

SUPPLEMENTARY INFORMATION

Hybrid magnetorheological elastomers enable versatile soft actuators

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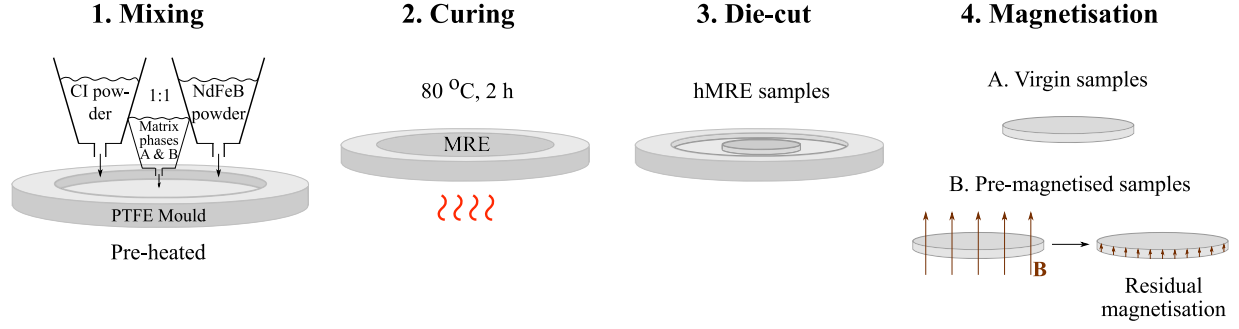
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Supplementary Methods 1: Materials and synthesis

The extremely soft hybrid MREs are manufactured combining three components: an elastomeric matrix, soft-magnetic particles and hard-magnetic particles. The volume ratio between both types of particles (soft-to-hard-magnetic particles volume ratio in the following) is modified according to the values of 1:0, 2:1, 1:1, 1:2 and 0:1. The total amount of magnetic particles, however, is fixed at 30 % volume ratio for all the specimens. Table 1 summarises the manufacturing conditions of the MREs. The soft-magnetic particles consist of soft SQ carbonyl iron powder (CIP) (BASF, Germany). The hard-magnetic particles consist of NdFeB powder (MQP-S-11-9-grade powder, Neo Materials Technology Inc., Greenwood Village, Colorado, United States). Both types of particles are spherical-shaped, but they differ in size. While the CIP has a mean diameter of 3.9 - 5 μm , NdFeB particles are larger with a mean diameter of 35 - 55 μm . For the matrix, an extremely soft elastomer Dowsil CY52-276 (DowSil, Midland, MI, USA) (PDMS with stiffness below 10 kPa) is used. Such soft matrices have been proved ideal to activate the magneto-mechanical response with small magnetic fields [1, 2]. To initiate the crosslinking, the two elastomeric phases are mixed at the ratio 1:1. Supplementary Figure 1 shows the steps followed to manufacture the samples. Note that the initial blend is pre-heated to prevent the sedimentation of the NdFeB powder. A final step allows for the pre-magnetisation of the specimens. For further detail on the preparation method, the reader is referred to [2].

Supplementary Table 1. Manufacturing conditions of the MREs according to the soft-to-hard-magnetic particles volume ratio.

Soft-to-hard particles ratio [-]	$\alpha_{soft-mag}$ [vol.%]	$\alpha_{hard-mag}$ [vol.%]	α_{total} [vol.%]
1:0	30	0	30
2:1	20	10	
1:1	15	15	
1:2	20	10	
0:1	0	30	



Supplementary Figure 1. **Scheme of the synthesis process of the MREs.** The manufacturing process is composed by: (1) mixing of the components; (2) curing in oven; (3) die-cutting of 1 mm height and 20 mm diameter geometries; (4) pre-magnetisation of specimens with homogeneous magnetic fields.

20 **Supplementary Methods 2: Computational framework**

The structural response of hybrid MREs is the outcome of microstructural interactions between the soft- and hard-magnetic particles and the carrier matrix. In order to account for such interplays and to obtain the homogenised material response, we extend the formulation proposed by the authors in [3] to account for the magnetic remanence of the particles. We postulate the model of the

25 hard-magnetic particles according to a non-dissipative simplification of the framework reported by Rambauck et al. [4].

Kinematics and balance laws

The constitutive framework is formulated considering finite deformations due to the ability of the MREs used in this work to undergo large strains. A deformation map $\mathbf{x} = \varphi(\mathbf{X})$ links the

30 continuum coordinates in the reference configuration $\mathbf{X} \in \Omega_0$ with the current configuration $\mathbf{x} \in \Omega$. Therefore, the displacement field is defined as $\mathbf{u} = \varphi(\mathbf{X}) - \mathbf{X}$. The gradient of this primary field yields the deformation gradient as

$$\mathbf{F} = \nabla_0 \varphi = \nabla_0 \mathbf{u} + \mathbf{I}, \quad J := \det \mathbf{F} \geq 0, \quad (1)$$

where ∇_0 is the gradient operator with respect to the reference configuration and $\mathbf{I}$ is the second-order identity tensor. Alternatively, one may use rotation-free strain tensors, namely the left and

35 right Cauchy-Green tensors, respectively

$$\mathbf{b} := \mathbf{F} \cdot \mathbf{F}^T, \quad \mathbf{C} := \mathbf{F}^T \cdot \mathbf{F}. \quad (2)$$

The conjugate fields $\mathbf{P}$ and $\boldsymbol{\sigma}$ are the Piola and Cauchy stress tensors, respectively referred to the reference and current configurations. These are related according to

$$\boldsymbol{\sigma} = J^{-1} \mathbf{P} \cdot \mathbf{F}^T. \quad (3)$$

The magnetic problem incorporates three main additional variables. These are the magnetic field intensity $\mathbb{H}$, the magnetisation $\mathbb{M}$ and the magnetic induction $\mathbb{B}$ in the reference configuration, and
40 their counterparts $\mathbb{h}$, $\mathbb{m}$ and $\mathbb{b}$ in the current configuration. Both configurations are related by

$$\mathbb{h} = \mathbf{F}^{-T} \cdot \mathbb{H}, \quad \mathbb{b} = J^{-1} \mathbf{F} \cdot \mathbb{B}. \quad (4)$$

The magnetic vectors are connected through the standard constitutive relation

$$\mathbb{b} = \mu_0 [\mathbb{h} + \mathbb{m}]. \quad (5)$$

The field equation for the mechanical displacement is

$$\nabla_0 \cdot \mathbf{P} = \mathbf{0}. \quad (6)$$

For the magnetic problem, the Maxwell equations for the magnetostatic case state that

$$\nabla_0 \times \mathbb{H} = \mathbf{0}, \quad \nabla_0 \cdot \mathbb{B} = 0. \quad (7)$$

Note that 7₁ is automatically satisfied when $\mathbb{H}$ is derived from a magnetic scalar potential field ϕ

45 as

$$\mathbb{H} = -\nabla_0 \phi, \quad \text{in } \Omega_0. \quad (8)$$

Constitutive formulation at the microscale

In the present work, we make use of a single total energy density function Ψ to describe the magneto-mechanical constitutive behaviour of both the matrix and magnetic fillers at the microstructural level. Given the nearly incompressible nature of the polymeric matrix, and the larger
50 stiffness of the magnetic particles with respect to the matrix, both phases are considered fully incompressible. The energy function is based on three contributions, i.e., mechanical, Maxwell and magnetisation energies, according to

$$\Psi(\mathbf{F}, \mathbb{H}) = \Psi_{\text{mech}}(\mathbf{F}) + \Psi_{\text{maxw}}(\mathbf{F}, \mathbb{H}) + \Psi_{\text{mag}}(\mathbf{F}, \mathbb{H}). \quad (9)$$

For the description of the elastic energetic contribution, the Neo-Hookean formulation is chosen

$$\Psi_{\text{mech}}(\mathbf{F}) = \frac{G}{2} [I_1 - 3], \quad (10)$$

with $I_1 = \text{tr}(\mathbf{F} \cdot \mathbf{F}^T)$ and G the shear modulus of the considered phase.

55 With respect to the magnetic contributions, the Maxwell energy describes the background magnetic energy due to the non-zero magnetic permeability of the vacuum as

$$\Psi_{\text{maxw}}(\mathbf{F}, \mathbb{H}) = -\frac{\mu_0}{2} I_5^e \quad (11)$$

with the energetic magnetic invariant¹ defined as

$$I_5^e = \mathbb{h} \cdot \mathbb{h} = \mathbf{F}^{-T} \cdot \mathbb{H} \cdot \mathbf{F}^{-T} \cdot \mathbb{H}. \quad (12)$$

The contribution of the magnetisable media is additively decomposed in an energetic term $\Psi_{\text{mag,e}}$

¹The superscript “e” means that the invariant depends only on the energetic magnetic field $\mathbb{h}$, and not on the remanent contribution $\mathbb{h}_r$.

and a remanent contribution $\Psi_{\text{mag,r}}$ as

$$\Psi_{\text{mag}}(\mathbf{F}, \mathbb{H}) = \Psi_{\text{mag,e}} + \Psi_{\text{mag,r}} = -\mu_0 \left[\frac{m_s^2}{\chi_e} \log \left(\cosh \left(\frac{\chi_e}{m_s} \sqrt{I_5^e} \right) \right) \right] + \mu_0 [1 + \chi_e] I_5^{\text{er}} + \frac{\mu_0}{2\chi_r} I_5^{\text{r}}, \quad (13)$$

60 where χ_e and χ_r are the energetic and remanent magnetic susceptibility, respectively, and m_s is the magnetic saturation of the magnetization curve of the corresponding phase.²

I_5^{er} is the magnetic invariant presented in [5]. It accounts for the energetic and remanent magnetic fields in the Eulerian configuration of the medium and it is defined as ³

$$I_5^{\text{er}} = \mathbb{h} \cdot \mathbb{h}_r = \mathbb{H} \cdot \mathbf{C}^{-1/2} \cdot \mathbb{H}_r. \quad (14)$$

Finally, the invariant I_5^{r} only considers the remanent field, reading as

$$I_5^{\text{r}} = \mathbb{h}_r \cdot \mathbb{h}_r = \mathbb{H}_r \cdot \mathbb{H}_r. \quad (15)$$

Note that the last term of Eq. 13 vanishes when differentiating with respect to $\mathbf{F}$ and $\mathbb{H}$. It would be necessary to define a dissipative magnetic response (i.e., evolution of $\mathbb{H}_r$ as an internal variable) to get the contribution of this term. In this work, we take the residual magnetisation constant and, therefore, such an energetic term is negligible.

The Piola stress tensor can be derived from the total energy potential and the incompressibility condition as

$$\begin{aligned} \mathbf{P}(\mathbf{F}, \mathbb{H}) &= \frac{\partial \Psi}{\partial \mathbf{F}} - p \mathbf{F}^{-\text{T}} = \mathbf{P}_{\text{mech}} + \mathbf{P}_{\text{maxw}} + \mathbf{P}_{\text{mag,e}} + \mathbf{P}_{\text{mag,r}} = \\ &= \mu \mathbf{F} - p \mathbf{F}^{-\text{T}} - \frac{\mu_0}{2} \frac{\partial I_5^e}{\partial \mathbf{F}} - \mu_0 \frac{m_s}{2\sqrt{I_5^e}} \tanh \left(\frac{\chi_e}{m_s} \sqrt{I_5^e} \right) \frac{\partial I_5^e}{\partial \mathbf{F}} + \mu_0 [1 + \chi_e] \frac{\partial I_5^{\text{er}}}{\partial \mathbf{C}} : \frac{\partial \mathbf{C}}{\partial \mathbf{F}} \end{aligned} \quad (16)$$

²For the matrix $\chi_e = 0$, for the soft-magnetic particles $\chi_e = 30$ and for the hard-magnetic particles $\chi_e = 0.05$. The parameter χ_r does not affect our simulations as the remanent magnetic field is assumed constant during the tests.

³The magnetic field is additively decomposed into an energetic and a remanent part as $\mathbb{H} = \mathbb{H}_e + \mathbb{H}_r$.

65 with

$$\frac{\partial I_5^e}{\partial \mathbf{F}} = -2 [\mathbf{F}^{-T} \cdot \mathbb{H}] \otimes [[\mathbb{H} \cdot \mathbf{F}^{-1}] \cdot \mathbf{F}^{-T}]. \quad (17)$$

The variable $-p\mathbf{F}^{-T}$ is a Lagrange multiplier that accounts for the incompressibility condition. In turn, the derivative of I_5^{er} reads as

$$\begin{aligned} \frac{\partial I_5^{\text{er}}}{\partial \mathbf{C}} = & -\frac{4}{\Delta} \left[I_1^{\text{U}} \mathbf{C}^{1/2} \text{sym}(\mathbb{H} \otimes \mathbb{H}_r) \mathbf{C}^{1/2} - (I_1^{\text{U}})^2 [\mathbf{C}^{1/2} \text{sym}(\mathbb{H} \otimes \mathbb{H}_r) + \text{sym}(\mathbb{H} \otimes \mathbb{H}_r) \mathbf{C}^{1/2}] + \right. \\ & [I_1^{\text{U}} I_2^{\text{U}} - I_3^{\text{U}}] [\mathbf{C}^{1/2} \text{sym}(\mathbb{H} \otimes \mathbb{H}_r) \mathbf{C}^{-1/2} + \mathbf{C}^{-1/2} \text{sym}(\mathbb{H} \otimes \mathbb{H}_r) \mathbf{C}^{1/2}] + [(I_1^{\text{U}})^3 + I_3^{\text{U}}] \text{sym}(\mathbb{H} \otimes \mathbb{H}_r) - \\ & (I_1^{\text{U}})^2 I_2^{\text{U}} [\text{sym}(\mathbb{H} \otimes \mathbb{H}_r) \mathbf{C}^{-1/2} + \mathbf{C}^{-1/2} \text{sym}(\mathbb{H} \otimes \mathbb{H}_r)] + \\ & \left. [(I_1^{\text{U}})^2 I_3^{\text{U}} + [I_1^{\text{U}} I_2^{\text{U}} - I_3^{\text{U}}] I_2^{\text{U}}] \mathbf{C}^{-1/2} \text{sym}(\mathbb{H} \otimes \mathbb{H}_r) \mathbf{C}^{-1/2} \right], \end{aligned} \quad (18)$$

and $\Delta = 8 [I_1^{\text{U}} I_2^{\text{U}} - I_3^{\text{U}}] I_3^{\text{U}}$. Note that I_1^{U} , I_2^{U} and I_3^{U} are the three principal invariants of $\mathbf{U} = \mathbf{C}^{1/2}$. Likewise, the magnetic induction is obtained from the energy density function as

$$\mathbb{B}(\mathbf{F}, \mathbb{H}) = -\frac{\partial \Psi}{\partial \mathbb{H}} = \mu_0 \left[1 + \frac{m_s}{\sqrt{I_5^e}} \tanh \left(\frac{\chi_e}{m_s} \sqrt{I_5^e} \right) \right] \mathbf{F}^{-1} \cdot \mathbf{F}^{-T} \cdot \mathbb{H} - \mu_0 [1 + \chi_e] \mathbb{H}_r \mathbf{C}^{-1/2}. \quad (19)$$

70 In the above expression, one can read the contribution of a linear term proportional to the magnetic intensity vector (background Maxwell contribution) and a term due to the magnetisation of the medium by the hyperbolic tangent profile. Note that the remanent magnetic field vector $\mathbb{H}_r$ is imposed as a constant within the hard-magnetic particles. It can be straightforwardly understood that the prescription of $\mathbb{H}_r$ leads to a permanent magnetisation with opposite sign.

75 Setting $\chi_e = 0$ in the previous expressions gives the response of a non-magnetic material (e.g., the polymeric incompressible matrix phase). In this scenario, the magnetic induction simplifies to $\mathbb{B} = \mu_0 \mathbb{H}$, i.e., the magnetisation becomes $\mathbb{M} = \mathbf{0}$.

Supplementary Table 2. Mechanical and magnetic properties used for matrix and particle behaviours.

Phase	Mechanical properties	Magnetic properties		
	Elastic shear modulus G [kPa]	Magnetic susceptibility χ [-]	Saturation magnetization $\mu_0 m_s$ [T]	Remanent magnetic field $\mu_0 \mathbb{H}_r$ [T]
Matrix	1.03	0	-	-
Soft-mag. particles	$81.78 \cdot 10^6$	30	2.5	-
Hard-mag. particles	$81.78 \cdot 10^6$	0.05	-	0.015

All variables are taken directly from experimental data, either originated from this work or from the literature. Regarding the magnetic remanence, it has been evaluated measuring the apparent residual magnetic field in pre-magnetised hMRE samples (not fully magnetised but imposing a 1 T field). This experimental value has been taken to fit the residual magnetisation of the particles comparing it with the homogenised residual magnetisation under null external field conditions.

Supplementary Methods 3: Computational homogenisation formulation

The homogenisation framework solves the magneto-mechanical equilibrium at the microscale and homogenises the results to macroscale variables. To this end, the RVE is created to be periodic at the boundaries of the domain Ω_0 . However, inside the domain the arrangement of the phases is random. This idealises a infinite random arrangement of particles by a repetition of the RVE.

The primary variables of the homogenisation problem are the microscopic displacement field $\mathbf{u}(\mathbf{X})$ and the microscopic magnetic potential field $\phi(\mathbf{X})$, which yield into the definition of the microscopic deformation gradient $\mathbf{F}(\mathbf{X}) = \mathbf{I} + \nabla_0 \mathbf{u}(\mathbf{X})$ and the microscopic magnetic field $\mathbb{H}(\mathbf{X}) = -\nabla_0 \phi(\mathbf{X})$. The macroscopic deformation gradient $\bar{\mathbf{F}}$ and the macroscopic magnetic field $\bar{\mathbb{H}}$ are obtained as the volume average of the microscopic counterparts as

$$\bar{\mathbf{F}} = \frac{1}{|\Omega_0|} \int_{\Omega_0} \mathbf{F}(\mathbf{X}) \, d\Omega_0, \quad \bar{\mathbb{H}} = \frac{1}{|\Omega_0|} \int_{\Omega_0} \mathbb{H}(\mathbf{X}) \, d\Omega_0, \quad (20)$$

respectively.

The microscopic (local) displacement vector $\mathbf{u}(\mathbf{X})$ and magnetic scalar potential $\phi(\mathbf{X})$ are additively decomposed into a linear contribution (macroscopic) and a higher order contribution (mi-

croscopic fluctuation) as

$$\mathbf{u}(\mathbf{X}) = [\bar{\mathbf{F}} - \mathbf{I}] \cdot \mathbf{X} + \tilde{\mathbf{u}}(\mathbf{X}) \quad \text{and} \quad \phi(\mathbf{X}) = -\bar{\mathbb{H}} \cdot \mathbf{X} + \tilde{\phi}(\mathbf{X}), \quad \forall \mathbf{X} \in \Omega_0, \quad (21)$$

with $\tilde{\mathbf{u}}(\mathbf{X})$ and $\tilde{\phi}(\mathbf{X})$ the fluctuation fields. Note that the average of the fluctuation fields over the RVE domain Ω_0 must vanish to satisfy (20). This requirement is automatically fulfilled with the imposition of periodic fluctuation fields on the boundaries of Ω_0 .

Given the constitutive equations of the different phases and the local incompressibility constraint, the admissible sets of solutions spaces $\mathcal{K}$ and $\mathcal{G}$ for $\mathbf{u}$ and ϕ , respectively, are

$$\mathcal{K}(\bar{\mathbf{F}}) = \{ \mathbf{F} = \mathbf{I} + \nabla_0 \mathbf{u}, \det \mathbf{F} = 1, \mathbf{u} = (\bar{\mathbf{F}} - \mathbf{I}) \cdot \mathbf{X} + \tilde{\mathbf{u}}, \tilde{\mathbf{u}} \text{ periodic in } \Omega_0 \} \quad (22)$$

$$\mathcal{G}(\bar{\mathbb{H}}) = \{ \mathbb{H} = -\nabla_0 \phi, \phi = -\bar{\mathbb{H}} \cdot \mathbf{X} + \tilde{\phi}, \tilde{\phi} \text{ periodic in } \Omega_0 \}. \quad (23)$$

Here, the fluctuation fields $\tilde{\mathbf{u}}(\mathbf{X})$ and $\tilde{\phi}(\mathbf{X})$ are the unknowns of the problem.

After imposing the incompressibility condition by a Lagrange multiplier p , the following homogenised energy potential $\bar{\mathcal{W}}$ is posed for the homogenisation problem

$$\bar{\mathcal{W}}(\bar{\mathbf{F}}, \bar{\mathbb{H}}) = \inf_{\mathbf{u} \in \mathcal{K}(\bar{\mathbf{F}})} \sup_{\phi \in \mathcal{G}(\bar{\mathbb{H}})} \sup_p \left[\frac{1}{|\Omega_0|} \int_{\Omega_0} [\Psi(\mathbf{F}, \mathbb{H}, \mathbf{X}) - p[\det \mathbf{F} - 1]] d\Omega_0 \right]. \quad (24)$$

The balance equations of the homogenisation problem are obtained by optimization of the homogenised potential energy $\bar{\mathcal{W}}$ in Eq. (24). In the absence of body forces and under periodic boundary conditions, they result in

$$\begin{aligned} \frac{\partial \bar{\mathcal{W}}}{\partial \tilde{\mathbf{u}}} &= \nabla_0 \cdot \mathbf{P}(\mathbf{F}, \mathbb{H}, p, \mathbf{X}) = 0, \\ \frac{\partial \bar{\mathcal{W}}}{\partial \tilde{\phi}} &= \nabla_0 \cdot \mathbb{B}(\mathbf{F}, \mathbb{H}, \mathbf{X}) = 0, \\ \frac{\partial \bar{\mathcal{W}}}{\partial p} &= \det \mathbf{F} - 1 = 0. \end{aligned} \quad (25)$$

Supplementary Methods 4: Macroscopic boundary conditions in the homogenisation prob-

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The proposed computational framework enables controlling mixed boundary conditions by means of macroscopic mechanical stress/strain and magnetic induction/intensity in the current or reference configurations. This means controlling either the independent macroscopic (average) fields $(\bar{\mathbf{F}}^{\text{app}}, \bar{\mathbb{H}}^{\text{app}})$, and the conjugate macroscopic fields $(\bar{\mathbf{P}}^{\text{app}}, \bar{\mathbb{B}}^{\text{app}})$, or a combination of them by im-

110 posing certain components of the primary fields and certain components of the conjugate fields. It is important to note that the macroscopic magnetic induction to be prescribed as boundary conditions (BCs) of the homogenisation problem contains both the energetic and remanent contributions according to Eq. 19. An alternative approach is to prescribe just the energetic part of the magnetic induction $\mathbb{B}_e$. Results illustrating this last approach are shown in a following section.

115 The average Maxwell (magnetic) stresses are automatically equilibrated by imposing continuity of the magnetic fields in neighbouring RVEs. Thus, only the average Maxwell stress contribution needs to be neglected at the macroscopic boundary conditions.

When one controls the conjugate fields $\bar{\mathbf{P}}$ and $\bar{\mathbb{B}}$, the average potential energy for the RVE is defined as

$$\mathcal{P}(\bar{\mathbf{F}}, \bar{\mathbb{H}}) = \bar{\mathcal{W}}(\bar{\mathbf{F}}, \bar{\mathbb{H}}) - \bar{\mathbf{P}}^{\text{app}} \cdot [\bar{\mathbf{F}} - \mathbf{I}] - \bar{\mathbb{H}} \cdot \bar{\mathbb{B}}^{\text{app}}. \quad (26)$$

120 In Eq. (26) the first term stands for the internal energy and the two last terms, for the work of the external “force” loads. These last terms vanish if the primary fields $\bar{\mathbf{F}}$ and $\bar{\mathbb{H}}$ are prescribed instead. Note that $\bar{\mathbf{P}}^{\text{app}}$ refers to the stress including both mechanical and magnetisation average stresses applied (not the Maxwell component). If a combination of components of the primary and conjugate average fields is imposed, the potential energy $\mathcal{P}$ needs to be amended accordingly just

125 for such components. To prescribe such loading conditions on the conjugate macroscopic fields, it is necessary to additionally fulfil

$$\begin{aligned} \frac{\partial \mathcal{P}}{\partial \bar{\mathbf{F}}} &= \int_{\Omega_0} \mathbf{P}(\mathbf{F}, p, \mathbf{X}) \, d\Omega_0 - \bar{\mathbf{P}}^{\text{app}} = 0 \\ \frac{\partial \mathcal{P}}{\partial \bar{\mathbb{H}}} &= \int_{\Omega_0} \mathbb{B}(\mathbf{F}, \mathbb{H}, \mathbf{X}) \, d\Omega_0 - \bar{\mathbb{B}}^{\text{app}} = 0, \end{aligned} \quad (27)$$

which are derived from (26).

To solve the non-linear equilibrium state, Eqs. (25), (27) are formulated in their integral weak form to be solved in the discretised Ω_0 domain. A Newton-Raphson scheme implemented in the open source code FEniCS is used to solve the non-linear problem under the prescribed boundary conditions. To enhance the convergence, the loading conditions are progressively applied via a time discretisation.

The main macroscopic boundary conditions imposed in the homogenisation framework for the confined deformation are

$$\bar{\mathbf{b}}(t) = \begin{Bmatrix} 0 \\ 0 \\ b_{\text{end}}t/t_{\text{ramp}} \end{Bmatrix}, \quad \bar{\mathbb{H}}_r(t) = \begin{Bmatrix} 0 \\ H_r \\ 0 \end{Bmatrix}, \quad \bar{\mathbf{F}}(t) = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}. \quad (28)$$

Note that the prescription of the Eulerian magnetic induction instead of its Lagrangian counterpart is just a mere modelling decision to better link to the real deformed state of the MRE [6].

Supplementary Methods 5: Treating the air domain in the virtual testing framework - the auxiliary weight function

The air domain within the virtual testing framework is modelled as a rectangular box ten times larger than the beam. This enables the application of a homogeneous far-field imposing the magnetic potential on the external faces of this domain. The air is modelled as a Neo-Hookean compressible media with a negligible stiffness, i.e., shear modulus of $G = 80$ Pa. To model its compressible nature, the degrees of freedom due to the Lagrange multiplier p in the air domain are removed and, instead, a volumetric contribution is added to the mechanical contribution to the energy potential as

$$\Psi_{\text{mech}}(\mathbf{F}) = \Psi_{\text{mech,iso}} + \Psi_{\text{vol}} = G\mathbf{F} - G\mathbf{F}^{-\text{T}} + \lambda \ln(J)\mathbf{F}^{-\text{T}}, \quad (29)$$

with $\lambda = \kappa - \frac{2G}{3}$ as a function of the bulk modulus κ .

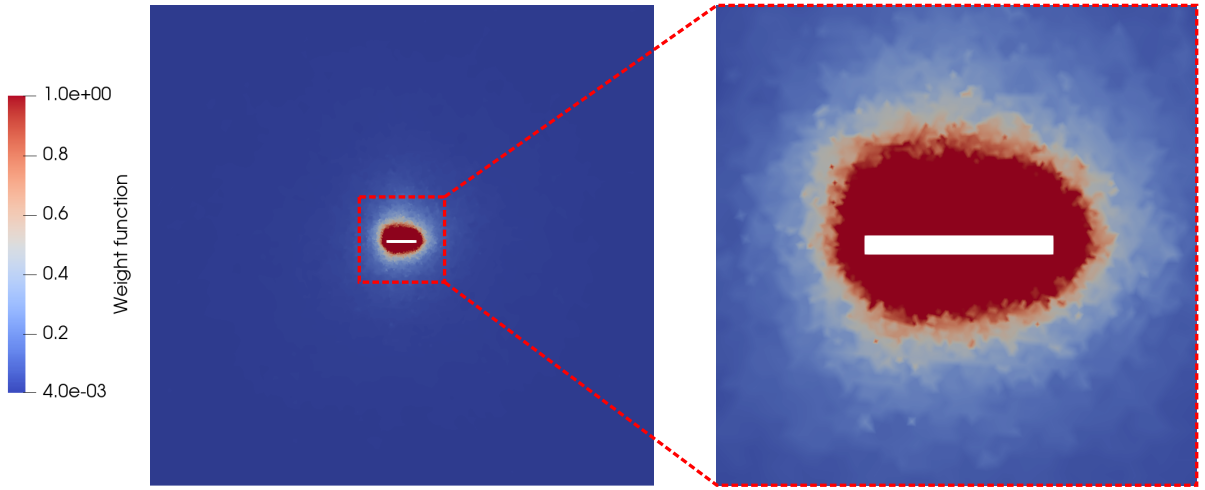
To minimise the additional stiffness posed by the air domain and to enhance its behaviour, we

implement the auxiliary energy function approach reported by Rambašek et al. [4]. The weight function gauges the mechanical contribution to the mechanical Piola stress tensor in the weak form.

150 It takes values close to one in the smaller elements surrounding the beam, and smaller values for the larger elements further from the beam. The function reads as

$$w(\mathbf{X}) = \max \left[w_{min}, \min \left(\frac{V_{ref}}{V_0}, w_{max} \right) \right], \quad (30)$$

with $w_{min} = 1 \times 10^{-5}$, $w_{max} = 1$ and $V_{ref} = 2 \times 10^{-2}$. Supplementary Figure 2 shows the values of the weight function in the air domain.

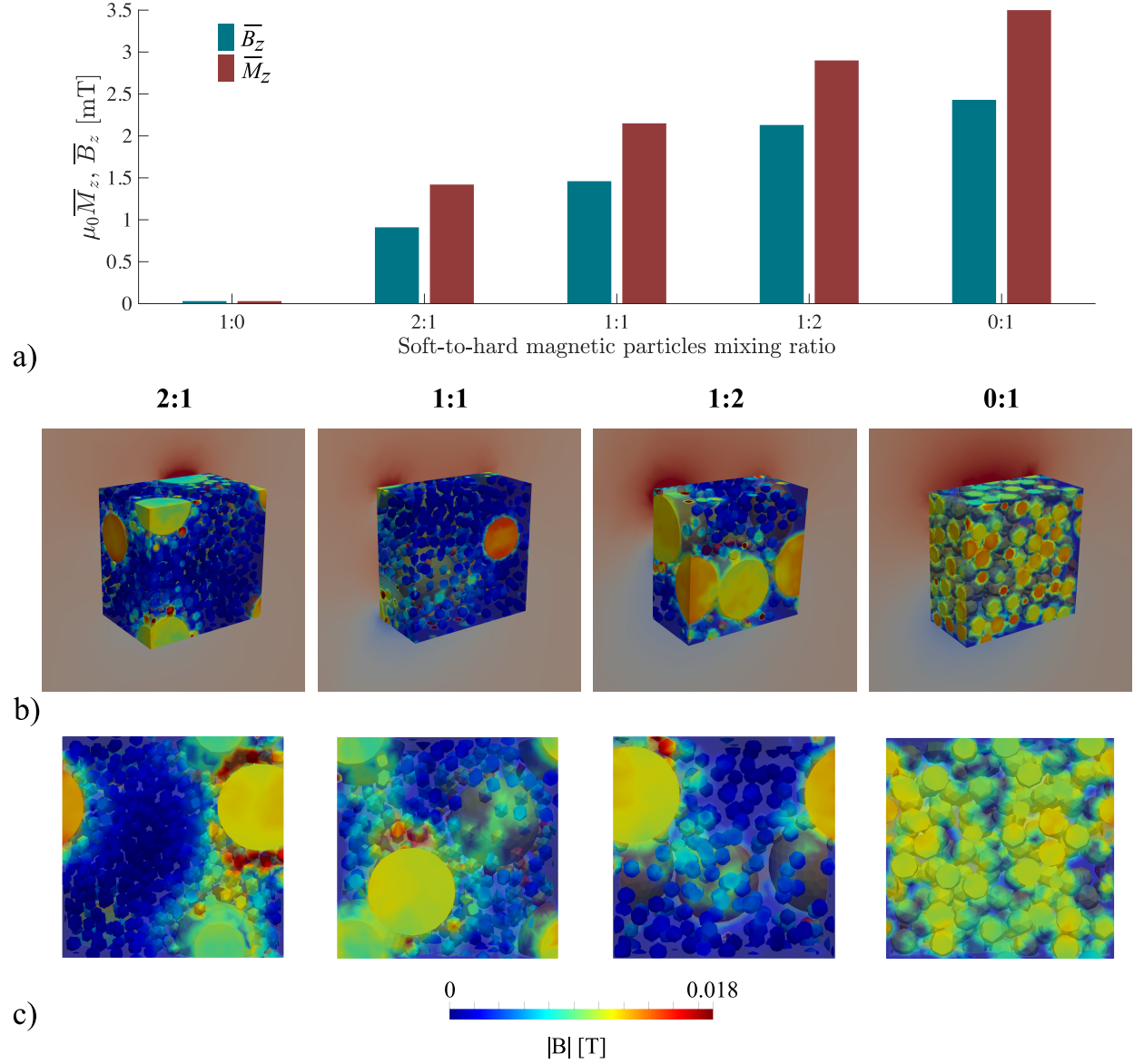


Supplementary Figure 2. **Distribution of the weight function in the air domain and a detail of the air domain in the proximity of the beam.** The weight function takes values close to one in the smaller elements surrounding the beam, and smaller values for the larger elements further from the beam.

Supplementary Methods 6: Experimental calibration of the remanent field in the hard- magnetic particles

The remanent magnetic field within hard-magnetic particles is prescribed as a constant value. To determine it, an inverse engineering approach is followed. After experimentally pre-magnetising a hMRE sample under 1 mT, we measured the magnetic induction on its surface. Then, the remanent field in the particles is adjusted so that the magnetisation of the homogenisation model fits the
160 macroscopic experimental measurement. To this end, we take a representative sample of hMRE

with 30 vol.%, according to the experiment. The sample is surrounded by air and uniquely subjected to the residual magnetisation of the particles via $\mathbb{H}_r$, see Supplementary Figure 3. Therefore, in this simulations, no external magnetisation is applied, no periodic boundary conditions are set and no macroscopic fields are prescribed. In addition, this methodology allows for estimating the
165 macroscopic remanent magnetisation of hybrid MREs, see results in Supplementary Figure 3.

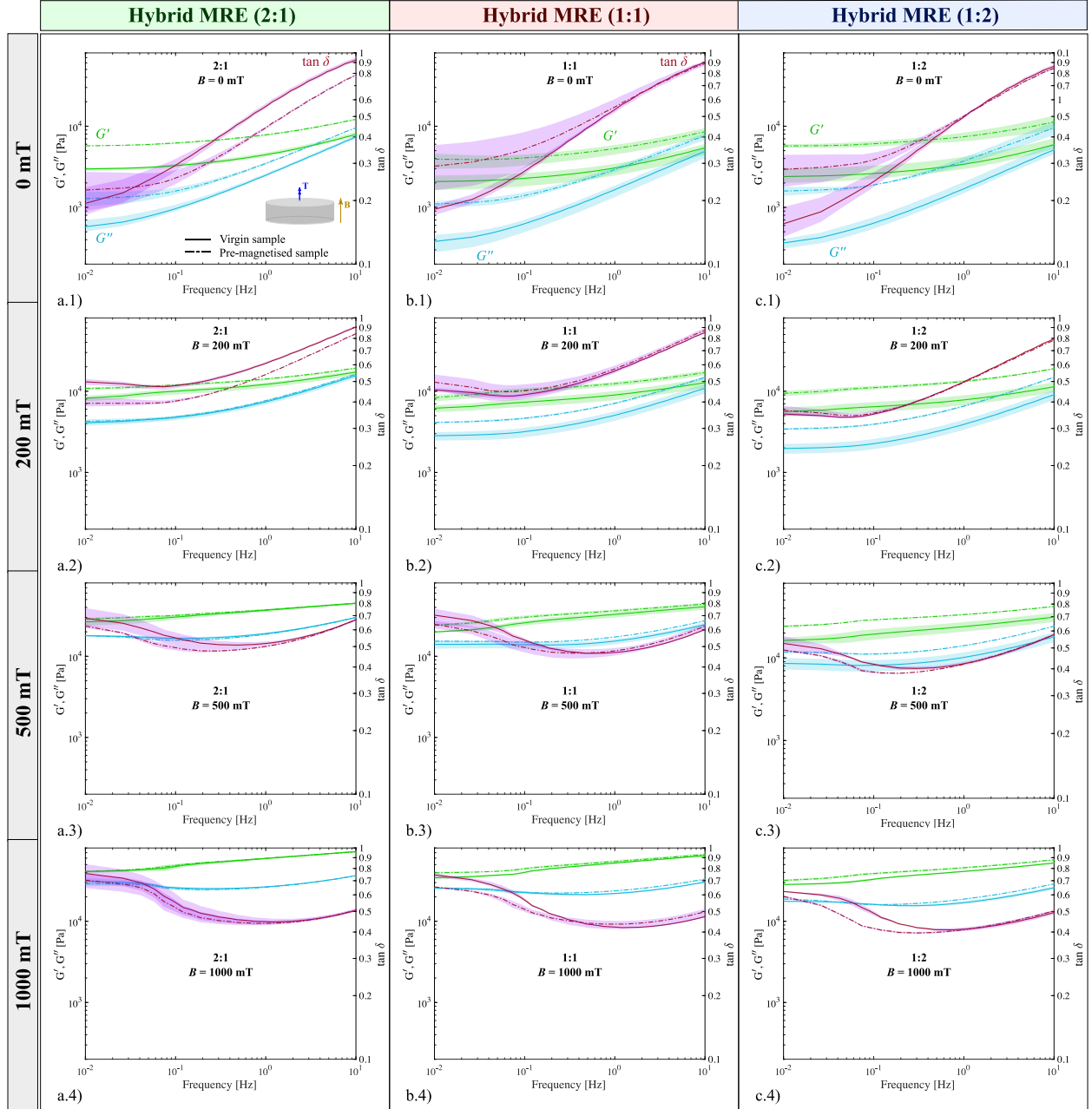


Supplementary Figure 3. **Calibration procedure of the remanent magnetic field.** In the absence of external magnetic actuation on a pre-magnetised MREs, the homogeneous magnetic induction within the pre-magnetised MREs is different than zero. (a) Homogeneous macroscopic magnetic induction and magnetisation within the RVE in the direction of the pre-magnetisation of the hard-magnetic particles and depending on the soft-to-hard particles mixing ratio. (b) Magnetic field magnitude in MRE samples for different mixing ratios. The sample is surrounded by air to model the experimental measuring of the remanent macroscopic field with the teslameter and perform the calibration. (c) Clips of the cubes to visualise the magnetic induction magnitude inside the cube. Note that the opacity of the matrix has been reduced to visualise the particles in rear planes.

Supplementary Discussion 1: Additional results for the magneto-mechanical frequency sweep tests

This section presents the complete set of results for the frequency sweep tests on hybrid MREs. Each column contains the results for each soft-to-hard particles mixing ratio. In addition, each

170 sample condition is tested under different magnetic actuations.



Supplementary Figure 4. **Dynamic mechanical analysis (DMA) of hybrid MREs.** Curves for the storage modulus, loss modulus and loss factor from shear frequency sweeps for soft-to-hard-magnetic particles volume mixing ratios of (a) 2:1, (b) 1:1 and (c) 1:2. The total amount of magnetic particles is 30 vol.%. The experiments are performed on both virgin and pre-magnetised samples under magnetic actuation of $B = \{0, 200, 500, 1000\}$ mT.

Supplementary Discussion 2: Influence of the magnetic particles' relative size ratio on the homogeneous behaviour

This section provides additional results to account for the influence of the relative size between soft- and hard-magnetic particles on the homogeneous behaviour of hybrid MREs. While the soft-
175 to-hard-magnetic particles' mixing ratio is fixed at a ratio of 1:1, the size ratio (soft:hard) is set to 1:6, 1:3, 1:1, 3:1 and 6:1.



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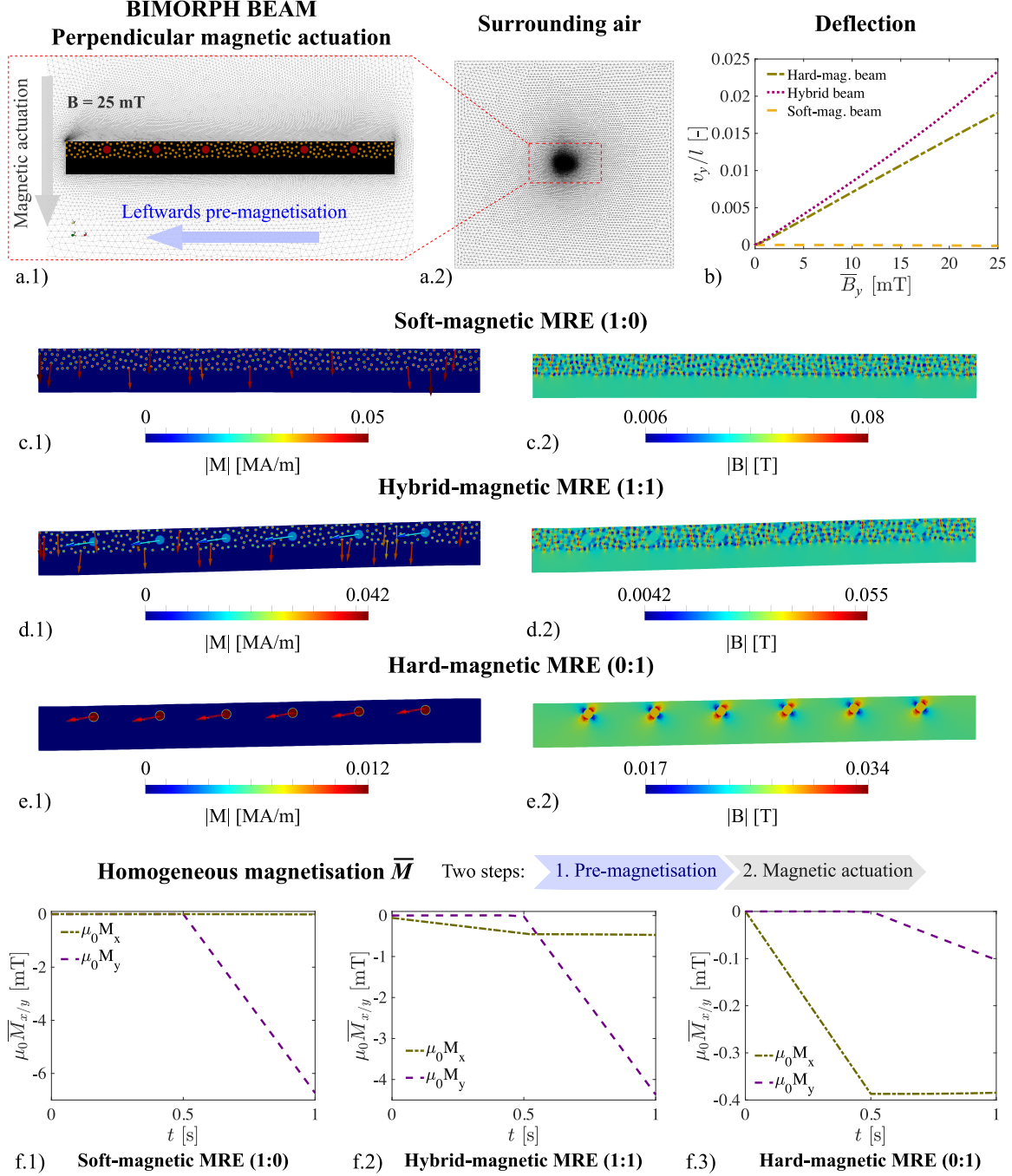
Supplementary Discussion 3: Additional results for the cantilever actuator under pre-magnetisation and magnetic actuation conditions

Additional results for the bimorph and homogeneous cantilever beams are presented in this section

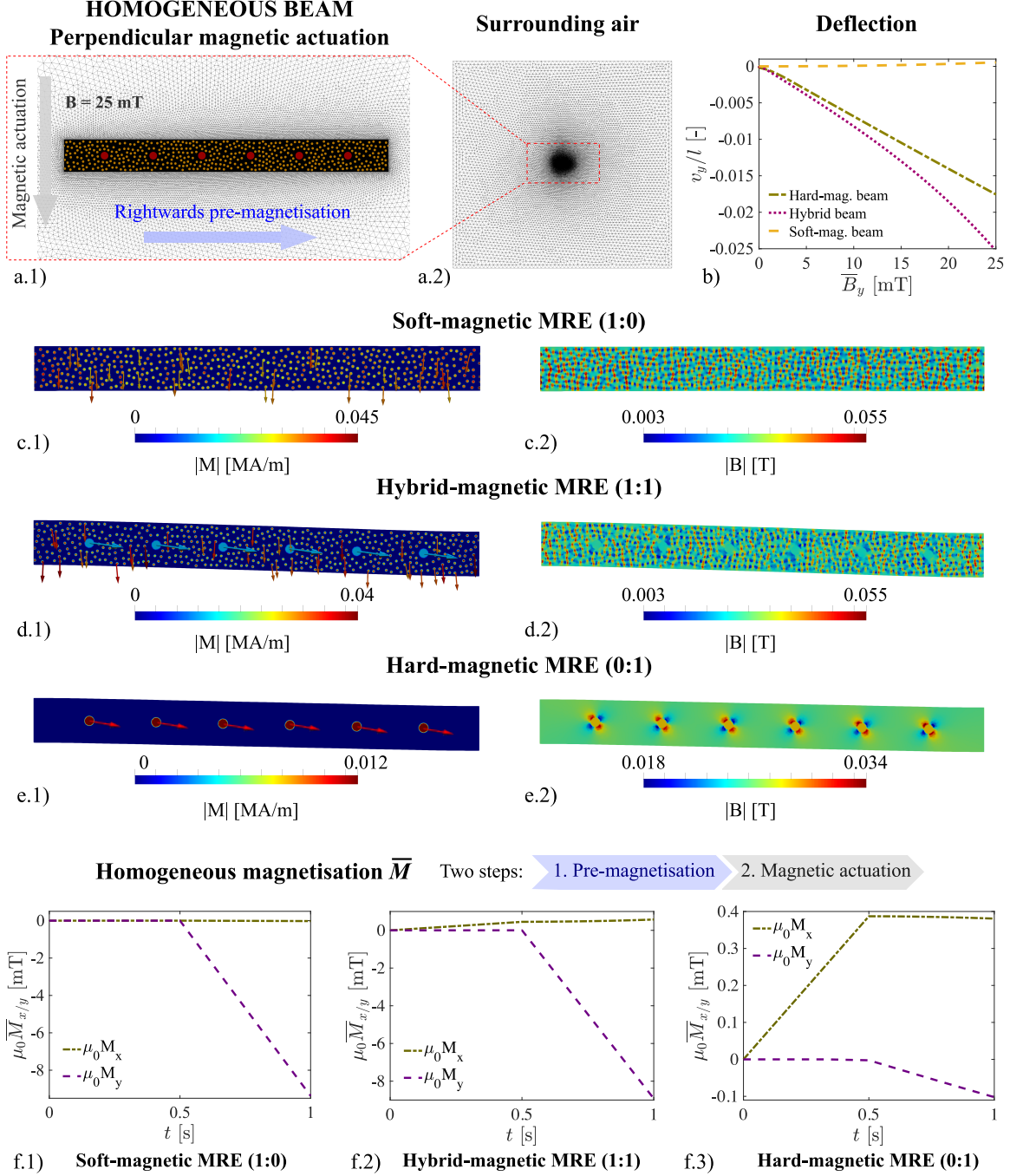
180 under different actuation conditions (see Table 3 for a summary of all the conditions).

Supplementary Table 3. Set of boundary conditions (magnetic actuation and pre-magnetisation) used for the cantilever actuation within the virtual testing framework. Note that (MT) means that the results are presented in the Main Text, and (SI), in Supplemental Information.

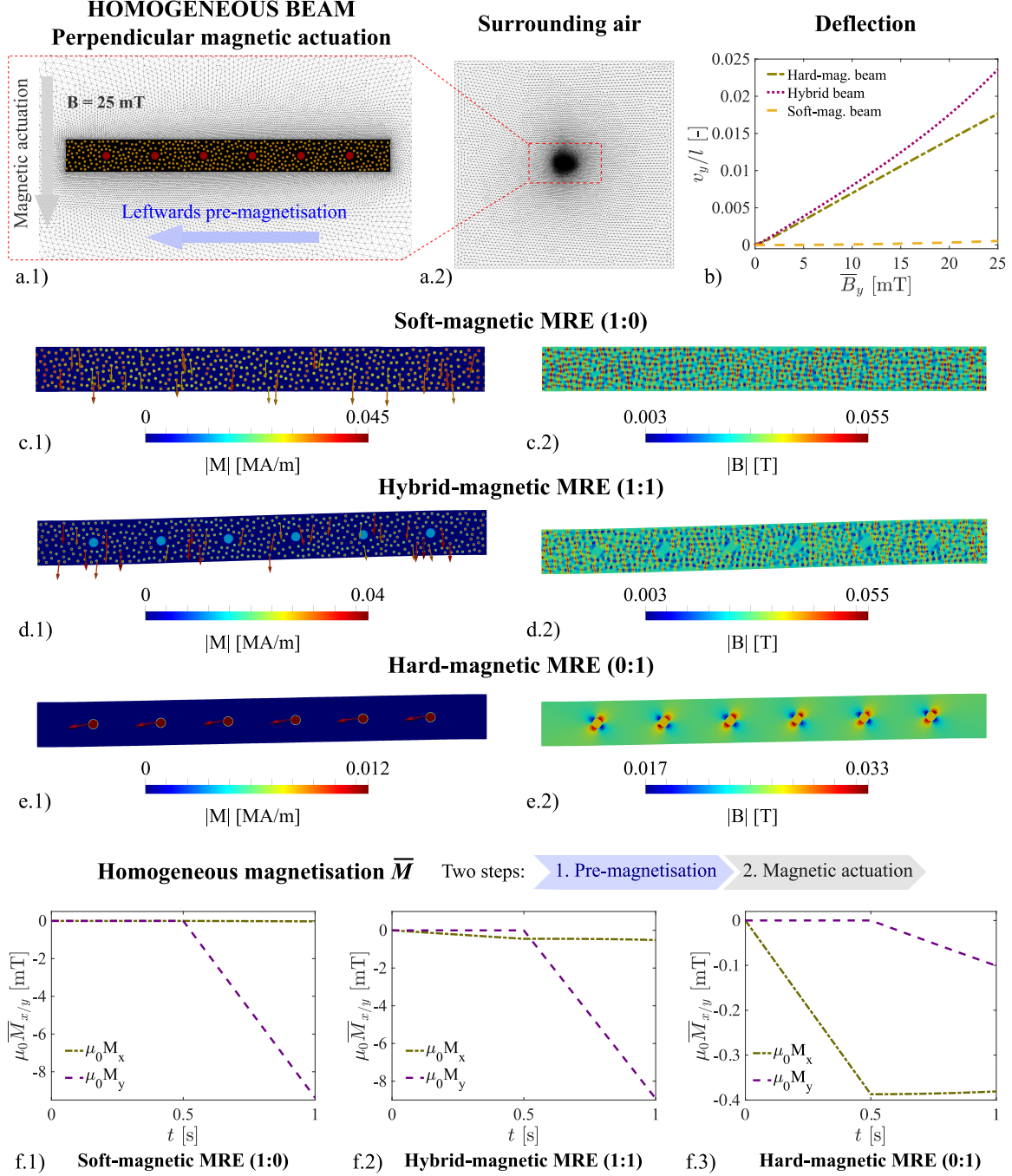
Type of beam	Magnetic actuation	Pre-magnetisation
Bimorph	Perpendicular	Rightwards (MT) / Leftwards (SI)
	Parallel	Rightwards (MT)
Homogeneous	Perpendicular	Rightwards (SI)/ Leftwards (SI)
	Parallel	Rightwards (MT)



Supplementary Figure 6. **Numerical application of the microscopic model on a bimorph cantilever beam with leftwards pre-magnetisation.** (a.1) Magnetic actuation conditions on a bi-layered cantilever beam with hybrid magnetic fillers (soft- and hard-magnetic particles) in the top layer and pure elastomeric matrix in the bottom layer. Hard-magnetic particles are leftwards pre-magnetised with a remanent field of $\mathbb{H}_r = 0.015$ mT. (a.2) Complete FE problem meshed including the air surrounding the beam to guarantee the proper application of the magnetic actuating field. (b) Normalised beam deflection against the external applied magnetic field depending on the filler content used in the upper layer: sMRE, 1:1 hybrid MRE and hMRE. The hMRE beam and the hybrid beam bend under external magnetic actuation as a consequence of the torque transmission from the hard-magnetic particles. The z-deflection is normalised by the length of the beam. (c.1) and (c.2) Magnetisation and magnetic induction fields within the sMRE beam after magnetic actuation (25 mT), respectively. (d.1) and (d.2) Magnetisation and magnetic induction fields within the hybrid MRE beam after magnetic actuation (25 mT), respectively. (e.1) and (e.2) Magnetisation and magnetic induction fields within the hMRE beam after magnetic actuation (25 mT), respectively. (f.1-3) Evolution of the homogenised horizontal and vertical magnetisation components of the sMRE, hybrid MRE and hMRE beams, respectively. In this simulations, a pre-magnetisation step is applied first, followed by the magnetic actuation.



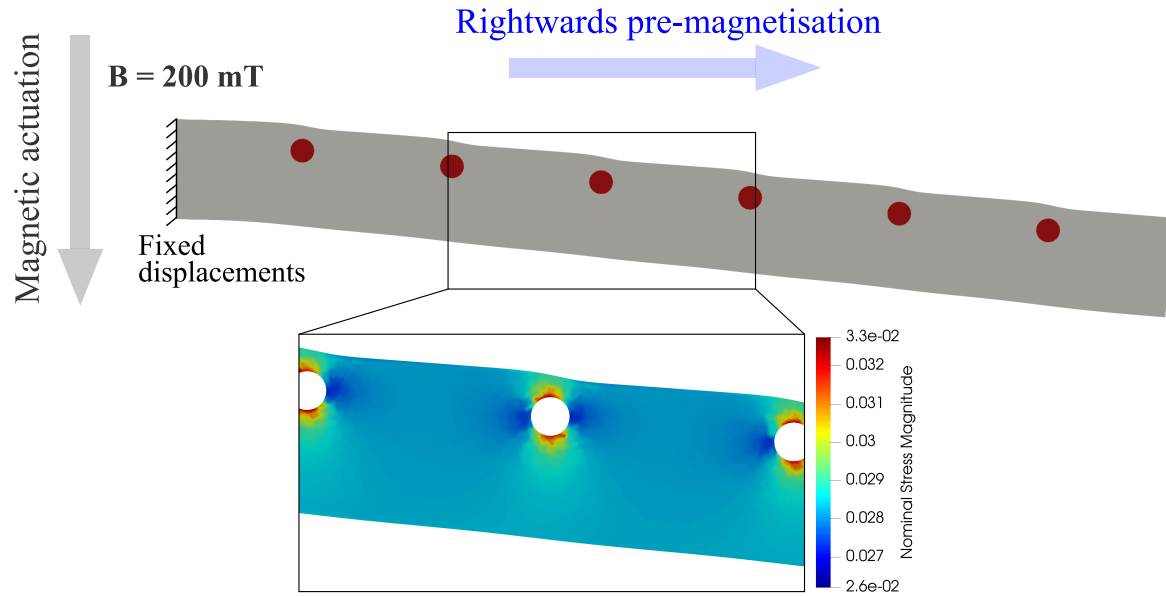
Supplementary Figure 7. **Numerical application of the microscopic model on a homogeneous cantilever beam with rightwards pre-magnetisation.** (a.1) Magnetic actuation conditions on a homogeneous cantilever beam with hybrid magnetic fillers (soft- and hard-magnetic particles). Hard-magnetic particles are rightwards pre-magnetised with a remanent field of $H_r = -0.015 \text{ mT}$. (a.2) Complete FE problem meshed including the air surrounding the beam to guarantee the proper application of the magnetic actuating field. (b) Normalised beam deflection against the external applied magnetic field depending on the filler content used in the upper layer: sMRE, 1:1 hybrid MRE and hMRE. The hMRE beam and the hybrid beam bend under external magnetic actuation as a consequence of the torque transmission from the hard-magnetic particles. The z-deflection is normalised by the length of the beam. (c.1) and (c.2) Magnetisation and magnetic induction fields within the sMRE beam after magnetic actuation (25 mT), respectively. (d.1) and (d.2) Magnetisation and magnetic induction fields within the hybrid MRE beam after magnetic actuation (25 mT), respectively. (e.1) and (e.2) Magnetisation and magnetic induction fields within the hMRE beam after magnetic actuation (25 mT), respectively. (f.1-3) Evolution of the homogenised horizontal and vertical magnetisation components of the sMRE, hybrid MRE and hMRE beams, respectively. In this simulations, a pre-magnetisation step is applied first, followed by the magnetic actuation.



Supplementary Figure 8. **Numerical application of the microscopic model on a homogeneous cantilever beam with leftwards pre-magnetisation.** (a.1) Magnetic actuation conditions on a homogeneous cantilever beam with hybrid magnetic fillers (soft- and hard-magnetic particles). Hard-magnetic particles are leftwards pre-magnetised with a remanent field of $H_r = 0.015 \text{ mT}$. (a.2) Complete FE problem meshed including the air surrounding the beam to guarantee the proper application of the magnetic actuating field. (b) Normalised beam deflection against the external applied magnetic field depending on the filler content used in the upper layer: sMRE, 1:1 hybrid MRE and hMRE. The hMRE beam and the hybrid beam bend under external magnetic actuation as a consequence of the torque transmission from the hard-magnetic particles. The z-deflection is normalised by the length of the beam. (c.1) and (c.2) Magnetisation and magnetic induction fields within the sMRE beam after magnetic actuation (25 mT), respectively. (d.1) and (d.2) Magnetisation and magnetic induction fields within the hybrid MRE beam after magnetic actuation (25 mT), respectively. (e.1) and (e.2) Magnetisation and magnetic induction fields within the hMRE beam after magnetic actuation (25 mT), respectively. (f.1-3) Evolution of the homogenised horizontal and vertical magnetisation components of the sMRE, hybrid MRE and hMRE beams, respectively. In this simulations, a pre-magnetisation step is applied first, followed by the magnetic actuation.

Additional results for the hard-magnetic cantilever actuator under larger magnetic actuation

Supplementary Figure 9 presents additional results for the pure hard-magnetic bimorph cantilever beam under a larger magnetic actuation to extend the study to a larger deformation range. A perpendicular magnetic field of 200 mT allows to see high local torsional deformations of the matrix in the particles' vicinity. This owes to the extremely soft nature of the matrix and explains the a non perfect parabolic macroscopic structural bending.



Supplementary Figure 9. **Numerical detail of local stress concentration in the bimorph beam.** Applying a larger perpendicular magnetic field of 200 mT on the hard-magnetic bimorph cantilever beam allows to see high local torsional deformations of the matrix in the particles' vicinity. The particles are pre-magnetised in the horizontal direction and try to turn to align their magnetisation with the external field. A detail shows the stress state of the matrix near the particles.

Supplementary References

- [1] Moreno, M. A., Gonzalez-Rico, J., Lopez-Donaire, M. L., Arias, A. & Garcia-Gonzalez, D. New experimental insights into magneto-mechanical rate dependences of magnetorheological elastomers. *Composites Part B: Engineering* **224**, 109148 (2021).
- [2] Moreno-Mateos, M. A., Lopez-Donaire, M. L., Hossain, M. & Garcia-Gonzalez, D. Effects of soft and hard magnetic particles on the mechanical performance of ultra-soft magnetorheological elastomers. *Smart Materials and Structures* **31**, 065018 (2022).
- [3] Lucarini, S., Hossain, M. & Garcia-Gonzalez, D. Recent advances in hard-magnetic soft composites: Synthesis, characterisation, computational modelling, and applications. *Composite Structures* **279**, 114800 (2022).
- [4] Rambašek, M., Mukherjee, D. & Danas, K. A computational framework for magnetically hard and soft viscoelastic magnetorheological elastomers. *Computer Methods in Applied Mechanics and Engineering* **391**, 114500 (2022).
- [5] Mukherjee, D., Rambašek, M. & Danas, K. An explicit dissipative model for isotropic hard magnetorheological elastomers. *Journal of the Mechanics and Physics of Solids* **151**, 104361 (2021).
- [6] Mukherjee, D., Bodelot, L. & Danas, K. Microstructurally-guided explicit continuum models for isotropic magnetorheological elastomers with iron particles. *International Journal of Non-Linear Mechanics* **120**, 103380 (2020).