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ANISOTROPIC MAGNETIC COMPOSITE WITH CUSTOMIZED MICROSTRUCTURATION FOR MICROFLUIDIC DEVICES

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SHORT ABSTRACT
Anisotropic magnetic composite polymers are promising materials to implement magnetophoretic functions in microfluidic devices. Besides microfabrication advantages, they offer interesting modular magnetic properties. In particular, the custom microstructuration of particles within the polydimethylsiloxane (PDMS) matrix, permits to create powerful magnetic functions. In the present work, we report that self-ordering of particles within the matrix permits either to improve existing magnetophoretic functions, or to create original ones, such as arrays of magnetic micro-traps resulting from individual 1D-magnetic flux micro-concentrators embedded in the matrix.

EXTENDED ABSTRACT
Magnetophoresis, which refers to the motion of an object in a magnetic field gradient, permits to address numerous biomedical applications when implemented in microfluidic devices [1-3]. The implementation of microconcentrators of magnetic field is particularly suitable in this context as it permits to modulate or even cancel the magnetic force in order to release on demand the trapped objects. In order to achieve high concentration of magnetic flux, the soft magnetic material should combine a large magnetization and preferentially a high effective magnetic susceptibility $\chi_{eff}$ to operate at relatively low magnetic fields, while maintaining no hysteresis losses. Moreover, a way to improve experienced magnetic forces is to pattern soft magnetic structures in the submillimeter range [4]. To overcome the technological challenges owing to heterogeneous integration of metallic material with PolyDiMethylSiloxane (PDMS), as well as limitations related to pure metals micropatterning using time consuming, costly and complex UV-LIGA process, the approach of PDMS-based composite is promising [5-7]. The first benefit concerns technological aspects. The composite preserves some PDMS properties such as soft-lithography micropatterning and O$_2$ surface activation

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for plasma bonding with glass and PDMS. In addition, the composite can also be directly integrated into microchannels avoiding cumbersome alignment procedures. The second interest relates with the magnetic properties achieved with composite. Indeed, the saturation magnetization of Carbonyl Iron - PDMS (I-PDMS) at high concentration, 83 wt% for instance, is higher than the one of pure Ni and comparable with the ones of the benchmark soft magnetic metallic alloys such as Mumetal, Supermalloy or Permalloy. The third advantage relates to the custom microstructuration of particles within the matrix. Thereby, we recently reported on the interest of microstructuring the composite by applying a magnetic field during the polymer reticulation in order to self-order particles in the polymer matrix [8]. Created agglomerates of chains of particle in the matrix induce a uniaxial magnetic anisotropy that was assessed by susceptibility measurements, over a large range of compositions, from 1 to 83 wt%. Such structure engineering can be exploited in different ways in order to create powerful magnetic functions in microsystems: either at high magnetic particle concentration, I-PDMS microstructuration permits to improve magnetophoretic efficiency, or at low concentration, in order to create arrays of individual 1D-magnetic flux micro-concentrators.

**Improved magnetophoretic efficiency through I-PDMS microstructuration**

I-PDMS microstructuration was obtained by submitting the composite to a uniform magnetic field of 130 mT during cross-linking. At high concentration, the particles form ramified chains with a global orientation along the flux lines. This induced a uniaxial anisotropy that gave rise to a nearly 20% increase in the susceptibility. In order to study the resulting impact on generated magnetophoretic force in microfluidic device, we developed a microfluidic channel comprising a wall made of I-PDMS. For one batch of devices we implemented isotropic I-PDMS and for the second batch anisotropic I-PDMS. A schematic of the channel is represented on Fig. 1a. Microbeads flowing in the channel were submitted to different forces: magnetophoretic force $\overline{F}_m$, fluidic drag force $\overline{F}_d$, gravitational force $\overline{F}_g$, and buoyancy forces $\overline{F}_{buo}$. According to the Newton’s second law, motion of a magnetic bead in a laminar flow can be determined by the balance of forces:

$$m_b \frac{d\overline{v}_b}{dt} = \overline{F}_m + \overline{F}_d + \overline{F}_g + \overline{F}_{buo}$$

(1)

$m_b$ being the mass of the bead and $\overline{v}_b$ its velocity. Generally, the inertial term and gravitational and buoyancy forces are neglected, which was confirmed in the particular case of our study. In order to measure the magnetophoretic force experienced by the beads in the channel for each batch of devices we implemented the following protocol. The channel was first filled up with beads. By stopping the flow rate (fluid velocity, $\overline{v}_f = 0$), the initial position of the beads in the channel was defined. The Plexiglas™ holder, that integrated two permanent magnets, was then positioned over the microchannel. Submitted to a magnetic field gradient, the superparamagnetic beads were attracted toward the I-PDMS composite wall of the channel. Fig. 1b displays a stacking of video frames revealing microbeads trajectories. From beads velocities measurements, and based on Eq.(2), we mapped the magnetophoretic force in the channel:

$$\overline{F}_m = \mu_0 V_b (\overline{M}_b \cdot \nabla) \overline{H}_a = 6 \pi \eta R_b \overline{v}_b f_D$$

(2)

$H_a$ being the applied magnetic field, $V_b$ the volume of the bead, $R_b$ its radius, $M_b$ its magnetization, which can be expressed as $\overline{M}_b = f (H_a) \overline{H}_a$, $\eta$ the viscosity of the solution and $f_D$ the drag coefficient of the particle that accounts for the influence of a solid wall in the vicinity of the particle [9]. Fig. 1c shows the deduced $F_m$ at different distances from the composite wall, i.e., 150 µm, 200 µm and 250 µm. At a distance of 150 µm form the microconcentrator, we measured a force reaching 60 pN using the anisotropic I-PDMS, a force twice as large as the one measured using isotropic composite. Based on the measured variation of the magnetophoretic force and on finite element simulations, we highlighted that the benefit of using anisotropic composite does not only rely on the global
susceptibility increase, but also on local magnetic field gradients originating from its fine periodic microstructure [10].

Figure 1: (a) Schematic of the microfluidic device, (b) Z-stack projection of videomicroscopic images representing superparamagnetic particles moving toward the composite wall of the microsystem. Inset: Schematic of forces experienced by microbeads in the microchannel, (c) Mean magnetic force generated by anisotropic (red, left bars) or isotropic (blue, right bars) composite microconcentrators at distances of 150 µm, 200 µm and 250 µm from the composite. The right Y-axis shows the corresponding value of field gradient. Inset: force ratio at these distances.

Array of 1D magnetic flux concentrators

In contrast, at low particle concentrations (1 and 5 wt%), the applied field during the PDMS cross-linking leads to the formation of well-defined and isolated chains, as revealed by X-Ray Tomography. We observed the coexistence of two types of organizations: chain-like agglomerates and isolated clusters of particles concentrated at the bottom surface of the composite membrane, as shown on Fig. 2a. Considering the membrane surface, the two types of organizations gave rise to two populations of traps of average diameter of 1.5 and 6.7 µm, larger ones mostly corresponds to buried particle chains. We took advantage of the 1D magnetic flux concentrator created by individual chain-like agglomerate, and able to generate high gradients of magnetic fields at the micrometer scale. We integrated 1 wt% and 5 wt% I-PDMS membranes in microfluidic systems and studied their magnetic trapping performances.

Figure 2: 1 wt% I-PDMS composite : (a) X-Ray tomography of composite membrane, (b) traps on the surface of the composite membrane before and after beads trapping.

Micrometer trap enabled to isolate magnetic beads and to organize them on large surfaces. Fig. 2b reports microscopic images of microfluidic channel integrating a trapping membrane (1 wt%), before and after beads injection at 250 µL/h. Beads were trapped in presence of an external magnetic field. We tested the persistence of trapping when increasing the flow rate. Using 5 wt% composite, 50 % of beads remain trapped at 2 mL/h. We have also shown that these dense arrays of traps permit to create a monolayer of beads, with a density of 1200 beads/mm², 89 % of large traps being occupied. This original array of 1D magnetic flux concentrator is promising as the implementation is quite simple while it permit to address biological characterization that require the controlled immobilization of a target population, such as drugs assays.
Conclusion

Anisotropic I-PDMS is a promising material in the general context of microfluidic devices, in particular regarding the keen interest for lab on a chip dedicated to biomedical applications. This material permits to solve issues linked to heterogeneous integration of metallic material with polymers. Most interestingly the custom microstructuration of particles within the matrix allows to create magnetic anisotropy. Such structure engineering can be exploited in different ways in order to create powerful magnetic functions in microsystems. At high composite concentration we demonstrated that anisotropic I-PDMS allows to increase magnetophoretic performances compared to isotropic I-PDMS. At low concentration, we created arrays of 1D-magnetic flux micro-concentrators through chain like microstructures diluted in the matrix. These original structures were used as traps in microfluidic devices.

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