Supplementary Information to

Dynamic behavior of floating magnetic liquid marbles under steady and pulse-width-modulated magnetic fields

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2D magnetic LMMS manipulation

To further extend the LMMS manipulation, the 2D position controlling of the floating LMMS under DC and PWM magnetic fields is presented in this section. The motion of the LMMS follows a linear set of equations, Equations 1-3 expressed below:

$$F_{net} = ma = F_m - F_f \tag{1}$$

$$F_f = 6\beta\pi r\mu v_x \tag{2}$$

$$F_m = \frac{\chi V}{\mu_0}(B.\nabla)\mathrm{B} \tag{3}$$

As a result, the superposition principle applies to this motion. Hence the motion in 2D coordinates could be defined as the summation of motion in two perpendicular directions. To support this argument, we further designed a set of experiments to observe if the LMMS follows a pre-defined 2D path line by adjusting the magnetic coil currents of three magnetic coils arranged in a triangle configuration. The 2D manipulation of the LMMS under the DC magnetic field is shown in Fig. S1(a). In this figure, green and red circles correspond to the magnetic coils when the current is turned on and off, respectively. This figure shows the LMMS could travel in L-shaped or diagonal paths by switching the magnetic coil current in the magnetic coils at different times. This magnetic actuation is similar to magnetic microcoil configuration, as observed in the research of Lehmann et. al. [1]. The advantage of using large magnetic coils with DC current instead of an array of micro coils is the ability to increase the distance between magnetic coils; therefore, the LMMS could be transferred with fewer magnetic coils and a simpler setup. The effect of the PWM magnetic field on the 2D manipulation is presented in Fig. S1(b). As shown, the PWM magnetic field proposes more

controllability on the LMMS 2D motion, offering manipulation with small step lengths by tuning the PWM frequency and duty cycle. The ability to stop the LMMS between the magnetic coils is achieved by the PWM magnetic field, which has not been feasible under the DC magnetic field. In this way, the number of magnetic actuators (in this case, large magnetic coils) could also decrease significantly.

a)

b)

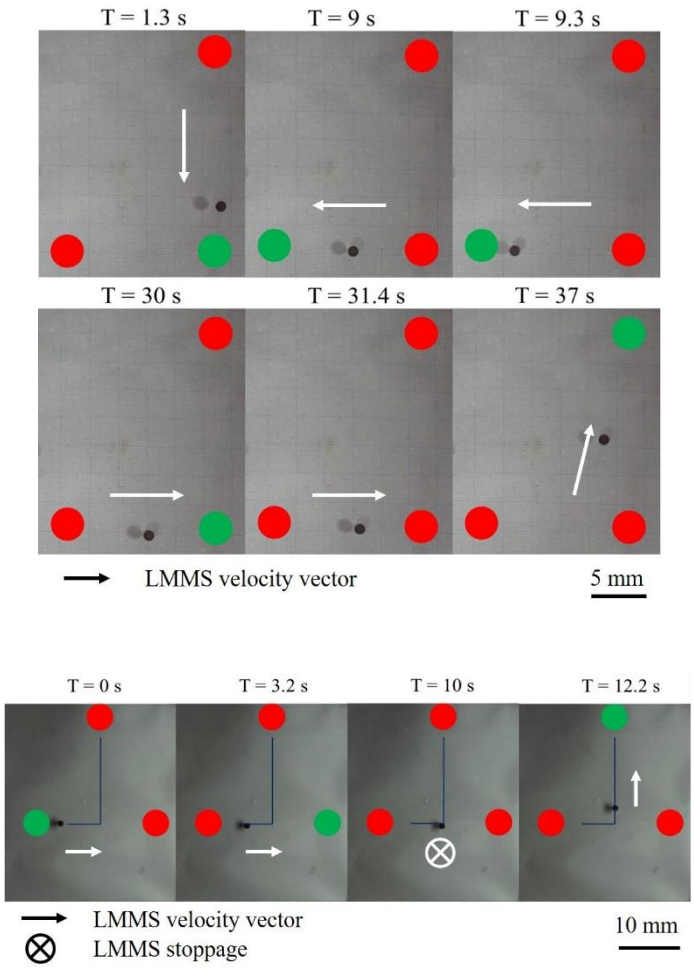

Figure S1. The 2D-manipulation of the floating LMMS under (a) DC magnetic field; (b) PWM magnetic field

Comparison of LMMS and FM under the PWM magnetic field

The comparison between the movement of LMMS and FM has been carried out for different duty cycles, volumes, and magnetic flux densities and the results are shown in Fig S2, S3, S4, respectively. These figures show that for different conditions, the LMMS always moves slower than the FM and the LMMS reaches the magnetic coil with more number of steps and fewer step lengths. Hence, the operator has more control over the LMMS relative to FM. Also, the results show that in both LMMS and FM, when the duty cycle,

volume, and magnetic flux density increase, the marble reaches the coil in a shorter time and smaller step length.

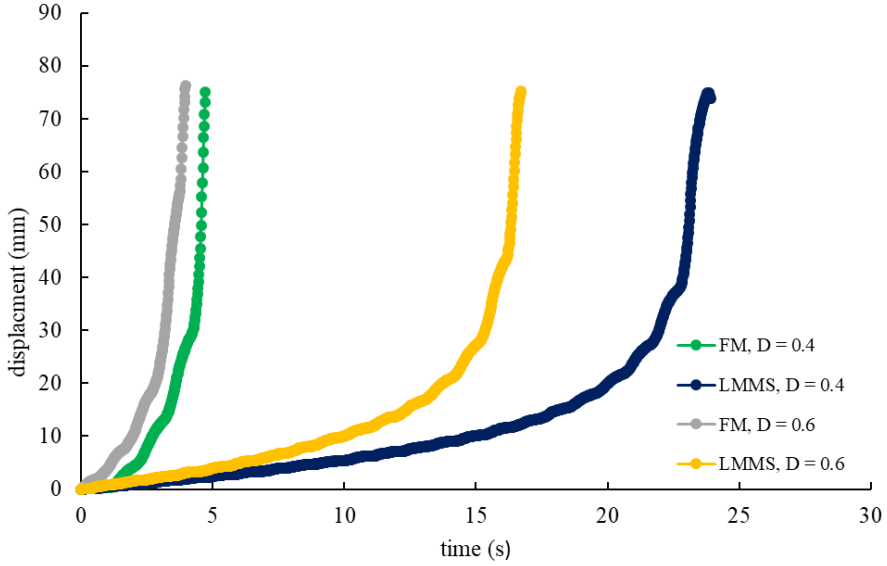

Fig S2: The LMMS and FM displacements for under PWM magnetic field over time for different duty cycles. The magnetic coil current, the volume, the initial distance, and the PWM frequency are 1 A, 20µl, 75 mm and 1 Hz, respectively.

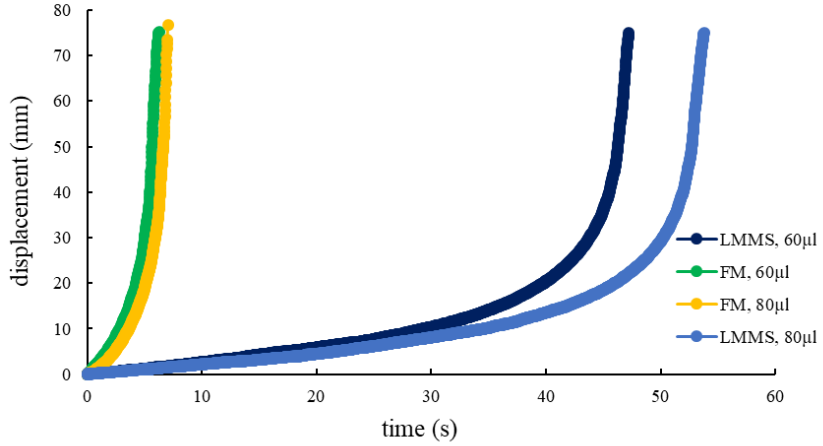

Fig S3: The LMMS and FM displacements under PWM magnetic field over time for different volumes. The magnetic coil current, the initial distance, and the PWM frequency and duty cycle are 1 A, 75 mm, 1 Hz, and 0.5 respectively.

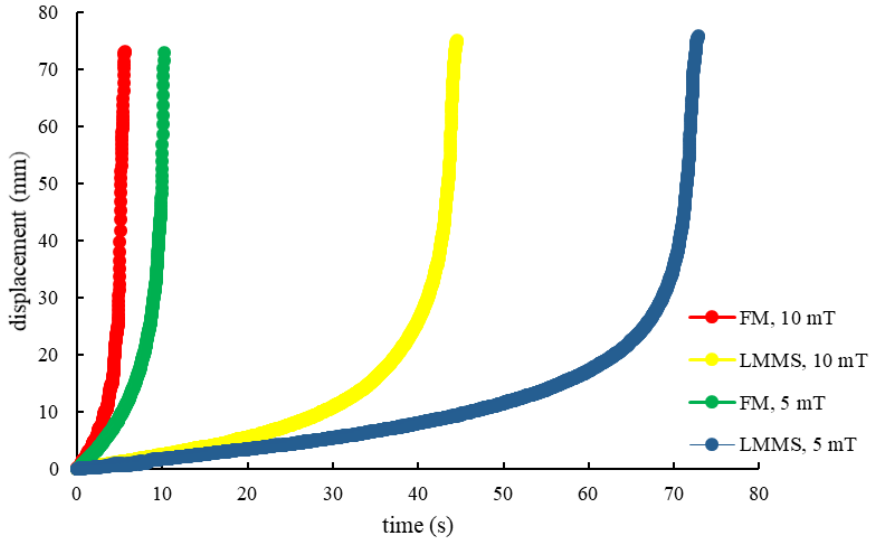

Fig S4: The LMMS and FM displacements under PWM magnetic field over time for different magnetic flux densities. The volume, initial distance, the PWM frequency and duty cycle are 20 µl, 75 mm, 1 Hz, and 0.5, respectively.

Biocompatibility Test

In this section, the cell viability and proliferation inside an LMMS and an FM were investigated and compared using the in-vitro MTT (3-[4, 5-diphenyl-2-yl]-2, 5-diphenyl-tetrazolium bromide) as an indirect evaluation of cell survival and growth. As explained, an LMMS consists of water in the core and magnetic nanoparticles on the shell, whereas an FM is a ferrofluid solution wrapped with magnetic nanoparticles. To evaluate and compare the cytotoxicity of FMs and LMMSs, the MTT assay was conducted on the magnetic nanoparticles and the ferrofluid. For this purpose, LTT99 cell line suspension at a 5000 cell/cm2 concentration were cultured in Petri dishes containing DMEM-F12 supplement with 10% fetal bovine serum (FBS) (Gibco, Gaithersburg, USA) and 1% penicillin/streptomycin (Gibco, Gaithersburg, USA). The Petri dishes were then incubated at 37 °C for two days. After passing 24 and 48 hours, the cell viability of samples was determined by measuring the optical density (OD) at 600 nm evaluated by a spectrometer using the following equation [2]:

$$Cell\ viability\ (\%) = \frac{OD\ of\ the\ sample}{OD\ of\ the\ control} \times 100$$

As illustrated in Fig. S5, having a magnetic solid/liquid could decrease the biocompatibility of the sample. To be more specific, the OD of the samples containing magnetic solid decreased by about 30% and 54% after passing 24 and 48 hours, respectively. On the other hand, the magnetic liquid (ferrofluid) reduces the cell viability substantially by 81% after 24 hr and 75% after 48 hr. This effect is because a hydrophilic solid

substrate is required for cell growth and proliferation. As a result, the ferrofluid should be replaced by less toxic solution for cells to increase the biocompatibility of the structure. The difference in the core structure of the LMMS and FM causes the LMMS to be more biocompatible.

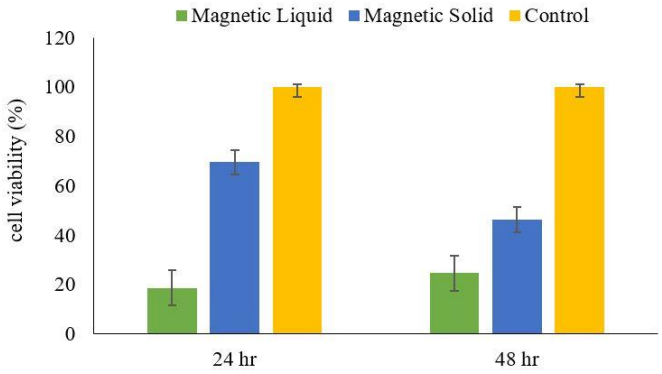

Figure S5: The cell viability assay performed on the magnetic nanoparticles (magnetic solid) and ferrofluid (magnetic liquid)

Reference

[1] Maleki, S., Shamloo, A., & Kalantarnia, F. (2022). Tubular TPU/SF nanofibers covered with chitosan-based hydrogels as small-diameter vascular grafts with enhanced mechanical properties. Scientific Reports, 12(1), 6179.

[2] Lehmann, U., Vandevyver, C., Parashar, V. K., & Gijs, M. A. (2006). Droplet-based DNA purification in a magnetic lab-on-a-chip. Angewandte Chemie International Edition, 45(19), 3062-3067.