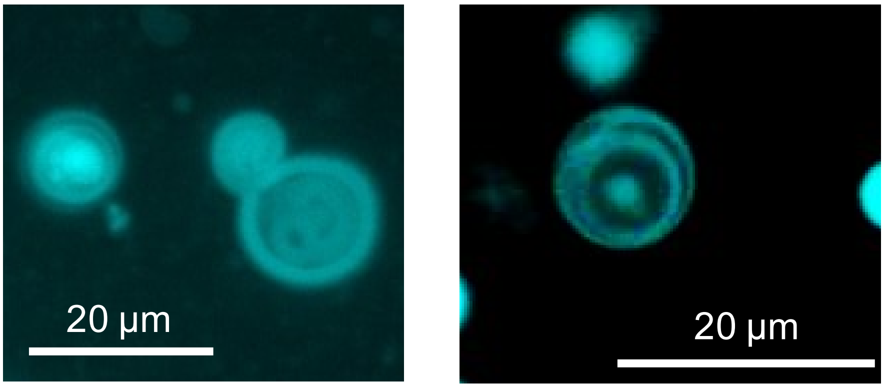
Fig. S1



(a) DDAB and Ca^{2+} (no oleate) (b) DDAB and oleate (no Ca^{2+})

Fig. S2

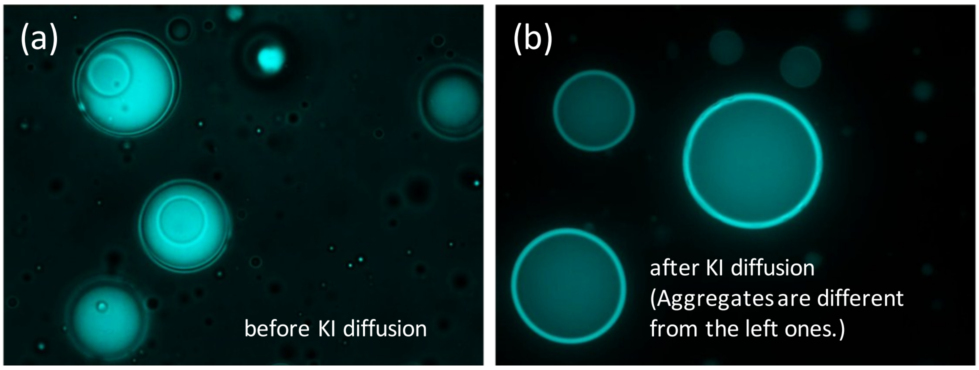


Table Dynamic behavior of the aggregates. Each chemical concentration was double the standard concentration in the Experimental section, except for (a) the amount of added NaOH and (b) the Ca^{2+} concentration. (c) The type of cation was varied.

(a)

pH	7.5	7.6	7.7	7.8	7.9	8	8.1	8.2	8.3	8.4
translation and coalescence	0	0	0	0	0	0	4	2	3	2
only coalescence	0	0	0	0	2	0	0	0	0	0
no motion	4	4	4	4	2	4	0	2	1	2

(b)

[Ca^{2+}]/mM	0	10	20	40	60	80
translation and coalescence	1	2	4	3	2	0
only coalescence	5	2	0	0	2	2
no motion	2	0	0	1	0	2

(c)

cation type	**Ca**	**Mg**	**Sr**	**Co**
translation and coalescence	4	3	0	1
only coalescence	0	0	0	2
no motion	0	1	4	1

Mechanism of sustainable motion

Here, we assume that the inner structure of VA is composed of numerous dehydrated vesicles. VA absorbs the surrounding smaller vesicles and discharges the inner vesicles via the reaction with KI. These discharged vesicles form a thin film that is spread over the glass surface. The driving force of translational motion is generated by the transformation from the inner vesicles into the film. For this discharge, VA decreases its size. However, the size is recovered by the absorption of the surrounding fresh smaller vesicles. The chemotactic nature along the concentration gradient makes the motion sustainable.

In the present experiment, both the VA and film were adsorbed on the glass surface. DDAB is a cationic surfactant. Therefore, the adsorption of DDA^+ on the glass surface is favoured at $pH \approx 8$. In a previous study, however, DDAB vesicles without oleate did not adsorb as readily on the glass surface. This is probably because DDAB vesicles without oleate are almost spherical. The adsorption of the vesicle on glass surface must accompany deformation from the sphere. The energy cost for this deformation probably prevents the adsorption. In contrast, the membrane containing oleate is much more flexible. This flexibility enables adsorption with large deformation.

Figure S3 presents a postulated mechanism for VA motion. VA that contains numerous dehydrated vesicles is adsorbed on the glass surface (Fig.S3a). When an aqueous solution with KI is injected and reaches a VA, the aqueous solution surrounding the VA is replaced gradually with the KI solution: DDAB and oleate molecules around the VA are diluted by the injected solution. The osmotic pressure of the surrounding solution may be reduced, and water permeates the dehydrated vesicles. This inflates the dehydrated vesicles located at the periphery (Fig.S3b), which is suggested from Figs.6 and 9: Numerous swollen vesicles can be seen, particularly along the periphery of VA. These swollen vesicles form a film by the formation of DDAI, which is the reaction product of DDA^+ and I^-. For this film formation, the water contained in the swollen vesicles is drained (Fig.S3c). This process is quite fast, as shown in Fig.6. The draining process may then generate the driving force for VA motion by momentum conservation, and the VA may absorb the simpler VAs (Fig.S3d). As a result, the VA leaves the film behind it. The film formation is transient, and the most stable form of the reaction product is small aggregates. Therefore, the film is transformed into numerous small aggregates, as shown in Fig.8 (30 s). The VA decreases its size due to the discharge of collapsed vesicles, but the size is recovered by the absorption of fresh vesicles.

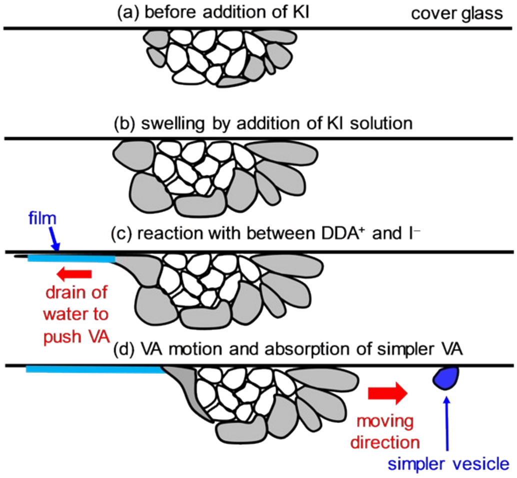


Figure S3 Illustration of the mechanism of the crawl motion of VA. (a, b) The dehydrated vesicles of the grey colored inflated by water permeation. (c) Some of them are transformed into a film. The drain of the inner water pushes the VA. (d) The moving VA may absorb smaller vesicles along the way.