Supplementary Information (SI) for RSC Applied Polymers.

Reversible magnetic soft actuators of thermoplastic polyurethane with Yttrium Iron Garnet

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1. Magnetic properties of the commercial and sol-gel synthesised YIG powders

Figure S1 depicts the hysteresis loops of YIG powders. Commercial powder (YIG < 100 nm particle size, 99.9 % trace metal basis, Sigma Aldrich, MO, USA) presents lower saturation magnetization (M_s) than sol-gel synthesised YIG particles (Table S1). Calcination at 1000 °C increases M_s of all powders.

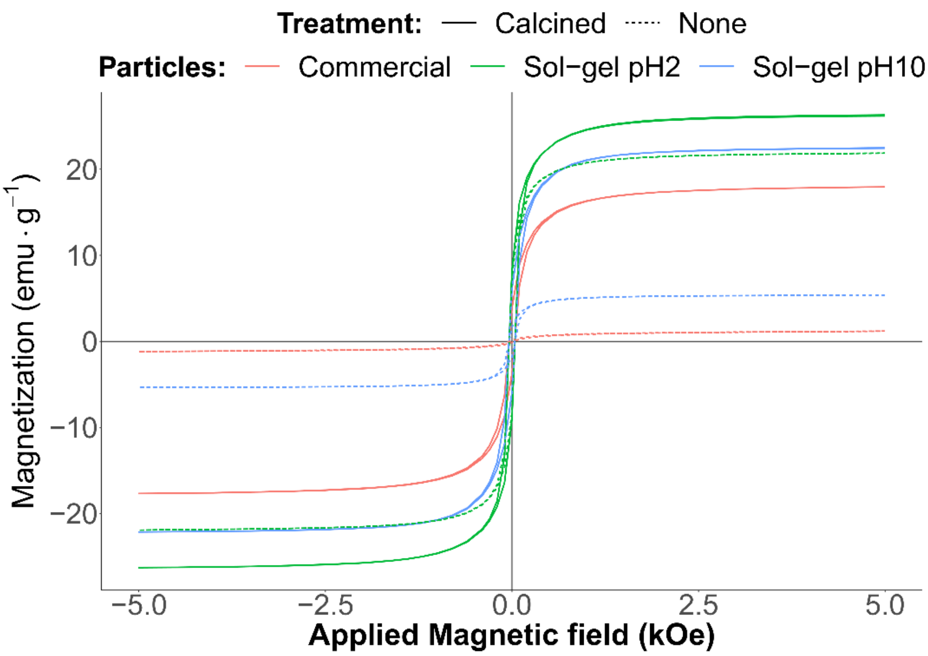

Fig. S1 Magnetic hysteresis loops, measured in a vibrating sample magnetometer (VSM), at room temperature, of the commercial and sol-gel synthesised YIG powders.

Table S1. Saturation magnetization (M_s, measured at 10 kOe, with an error of 0.1 emu·g^{-1}), total mass (m, with an error of 0.001 mg), coercivity (H_c, with an error of 1 Oe) and reduced remanence ($m_r = M_r/M_s$, with an error of 0.01, where M_r is the remanent magnetization value) of commercial and sol-gel synthesised YIG powders, before and after calcination at 1000 °C

	Commercial YIG		Sol-gel synthesised YIG			
			pH 10		pH 2	
	Untreated	Calcined	Untreated	Calcined	Untreated	Calcined
M_s (emu·g^{-1})	1.4	18.1	5.5	22.5	22.0	21.9
m (mg)	8.186	6.323	6.507	9.501	5.329	5.235
H_c (Oe)	31	37	39	40	38	47
m_r	0.10	0.21	0.31	0.27	0.36	0.32

2. Raman spectra of mTPU composites and morphology

Figure S2 depicts the Raman spectra of mTPU. The vertical dashed lines indicate the characteristic Raman shifts of YIG.

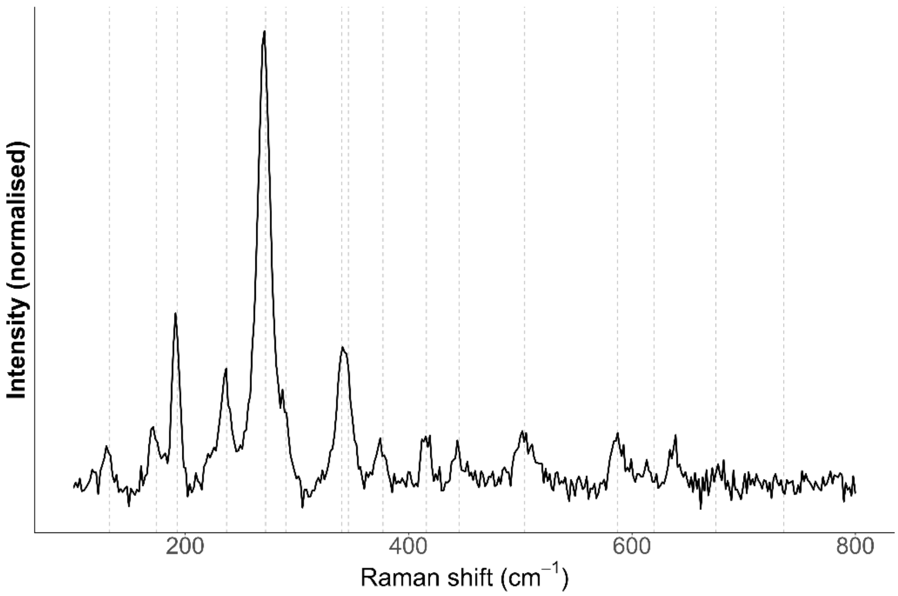

Fig. S2 Averaged Raman spectra of mTPU composites, vertical dashed lines indicate the characteristic Raman shifts of YIG.

Figure S3 shows a scanning electron microscopy (SEM) image of the mYPU composite, with measurements of the YIG agglomerates' size, illustrating a size range between 2 and 19 µm.

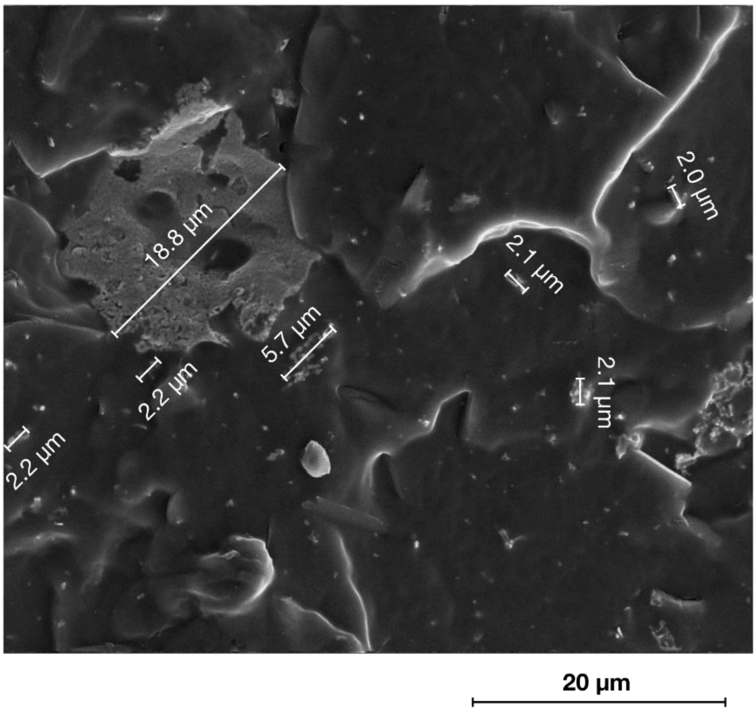

Fig. S3 SEM image (in secondary electron mode) of mTPU composite, with measurements of YIG agglomerates.

3. Simulations using COMSOL Multiphysics®

A 3D model was created to study the magnetic flux density (*B*) generated by a cylindrical permanent magnet with 5 mm in diameter and 10 mm in length, surrounded by air. The material used for the permanent magnet was *Sintered NdFeB (N45 grade)*, from the *Hard Magnetic Materials*' folder of the *AC/DC Material Library* of *COMSOL Multiphysics®* [1], with a remanent flux density norm of 1.35 T and a recoil permeability of 1.05. The surrounding air was modelled with a relative permeability of 1. The boundary of the model was set as an open boundary (using the 'Magnetic Insulation' condition) to simulate the behaviour of the magnetic field in free space. The boundary condition ensures that the normal component of the magnetic flux density at the outer boundary is zero. North and south magnetic poles were defined at the opposite ends (boundaries) of the cylindrical permanent magnet. The element size of the mesh was set to *finer*. *B* was calculated using the COMSOL *Magnetic Fields* module, solving the magnetostatic Maxwell's equations. Figure S4 shows the resulting magnetic field distribution, as a function of the distance from the magnet's end, along the axial direction.

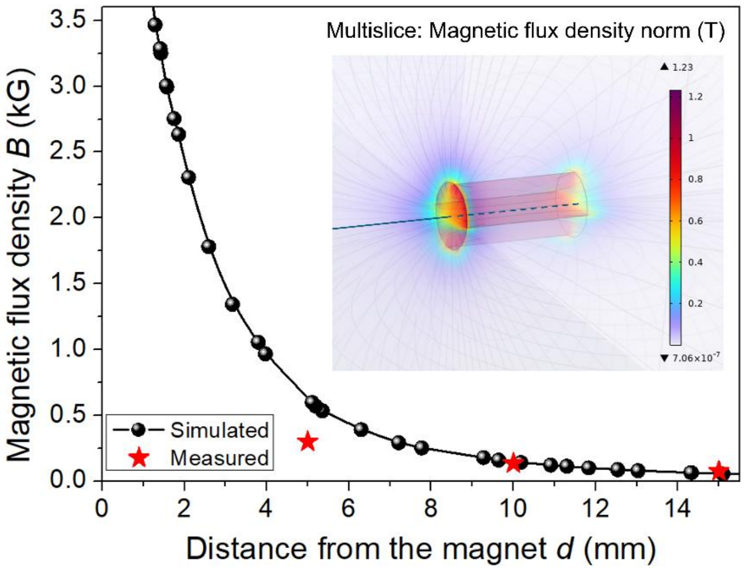

Fig. S4 Magnetic flux density (B) values simulated using COMSOL Multiphysics® (black dots) and measured using a gaussmeter (red stars) along the axis of the magnet, as a function of the distance d from the magnet's end. Inset shows a snapshot of the multislice magnetic flux density norm (1 T = 10 kG) plotted using COMSOL.

For the bending of mTPU rectangular tapes, a 3D model was created using the COMSOL *Solid Mechanics* module. A new material was created following the experimentally obtained values of the mTPU composites, with a Young's modulus of 122 MPa, a density of 940 kg/m^3 (calculated from the total mass and dimensions of the mTPU cylindrical pellet measured at VSM) and an estimated Poisson's ratio of 0.3 (assuming 0.45 for TPU, 0.2 for YIG, TPU density of 1.2 g·cm^{-3}, YIG density of 5.2 g·cm^{-3}, and a YIG wt.% of 10). The displacement of the upper free end of the tape was then studied as a function of the total applied force at one of the lateral boundaries of the upper domain, using the linear elastic material model. A free tetrahedral mesh with finer size was used for all domains.

Finally, for the estimation of the magnetic force acting on the upper domain of the tape, a 3D model was created using the COMSOL *Magnetic Fields, No Currents* module. Apart from the parameters mentioned before, we also estimated the relative permeability (μ_r) of the mTPU composite, using the M(H) values from the magnetic hysteresis measurements and the following relations [2]:

$$\mu_r = \frac{\mu}{\mu_0} = \frac{B}{\mu_0 H} = \frac{\mu_0(H + M)}{\mu_0 H} = 1 + \frac{M}{H}$$

where μ is the magnetic permeability of the material and μ_0 the vacuum permeability (4x10^{-7} H·m^{-1}). M and H are measured in A·m^{-1} using the following relations: 1 emu·cm^{-3} = 10^3 A·m^{-1}, and 1 Oe = 1 G = $(4\pi)^{-1}$ x 10^3 A·m^{-1}. Although an average value between 1 and 1.1 was obtained, the simulations were performed with μ_r = 13, which gave a better approximation to the experimentally measured values (Figure 9 a in the main text). The total magnetic force on the top domain of the tape was estimated by integrating the Maxwell stress tensor over the selected boundary area at different distances (along the x-axis) from the magnet. The x-component of the force was then applied to the lateral surface of the upper segment, as illustrated in Figure 9 c of the main text. Planar views of the geometry used are depicted in Figure S5.

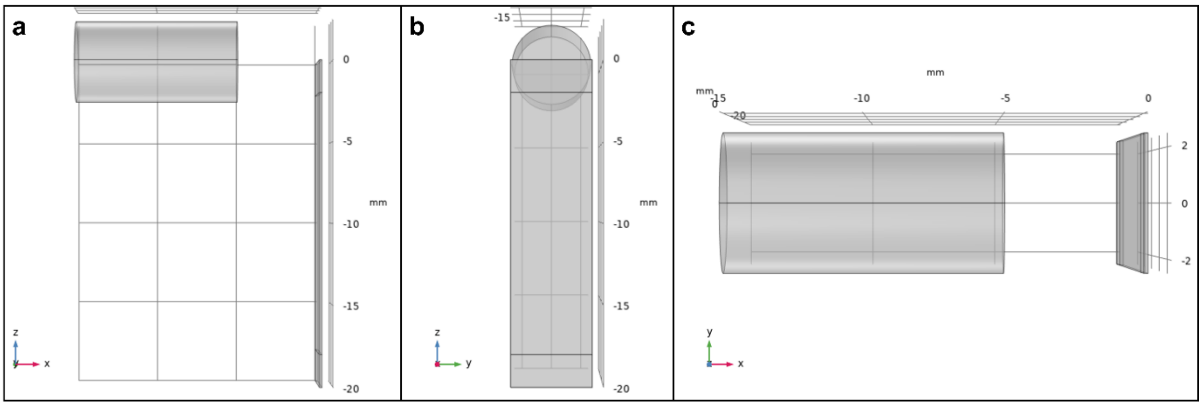

Fig. S5 COMSOL snapshots illustrating the geometries used for the cylindrical permanent magnet and the 150 μm-thick rectangular tape at different planar views: a) xz-plane, b) yz-plane and c) xy-plane.

4. Deformation of the mTPU/Au bi-layer actuator

The deformation of the bi-layer mTPU/Au actuator, upon the approach and withdrawal of a permanent magnet (5 mm ⌀ × 10 mm length) is depicted in Figure S6.

Fig. S6 Deformation of the mTPU/Au bi-layer upon the approach and withdrawal of a permanent magnet.

References

[1] COMSOL Multiphysics® v. 6.2. www.comsol.com. COMSOL AB, Stockholm, Sweden.

[2] S. Chikazumi and C. D. Graham, *Physics of Ferromagnetism*, Clarendon Press, Oxford, 1997.