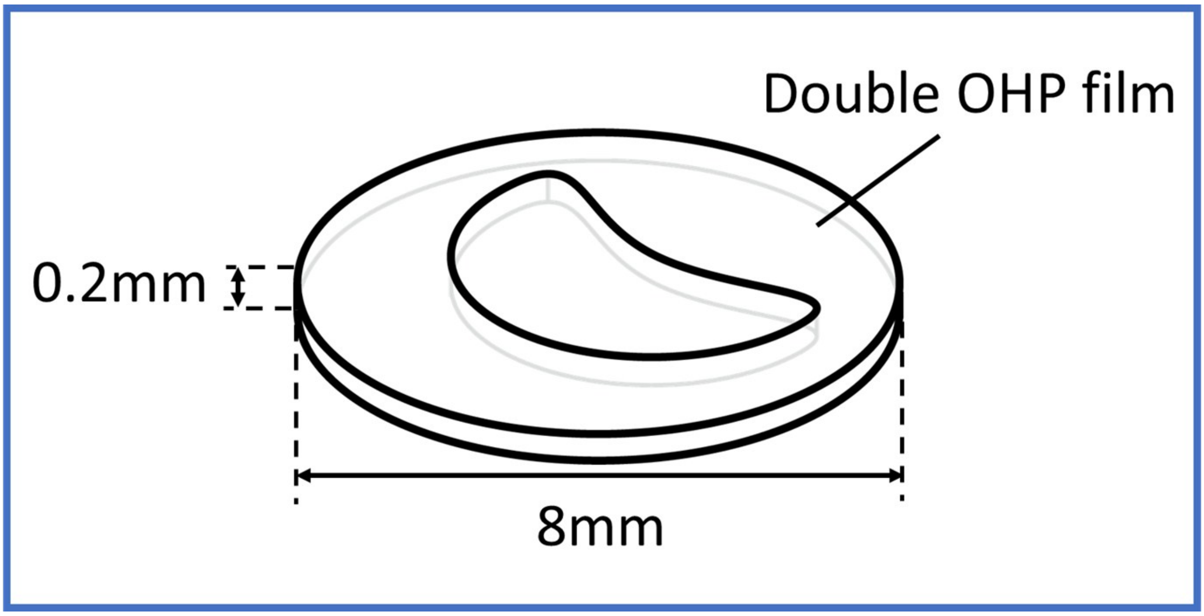

Fig. S2 Schematic diagram of the exoskeleton.

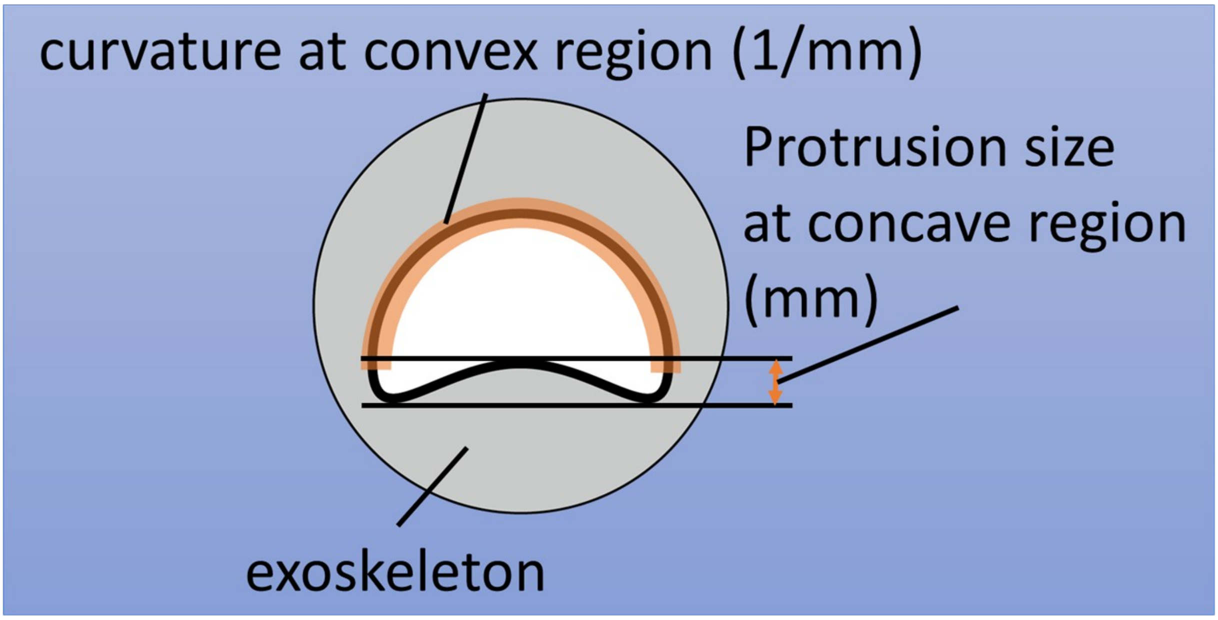

Fig. S3 Curvature and protrusion size parameters at the convex and concave regions of the exoskeleton.

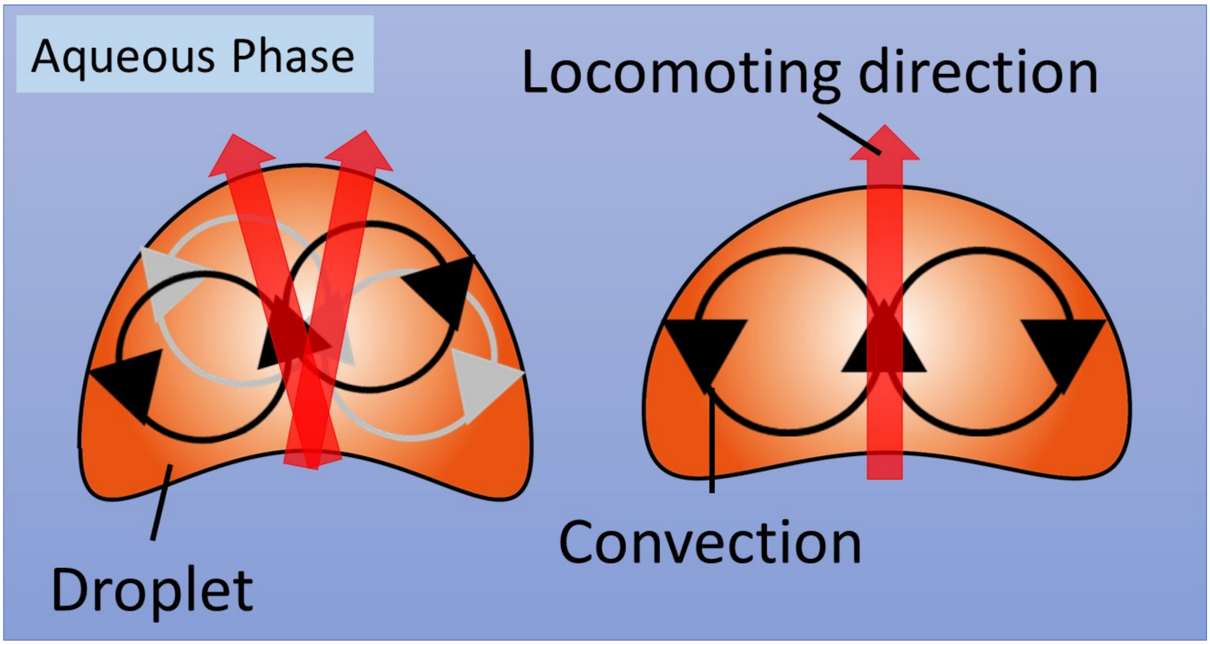

Fig. S4 Differences in locomotion style for exoskeletons IV and X. A self-propelled oil droplet in exoskeleton IV (left) is more spherical than the droplet in exoskeleton X (right). Spherical self-propelled oil droplets tend to locomote randomly. Furthermore, the resistance received from the aqueous phase is constant from all directions. Therefore, exoskeleton IV has greater stability with respect to velocity change, while exoskeleton X has greater stability in traveling direction.

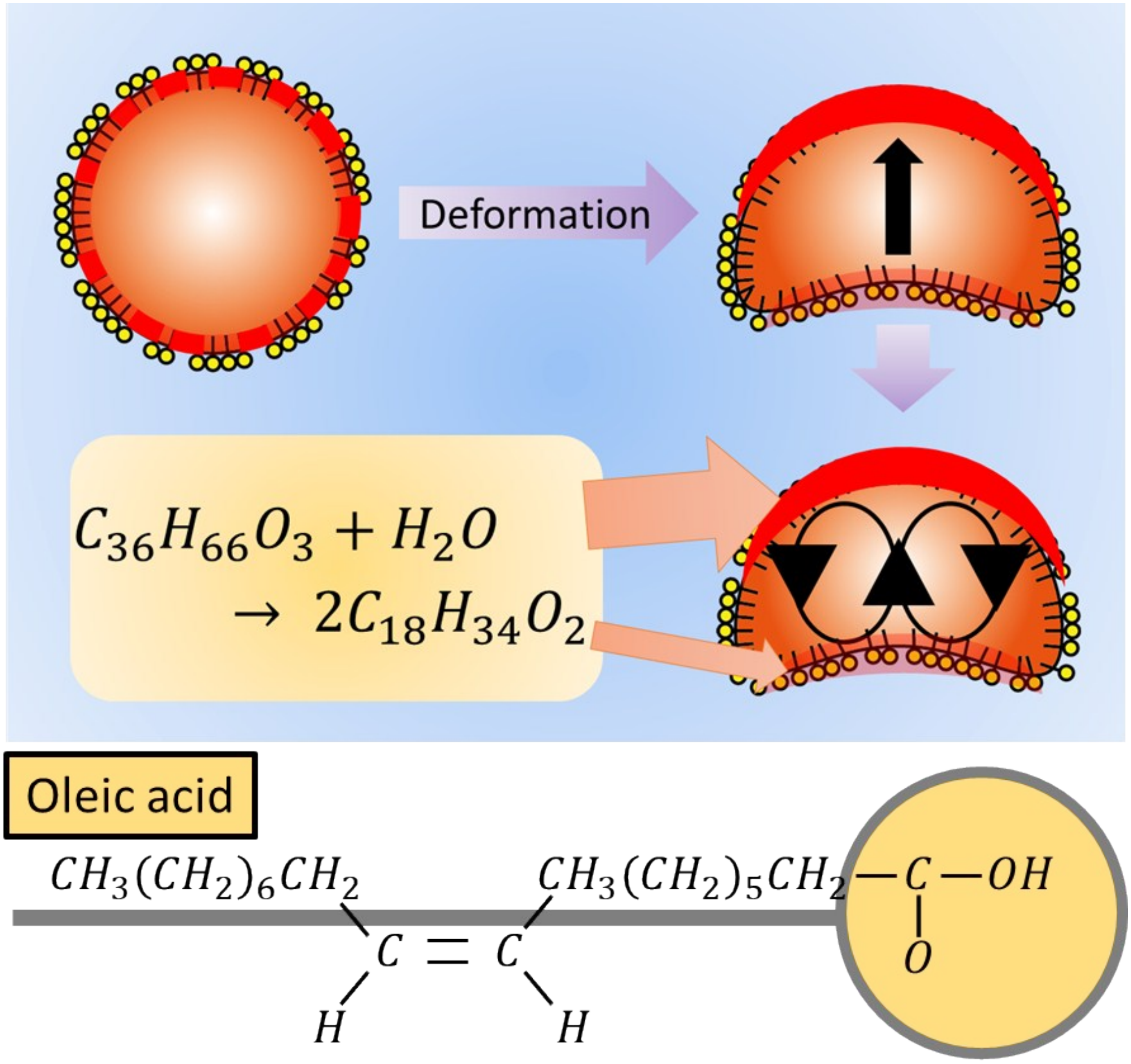

Fig. S5 Driving mechanism of self-propelled oil droplets using chemical reaction. (Upper left) The surfactant (oleic acid) is randomly adsorbed around the oil droplet. Variation occurs in the surfactant concentration at the oil droplet interface. Consequently, variations in the surface tension result from variations in the concentration, which leads to Marangoni instability. (Upper right) The oil droplet transforms into the boomerang shape under Marangoni instability.[41] The surface tension of the convex region is greater than that of the concave region in the boomerang-shaped oil droplet, and this difference changes inside of the oil droplet from a concave shape to a convex shape. (Lower right) Convection occurs inside the oil droplet as a result of Marangoni convection. Convection moves the surfactant anhydride (oleic anhydride) to the convex regions of the boomerang shape. The surfactant is generated more efficiently in the convex region than in the concave regions of the oil droplet. Therefore, the concentration gradient at the oil droplet interface is maintained, and the oil droplet continues to be self-propelled.[17] The bottom figure represents the chemical formula of oleic acid. Oleic acid is a surfactant, which has a hydrophilic group and a hydrophobic group.

Volume [µL]	1	2	3	4	5	6	7	8	9	10
Split [times]	0	0	5	4	5	5	6	6	7	All (24)
Try 1 Velocity [mm/s]	4.44	3.46	Split	Split	Split	2.37	2.37	Split	Split	-
Try 2 Velocity [mm/s]	4.25	3.41	Split	3.38	Split	2.38	Split	Split	Split	-
Try 3 Velocity [mm/s]	3.91	3.31	3.27	Split	Split	Split	Split	2.09	Split	-
Try 4 Velocity [mm/s]	3.72	3.41	Split	2.18	Split	Split	Split	Split	Split	-
Try 5 Velocity [mm/s]	3.68	3.36	3.58	Split	2.31	Split	Split	Split	Split	-
Try 6 Velocity [mm/s]	2.97	3.45	Split	Split	2.61	Split	Split	Split	Split	-
Try 7 Velocity [mm/s]	3.38	3.49	Split	3.62	Split	Split	Split	Split	Split	-
Average Velocity [mm/s]	3.76	3.41	3.42	3.06	2.46	2.37	2.37	2.09	-	-
SD [mm/s]	0.464	0.057	0.156	0.630	0.151	0.006	0	0	-	-

Table S1 Velocity data for oil droplets with different volumes (1–10 µL). We repeated the experiment 7 times for droplets with volumes of 1 to 9 µL and 24 times for the 10 µL oil droplets. The number of splits and average velocity were analyzed. SD is the standard deviation of the average velocity in 7 experiments.

Exoskeleton	I	II	III	IV	V	VI	VII	VIII	IX	X
Curvature [/mm]	0.0250	0.306	0.0360	0.0417	0.0472	0.0306	0.0306	0.0306	0.0306	0.0306
Protrusion size [mm]	0.628	0.628	0.628	0.628	0.628	1.050	0.791	0.628	0.465	0.302
Area [mm^2]	37.7	37.7	37.7	37.7	37.7	37.7	37.7	37.7	37.7	37.7

Table S2 Details of 10 types of exoskeletons. Exoskeletons I–V have different curvatures in convex region of the boomerang shape. Exoskeletons VI–X have protrusions in concave region of the boomerang shape. All exoskeletons have the same area (37.7 mm^2) with a fixed radius of 8 mm and thickness of 0.2 mm.