Electronic Supplementary Material (ESI) for Soft Matter.

Supplementary Information for:

Understanding and overcoming shear alignment of fibers during extrusion

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Calculation of calcium phosphate fiber susceptibility (χ_{ps})

The susceptibility of calcium phosphate (CaP) fibers was calculated using two methods: 1) SQUID magnetromy and 2) calculate surface coverage of Fe_3O_4 based on the experimental procedure.

1) Using SQUID magnetromy, a 1 mg sample of coated CaP fibers was measured. The susceptibility of the sample at low fields was found to be 0.07. However, the magnetic energy term in the paper takes into account the susceptibility of the magnetic shell, and not the entire particle. From SQUID it is only know that:

$$\phi_{Fe304}\chi_{Fe304} + \phi_{CaP}\chi_{CaP} = 0.07 \tag{S1}$$

Where ϕ and χ represent the volume fraction and unitless volume susceptibility, respectively. In this case, χ_{Fe304} was measured to be 14 using SQUID magnetometry on pure iron oxide nanoparticles and χ_{CaP} was taken as zero since it has a negligible susceptibility. The maximum volume percentage of iron oxide in a particle assembly is determined when the volume of the ellipsoidal shell around a particle is completely filled with iron oxide:

$$\phi_{Fe304-max} = \frac{V_{shell}}{V_{CaP} + V_{Shell}} = 0.021 \tag{S2}$$

Here the volume of the shell (V_{shell}) is equal to:

$$V_{shell} = V_{Total} - V_{CaP} \tag{S3}$$

Where $V_{Total} = \frac{4}{3}\pi(A+d)(B+d)^2$ and $V_{CaP} = \frac{4}{3}\pi AB^2$ (S4)

Here A and B are the long radii and short radii of the ellipsoid, respectively, while d is the diameter of the Fe_3O_4 nanoparticles coating the surface (12 nm). The calculations were based on the average fiber dimensions (A = 20 μm, B = 1 μm). Dividing $\phi_{Fe304-max}$ from S2 by the measured ϕ_{Fe304} from S1 leads to an approximate surface coverage of 21.2%. The susceptibility of the shell can then be calculated as:

$$\chi_{ps} = \chi_{Fe304}\phi_{Fe304} \tag{S5}$$

Using equation S5, the susceptibility of the shell is found to be approximately 2.97.

2) In addition, the susceptibility of the CaP fibers was also calculated based on the total surface area of the CaP fibers and the amount of iron oxide added to the solution during the labeling process. Using the density of CaP (3.18 g/ml) and taking the average fiber dimension to be A = 20 µm, B = 1 µm, the number of fibers/gram can be calculated according to:

$$\rho_{CaP\ fiber} V_{CaP\ fiber} = m_{Cap\ fiber} \tag{S6}$$

$$\frac{m_{CaP}}{m_{Cap\ fiber}} = N_{CaP\ fibers} \tag{S7}$$

The number of iron oxide nanoparticles added to the solution of water and CaP can be calculated based on the original 3.9 vol. % solids content. Therefore, the number of nanoparticles and CaP fibers is known, and an approximate coverage can be determined by:

$$\frac{N_{Fe3O4\ particles}}{N_{CaP\ fibers}} = K \tag{S8}$$

Here, K represents the number of particles per individual CaP fiber. Assuming that all of the Fe_3O_4 particles form a monolayer on the surface of the fibers, the filled percentage of the magnetic shell can be calculated as:

$$\frac{K * V_{np}}{V_{shell}} = \sim 18.8\% \tag{S9}$$

Interpreting the filled percentage of the magnetic shell to be approximately equal to the surface coverage, a surface coating of roughly 18.8% is calculated, leading to a shell susceptibility (χ_{ps}) = 2.63. The average of these two methods yields an approximate susceptibility χ_{ps} = 2.8. Good agreement between the numerical simulations discussed in Fig. 3 of the paper and experiments further validate the use of 2.8 as the shell susceptibility. These values were further corroborated with SEM micrographs and image analysis.

Supplementary Videos

The supplementary videos show the real-time response of magnetized calcium phosphate (CaP) rods suspended in isobornyl acrylate (IBOA). The suspension is flowed at 1 ml/hr through a 1mm diameter capillary.

Videos **S1** and **S2** show the real-time response of CaP fibers subjected to pipe flow without an applied field. It is clear that while shear forces tend to align the fibers in the direction of flow, there is disorder caused by Jeffery Orbits as well as particle-wall interactions. Video S1 and S2 were recorded using transmitted light microscopy at 10x and 20x, respectively.

Videos **S3** and **S4** show the real-time response of CaP fibers subjected to pipe flow with an applied field of approximately 120 mT in the direction of flow. The video depicts how the magnetic field is able to pin the Jeffery Orbits and create better alignment of CaP fibers. Video S3 and S4 were recorded using transmitted light microscopy at 10x and 20x, respectively.