Review of: "Hierarchical stabilization of emulsions with multi-scale interconnected droplets and ultra-low nanoparticle loadings"

Weichun Pan

Potential competing interests: The author(s) declared that no potential competing interests exist.

The manuscript of “Hierarchical stabilization of emulsions with multiscale interconnected droplets and ultra-low nanoparticle loadings” submitted by Cao et al. is of interest. Two-step strategy was adopted to prepare the multiscale interconnected droplets with an attractive feature of ultra-low nanoparticle loadings. But the main concern of my point of view is that the whole process is lack of control. Authors can’t control the structure, morphology, and size of the target objective. This is the main weak point of this manuscript. The second comment is the mechanism to stabilize large emulsion droplet. There is possible another scenario. A polyvalent cation affinity to PIONP is able to bridge two droplets, a similar one in Tofu production. So the key is the polyvalent cation. Indeed Tofu has the similar structure pattern as the one in this manuscript. Therefore, without this clarification, the novelty of this manuscript is not good enough for publication.

There are several serious questions which prevent this manuscript publication.

- Why do authors not input the line number in the manuscript? It is very unusually and hard to review and comment their work.
- There are many mistakes either typos or concept mistakse. For instance, where is the extended data figure 2; Pg26, “*Three were three different emulsion droplets of submicron droplets*”, *Three* should be *There*. All data in tables have not the standard deviation. And data in some figures, such as extended figure 14b and 15b, have not error bars. All these are not acceptable for scientific papers.

Minor problems.

- In abstract (Pg1), “creation of rigid interface and closed-cell structure with small total area”. I can’t understand the concept of “small total area”. How small is small?
- Eq. (1) (Pg 2) has several very strong assumptions, such as the contact angle of particles residing in oil-water interface is 90^{o}, and the arrangement of particles is a hexagonal close-packed on drop surface. Therefore, any deviation from the prediction of this equation is not surprise.
- Pg 3, “*In the second level, these submicron droplets in combination with polyvalent ions (e.g., Ca^{2+}, Co^{2+}, Ni^{2+}, and Fe^{3+}) spontaneously form macroemulsion small droplets, ca. 1.3 μm in diameter*” is contradict to Figure 1.
- Pg 3, “*The hierarchy greatly increase the surface-to-volume ratio*”. This statement strictly speaking is wrong. When the reference is large droplets, it is right. But if it shifts to small ones, it is not true. Since

there are multi-scale structures, this statement is no sense.

- Pg 3, “*Neither the large droplets nor the small droplets in macroemulsion require any coverage by individual nanoparticles to remain stable*.” This statement is right?
- Pg 4 and Extended Data Fig. 1, in order to state Fe_3O_4 crystalline, authors may provide more information, for instance, the Fourier transform of TEM image.
- Pg 4, “*these PIONPs can pin the oil–water interface after binding calcium ions through carboxylate group of surface polymers*”. This statement is different from the schematic picture of Fig. 1.
- Pg 5, “*These observations provide consistent evidence that the energy barrier creating small droplets is quite low*”. There are two questions. 1) How do authors build the relationship between these observations with the energy barrier to form small droplets? 2) Why is this barrier low? Less than RT?