Lab on a Chip

COMMENT

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# Reply to the 'Comment on "Robust scalable high throughput production of monodisperse drops"' by M. Nakajima, *Lab Chip*, 2017, 17, DOI: 10.1039/C7LC00181A

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This reply to the comment by Nakajima on our article that appeared in *Lab on a Chip* (E. Amstad, M. Chemama, M. Eggersdorfer, L. R. Arriaga, M. Brenner and D. A. Weitz, *Lab Chip*, 2016, **16**, 4163–4172) highlights the differences between the microchannel step emulsification devices developed by the Nakajima group and the millipede device reported by us in *Lab on a Chip*.

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The comment by M. Nakajima[1] on our article "Robust scalable high throughput production of monodisperse drops"[2] correctly points out that the first parallelized step emulsification device was developed in the Nakajima group in 1997 and that this very valuable contribution resulted in more than 100 follow-up papers.[3] Given the large number of papers, both by the Nakajima group and by others, we could not possibly cite all papers. Even though we did not cite this very first paper we did correctly reference the excellent work by the Nakajima group. Moreover, while we did not reference Nakajima explicitly, we did not reference any author explicitly by name. This seems the correct thing to do given the excellent contributions of so many authors. The step emulsification devices presented by Nakajima and all other groups are excellent examples of a method to parallelize drop formation with microfluidic devices, as we fully referenced in the paper. However, the drop break-up mechanism obtained in these devices is distinctly different than that in the millipede device introduced in our paper. Indeed, a main point of our paper is to introduce this new instability which leads to the drop break-up mechanism and which provides the very robust operation of the millipede device.

This difference in drop break-up mechanism is most clearly highlighted by the completely different scaling of the drop size with nozzle height, $h$: the diameter of drops formed by the millipede devices scales with $h$.[2] By contrast, the diameter of drops formed in the microfluidic step emulsification devices of the Nakajima group scales with $h^{\frac{1}{3}}$.[4] This difference is direct evidence of the new instability, which leads to drop formation in the millipede device and is why the device is so different than previously reported step emulsification devices.

The millipede nozzle geometry does have some resemblance with that of the microchannel step emulsification devices developed by the Nakajima group; however, there are distinct and very important differences. In the millipede device, the nozzles have a triangular opening that extends all the way to the step. By contrast, the nozzles of the microchannel step emulsification devices have a triangular opening followed by a rectangular extension up to the step. This extension, called a terrace by Nakajima, makes a profound difference in the behavior of the drop formation, changing the confinement of the inner fluid as the drop is formed: the inner fluid is primarily confined in the direction of the nozzle height. By contrast, in the millipede device, the inner phase is confined in the direction of the height of the nozzle and in its plane. This additional in-plane confinement of the inner phase in the millipede nozzles significantly changes the shape of the fluid tongue: it attains a semi-spherical shape during initial stages of drop formation and its radius continuously grows as the fluid flows towards the step.[2] By contrast, tongues formed in the microchannel step emulsification devices are nearly spherical and their shape remains almost constant when they flow towards the step.[5] This difference in the tongue shape has essential consequences for the mechanism of drop formation. For step emulsification devices with terraces, the hydrodynamic instability is a dynamic instability that forms in the flowing fluid. By contrast, for the millipede device, the instability that leads to drop formation is a quasi-static Rayleigh–Plateau instability that

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depends explicitly on the geometry of the device. This instability can only form if quasi-static conditions are established during early stages of drop formation. This is the case if the volume of the nozzle exceeds that of the forming drop such that the fluid flow into the nozzle is decoupled from the fluid flow into the drop. By contrast, the volumes of the terraces in Nakajima microchannel step emulsification devices are typically smaller than those of the forming drops such that no quasi-static conditions can be established. Instead, drop formation results from a the more typical dynamic Rayleigh–Plateau instability.[5] As was clearly pointed out in our paper, it is this new drop formation mechanism due to the quasi-static Rayleigh–Plateau instability, which is dependent solely on the device geometry, that provides the very robust behavior of the millipede device and makes large-scale parallelization so easily attained.

The coefficient of variation of drops produced with microchannel step emulsification devices with optimized terrace shapes, originally reported by Nakajima, can be narrow, as shown in Table 1 of our paper. Indeed, the CVs obtained with these devices are significantly narrower than those typically achieved with step emulsification devices with straight channels.[6] We included the excellent paper of Vladisavljevic *et al.*, because of the very high throughputs achieved even though it did not directly report values for the CV.[7] Instead, we estimated the CV from the bar graph shown in the paper. The new information provided in the comment by Nakajima indicates that the CV of the drops is lower than can be estimated from the bar graph. However, this low CV is only obtained if the flow rates in the channels are very low, resulting in a flow rate per nozzle that is much lower than those achieved by the millipede device having the same low CV. By contrast, if the Nakajima device is operated at similar flow rates per nozzle as the millipede device, the CV becomes much larger, as shown in Fig. 5 of their paper.[7]

The maximum throughput of the millipede device decreases with increasing viscosity, as reported in Fig. 5a in our paper. A very similar trend is observed for the mirochannel step emulsification devices.[8] This trend limits the amount of drops made of viscous fluids that can be produced per unit area and time for both devices.

## References

1 M. Nakajima, *Lab Chip*, 2017, 17, DOI: 10.1039/C7LC00181A.
2 E. Amstad, M. Chemama, M. Eggersdorfer, L. R. Arriaga, M. Brenner and D. A. Weitz, *Lab Chip*, 2016, **16**, 4163–4172.
3 T. Kawakatsu, Y. Kikuchi and M. Nakajima, *J. Am. Oil Chem. Soc.*, 1997, **74**, 317–321.
4 S. Sugiura, M. Nakajima and M. Seki, *Langmuir*, 2002, **18**, 3854–3859.
5 S. Sugiura, M. Nakajima and M. Seki, *Langmuir*, 2002, **18**, 5708–5712.
6 N. Mittal, C. Cohen, J. Bibette and N. Bremond, *Phys. Fluids*, 2014, **26**, 082109.
7 G. T. Vladisavljevic, I. Kobayashi and M. Nakajima, *Microfluid. Nanofluid.*, 2011, **10**, 1199–1209.
8 I. Kobayashi, S. Mukataka and M. Nakajima, *Langmuir*, 2005, **21**, 5722–5730.