Matching Magnetic Heating and Thermal Actuation for Sequential Coupling in Hybrid Composites by Design

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Sequentially coupling two material functions requires matching the output from the first with the input of the second function. Here, magnetic heating controls thermal actuation of a hybrid composite in a challenging system environment causing an elevated level of heat loss. The concept is a hierarchical design consisting of an inner actuator of nanocomposite material, which can be remotely heated by exposure to an alternating magnetic field (AMF) and outer layers of a porous composite system with a closed pore morphology. These porous layers act as heat insulators and as barriers to the surrounding water. By exposure to the AMF, a local bulk temperature of 71 °C enables the magnetic actuation of the device, while the temperature of the surrounding water is kept below 50 °C. Interestingly, the heat loss during magnetic heating leads to an increase of the water phase (small volume) temperature. The temperature increase is able to sequentially trigger an adjacent thermal actuator attached to the actuator composite. In this way it could be demonstrated how the AMF is able to initiate two kinds of independent actuations, which might be interesting for robotics operating in aqueous environments.

Multifunctional materials are designed to perform multiple tasks through judicious integration and logical connection of functions. These can be implemented on the same length scale or hierarchically organized. The performance of a function in a material system can be adjusted either by structural variations (structure–function relationship) or by changing parameters of fabrication or programming (process–function relationship). A unique feature of multifunctionality is the targeted determination of function–function relationships. Orthogonal functions can be addressed almost independently from each other, while, sequentially coupled functions enable linking functions as the output signals of one function acts as input for the activation of a second function. The central challenge is to couple the individual functions representing the flow of information allowing the temporal and quantitative control of the targeted performance.

Several multimaterial approaches combining soft and flexible polymers with functional fillers have been explored for creating multifunctionality with orthogonal or sequentially coupled functions. An example for orthogonal functions is a drug loaded polymer implant with shape-memory capability and degradation functionality. Such implants can be inserted into the body in a minimally invasive procedure. The shape-memory effect is induced by body temperature, once the implant is in contact with the physiological environment. The drug is released.

Finally, the polymer matrix degrades. Here, the functions shape-memory, drug release and degradability operate independent from each other as they are time wise separated. The incorporating of small amounts of active fillers such as iron oxide nanoparticles (ioNPs), graphene, nanowires, or carbon nanotubes into polymer matrices with stimuli responsive or actuation functionality have enabled sequentially linked functions. The fillers provide energy conversion functionality such as magnetic, photo or electric heating. For instance, the incorporation of magnetic nanoparticles (MNPs) in a semicrystalline polymer with oriented polymer chains enabled the coupling of magnetic heating with a thermally controlled function. The heat generated in the composite materials by exposure to an alternating magnetic field (AMF), resulted in the melting of a fraction of crystals in the polymer and initiated the melting-induced contraction. When the magnetic field is switched off, the oriented crystallization of the crystalline lamella leads to an expansion and in this way reverses the shape change. Recently, a multifunctional polymer implant systems for a local on-demand protein release with sequentially coupled functions was reported. Here, the laser light was used to locally heat the surface of heat shrinkable microcapsules with NIR dye, which sequentially triggered the on-demand protein release. Similarly, the incorporation of gold nanoparticles in a thermo-sensitive polymer enabled a photo initiated actuation of the polymer. Overall, the non-contact actuation of the polymers offer great potential for applications in drug delivery, soft robots, and biomedical

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Multifunctional materials hold great promise for their role in e-skins, wearable and implantable devices, soft actuators, and sensors with the potential for self-power and self-healing. Many of these applications require device operations in aqueous environments, which causes a great challenge. The high heat capacity of water compared to air results in a massive loss in heat energy. For this reason, remotely activated stimuli-sensitive hydrogels require significant energy inputs. The heat loss via the surfaces has also been identified as a hurdle for magnetically induced shape-memory composites. We hypothesized that a hierarchical design would enable the efficient coupling of magnetic heating and thermally controlled actuation.

Our concept is the introduction of a heat insulating component between the actuator and the aqueous environment (Figure 1). Our strategy to achieve such a hierarchical design involves a layered composite device, with the core of a bulk hybrid composite with crystallizable actuating units and the outer layers of porous architecture having a closed pore morphology. We also equipped the device with a thermally controlled actuator unit having a lower actuation temperature, to explore, whether a slight increase in temperature of the water due to the heat loss can remotely trigger this second actuator unit. A schematic demonstration of the sequentially linked magnetic and the additional thermal actuation functionalities of the fabricated device along with the molecular mechanism is shown in Figure 2.

Oligo(ω-pentadecalactone) (OPDL) based hybrid nanocomposites (Hy-NC) with covalently anchored ioNPs were selected for the design of the core of the device. These Hy-NC with the crystallizable domain of OPDL acting as actuating unit, enables a high transition temperature and fast crystallization and could be a suitable selection for magnetically triggered actuators. The synthesis of Hy-NC was carried out according to the methods reported in refs. [32,33]. Compared to the magnetic actuation in dry state, relatively higher amount of ioNPs (8 wt%) was required to enable the actuation of the Hy-NC in aqueous environment. Hy-NC has a glass transition temperature \( T_g \) at \(-70 \degree C\) as well as a melting temperature \( T_m \) of 74 \degree C to which a crystallization temperature \( T_c \) of 58 \degree C corresponds as determined by DSC. The same Hy-NC was used to fabricate the porous outer layers with closed pores by application of a template technique using polyethylene based microspheres (PE, \( d = 50 \pm 5 \mu m \)). The magnetically triggered actuation capability of these Hy-NC is based on directed crystallization of a fraction of crystals acting as actuating unit, while the remaining crystals with higher \( T_m \) act as geometry determining unit.[16,34] The thermal actuator with the lower actuation temperature was based on a multiphase polymer network of oligo(e-caprolactone) (OCL) and OPDL. Here, OCL crystalline domains \( T_m, OCL = 50 \degree C \) acted as actuating units while the OPDL \( T_m, OPDL = 82 \degree C \) crystalline domains acted as geometry determining units. The synthesis and actuation capability of such polymer network is reported in ref. [35]. The thermal analysis of the Hy-NC and the thermal actuator is shown in Figure S1, Supporting Information. The multilayer actuator is activated by exposure to AMF.

The hierarchical architecture of the fabricated device was investigated by using optical and scanning electron microscopy (SEM). The optical microscopic image of the cross section of the device \((40 \times 10 \times 5 \, mm^3)\) is shown in Figure 1b. The two outer layers of the porous composite (each thickness \( = 1.5 \, mm \)) and the inner layer of the bulk composite (2 mm) are visible in the microscopic image. The SEM image shown in Figure 1d confirmed the porous morphology of the outer layers with spherical pores of diameter \( d = 50 \pm 3 \, \mu m \), which is corresponding to the size of the polyethylene (PE) microspheres used as templates. As the PE microspheres were randomly distributed in the prepolymer mixture, the leaching of the PE spheres enabled the porous architecture with closed cell morphology which was confirmed by 3D microstructures obtained by X-ray computed microtomography (µ-CT). However, to avoid the percentage of interconnected pores in the composite, relatively lower weight ratio (10 wt%) of the PE spheres in the reaction mixture was used. The µ-CT measurements resulted in an overall porosity of 30% along with a mean void size of 50.1 ± 11.4 nm. Nevertheless, highly dense porous layers with the wall thickness of the spherical pores ranging from 100–200 \( \mu m \) were achieved. The pore size distribution obtained by µ-CT measurements is shown in...
in Figure 1c while, a 3D image of the porous layer is provided in Figure S2, Supporting Information. By further increasing the amount of PE spheres to 20 wt% in the reaction mixtures, the porosity was increased to 70%, however, most of the interconnected pores were achieved (Figure S3, Supporting Information).

As a precise heating and partial melting of the OPDL crystalline domain by exposure to an AMF is necessary for magnetic actuation, therefore, the bulk temperature \( T_{\text{bulk}} \) during magnetic heating of the composite device by application of different magnetic field strengths in water was determined. The exposure to an AMF of \( H = 25 \text{ kA m}^{-1} \) for 5 min enabled a maximum temperature of 71 °C, which is below the \( T_m \) of the OPDL crystalline domain and is sufficient to provide the maximum actuation capability.\(^{[16]}\) Switching the magnetic field strength to \( H = 0 \text{ kA m}^{-1} \) resulted in an instantaneous cooling of the composite to 37 °C, which is well below the crystallization temperature of the Hy-NC \( (T_c = 58 \text{ °C}) \). During magnetic heating, the heat loss to the surrounding water resulted in an indirect heating of the water and a maximum temperature of \( T_{\text{water}} = 48 \text{ °C} \) was observed. The significantly lower temperature of the water compared to the \( T_{\text{bulk}} \) of the composite was attributed to the heat insulating and water barrier characteristic of the closed cell morphology of the porous layers. The heating profiles of the Hy-NC and water for three heating and cooling cycles is shown in Figure 3a. The magnetically induced cyclic actuation of the device (without thermal actuator) in the stretching mode enabled a reversible change in the length with \( \varepsilon_{\text{rev}} = 4.5 \pm 0.2\% \). The magneto-mechanical experiments were carried out according to the method reported in ref. \(^{[16]}\). The resulting actuation cycles are provided in Figure S4, Supporting Information. To observe the sequential coupling of magnetic and thermal actuation of the fabricated device, both components, the Hy-NC and the embedded thermal actuator were heated to 90 °C, where the crystalline domains of both components are in molten state.
After heating, the device was programmed by bending both components of the device and cooling to ambient temperature as shown in Figure 3b. The programming step was followed by actuation experiments, which were conducted in the aqueous environment in the center of the magnetic coil. Exposure of the device to \( H = 25 \text{ kA m}^{-1} \) for 5 min resulted in a partial opening of the bend shaped Hy-NC and was attributed to the partial melting of the OPDL crystalline domain. When \( T_{\text{bulk}} \) of the composite reached 71 °C, the heat loss to the surrounding water enabled a rise in the temperature of the water to 48 °C in 5 min. The sequential heating of water initiated the actuation of the thermal actuator fabricated without addition of ioNPs. Here, a combination of the magnetic and thermal actuation functionalities was observed. By turning off the magnetic field strength to \( H = 0 \text{ kA m}^{-1} \), the composite device quickly returned back to the programmed bended shape within a short time span of 1 min. Nevertheless, as the temperature of the surrounding water was still high, the thermal actuator maintained the already open shape. To observe the recovery of the thermal actuator, the water was cooled to 15 °C (\( T_{\text{C,PCL}} = 22 \) °C) by application of cooled air provided from the lower side of the water beaker. The sequentially linked magnetic and thermal actuation cycles of the Hy-NC were repeated three times and the resulting changes in the bending angles of both components were recorded. The stepwise actuation of magnetic and thermal actuation of the fabricated multifunctional device is also shown in the movie (Movie S1, Supporting Information). The changes in the bending angles of the Hy-NC and the thermal actuator are provided in Figure 3c. The magnetic actuation of the Hy-NC resulted in a reversible change of the bending angle \( \Delta \theta = 28 \pm 1^\circ \) by application of \( H = 25 \text{ kA m}^{-1} \) and \( H = 0 \text{ kA m}^{-1} \). By further increasing the magnetic field strength to \( H = 27 \text{ kA m}^{-1} \), a complete opening of the Hy-NC was achieved and the actuation capability was lost because of the complete melting of the OPDL crystalline domain. In contrast, for the thermal actuator a \( \Delta \theta = 42 \pm 2^\circ \) was achieved by indirect heat transfer to the water, enabling a temperature of 48 °C and by cooling to 15 °C with cooled air. The IR camera images of the Hy-NC device and the surrounding water are shown in Figure S5, Supporting Information. The thermal actuators are not visible here as their temperature does not differ to the surrounding water.

To investigate the heat flow across different layers of the device, temperature-dependent thermal diffusivity (\( \alpha \)) and thermal conductivity (\( \lambda \)) measurements of the individual layers and the multilayer device were carried out. Figure 4a shows the thermal diffusivity measurements of the bulk Hy-NC core, porous layer, and the multilayer device by using the laser flash method in the temperature range of 25–100 °C. For all samples, a slight decrease in the value of \( \alpha \) by increasing the temperature was observed. However, the porous composite films showed the maximum \( \alpha \) compared to the bulk Hy-NC and the device. For example at 25 °C, \( \alpha = 0.10 \text{ mm}^2 \text{ s}^{-1} \) for bulk Hy-NC was increased to the value of 0.19 mm² s⁻¹ for the porous layers. The high value of the thermal diffusivity for porous composite layers is attributed to the low density of the porous system compared to the bulk composites.\(^{[36]}\) However, for the multilayer device containing two porous layers and a bulk composite layer, a slightly lower values of \( \alpha = 0.14 \text{ mm}^2 \text{ s}^{-1} \) at 25 °C was achieved. Similar pattern of the thermal diffusivity values for three samples in the whole temperature range was observed. In contrast to the thermal diffusivity measurements, the values of thermal conductivity (\( \lambda \)) are directly proportional to the density of the composite and are related to the thermal diffusivity by Equation (1).

\[
\lambda(T) = \alpha(T) \cdot \rho(T) \cdot C_p(T)
\]  

**Figure 3.** a) Bulk temperature profile of the Hy-NC during magnetic actuation (red), temperature profile of the surrounding water (black). b) Images showing the sequentially linked magnetic and thermal actuation of the Hy-NC and the thermal actuator by magnetic heating and by increase in the temperature of the water. c) Reversible change in the bending angle of the Hy-NC (red) and the thermal actuator (black).
Here, $\lambda$ is thermal conductivity, $\rho$ is the density, and $C_p$ is the specific heat capacity of the material at temperature of interest. The bulk Hy-NC showed the maximum value of heat conductivity while, the porous layer which acts as heat insulator showed the minimum heat conduction in the temperature range from 25 to 100 °C. However, by increasing the temperature, a significant rise in the $\lambda$ values for three samples was observed (Figure 4b).

In porous systems, it is assumed that the total $\lambda$ can be described by four different contributions, as expressed in Equation (2).

$$\lambda = \lambda_v + \lambda_a + \lambda_r + \lambda_c \tag{2}$$

where $\lambda_c$ represents the thermal convection between neighboring cells, which can be neglected as the porous layer has closed cells with cell sizes $\approx 50 \mu m$.[37] $\lambda_a$ is the thermal conduction along the thick cell walls; $\lambda_r$ is contributed by the thermal conduction across the cells by the impulse transfer of gas molecules (air in cells) to the cell walls; and $\lambda_v$ is the heat transfer by electromagnetic radiation emitted by all surfaces. The rate of heat transfer by radiation is dependent on the temperature of a surface showing an increase by a factor of 3 with the increasing temperature of a surrounding water. An adequate selection of the magnetic field strength generated a sufficient heat in the composites material and initiated the magnetic actuation. The poor heat conductivity of the porous layers resulted in a controlled heat transfer to the surrounding water, which acted as a mediator to sequentially initiate the actuation of thermal actuators. It is anticipated that such multifunctional designs will stimulate future work in the development of novel thermal actuating materials enabling the coupling of other functions such as electric conductivity, photosensitivity, and actuation functionality.

In summary, a hierarchically structured composite device along with embedded thermal actuators enabled a sequential coupling of magnetic and thermal actuation functionalities. The porous layers of the device with closed cell morphology acted as heat insulation and provided a barrier to the surrounding water. An adequate selection of the magnetic field strength generated a sufficient heat in the composites material and initiated the magnetic actuation. The porous layers resulted in a controlled heat transfer to the surrounding water, which acted as a mediator to sequentially initiate the actuation of thermal actuators. It is anticipated that such multifunctional designs will stimulate future work in the development of novel complex actuating materials enabling the coupling of other functions such as electric conductivity, photosensitivity, and actuation functionality.

### Experimental Section

**Materials:** \(\alpha\)-pentadecalactone (PDL), dibutyltin dilaurate (DBTDL), 1,6-hexane diisocyanate (HDI), and dichloroethane from Merck (Darmstadt, Germany) were of analytical grade and were used as received. Polyethylene microspheres were purchased from Cospheric LLC (Santa Barbara, USA).

**Fabrication of the Device:** The synthesis of Hy-NC with covalently integrated ioNPs (8 wt%) was carried out according to the reported procedure.[39] For the fabrication of device, two separate reaction mixtures, the porous composite and bulk composite, were prepared. For the synthesis of porous composite reaction mixture, precursors 0.7 g (0.015 mmol of hydroxyl groups) of OPDL coated ioNPs (Mn = 3300 g mol\(^{-1}\) of OPDL chains determined by GPC) reported in ref. [32] and 2.3 g (0.4 mmol) of three arm OPDL (Mn = 3300 g mol\(^{-1}\) determined by GPC) were sonicated in 5 mL dichloroethane (DCE) for 10 min. Afterward, 0.17 mL (1.1 mmol) HDI and 3 mL (0.005 mmol) DBTDL and 0.3 g PE microspheres were added. The reaction mixture was stirred vigorously at 200 rpm (Heidolph, RZR2102, Schwabach, Germany) for 1 h. The reaction mixture was divided into two parts. One part was poured in a teflon mold (40 x 10 x 5 mm\(^3\)) and the other part was saved. The teflon mold was kept at 80 °C for 3 h under a constant nitrogen flow at 100 mbar. Meanwhile, the reaction mixture for bulk composite layer was prepared. For the synthesis of bulk composite, the same precursors (as used for porous composite) were used. Nevertheless, no PE microspheres were added for the synthesis of bulk composite. 3 mL of the reaction mixture were poured in the Teflon mold over the...
layer of prepolymer mixture of the porous layer. After a waiting period of 3 h at 80 °C, the remaining reaction mixture with PE spheres was poured into the Teflon mold to form the third layer of the device. The teflon mold was kept at 80 °C for 24 h under a constant nitrogen flow at 100 mbar and then at 100 °C for 6 h. The PE template was removed by extraction of the fabricated device with toluene at 120 °C for 3 days. Porous outer layers were gained by stepwise exchange of toluene with THF and afterward THF with water.

The thermal actuators, which were synthesized according to the reported procedure,[15] were embedded into the edges of the composite device by making small cuts into the middle bulk composite layer.

Thermal Analysis: The thermal analysis of the composites was carried out by using NETZSCH (Selb, Germany) DSC 204 in the temperature range from −100 to 100 °C with a constant heating and cooling rate of 5 K min⁻¹ under nitrogen environment. To determine the specific heat capacity \( C_p(T) \) in the experimental temperature range, the DSC was calibrated using sapphire (Al₂O₃) sample with a well-known specific heat capacity as standard. As usual the base lines of aluminum pan and sapphire reference were measured and compared with the heat flow measurements of the composite samples. The accuracy of the measurements is 3%.

Inductive Heating: The fabricated device was put in a glass beaker filled with 10 mL of water. Inductive heating of the device was accomplished by positioning the beaker in an alternating magnetic field at a frequency of \( f = 258 \) kHz. The equipment consisted of a high-frequency generator (TIG 5/300; Huettinger Electronic, Freiburg, Germany) and a water-cooled copper coil (six loops, diameter 4 cm, height 4.5 cm). The magnetic field strength \( H \) could be adjusted in between 7 and 30 kA m⁻¹ by variation of the power output of the generator. The measurement of \( T_{\text{bulk}} \) and \( T_{\text{water}} \) were carried out by self-made Cu-CuNi thermocouples, which were incorporated in the device.

Thermal Diffusivity: The thermal diffusivity (\( \alpha \)) measurements were carried out by Laser flash analysis LFA447 (Netzsch, Selb, Germany). For the measurements, disc shaped samples with thickness ranging between 2 and 4 mm and diameter = 12 mm were used. All samples were coated (both faces) with a very thin layer of colloidal graphite to improve their absorbency. To eliminate the samples transparency a thin layer (10 nm) of gold was sputtered on the faces of the specimen prior to the graphite coating. During the measurement, one side of the sample disc was heated with a laser (Xenon lamp = 15 J per pulse), resulting in a temperature gradient and heat propagating through the sample toward the opposite side. On the opposite side an infrared detector read and reported the temperature change with respect to time in the form of a thermogram. An estimate of the thermal diffusivity was obtained when these results were compared and fitted to theoretical predictions using a Cowan’s model.[16] Later on the computer was programmed to measure the values of \( \alpha \) by using the following equation
\[
\alpha = 0.139 \frac{L^2}{t_{1/2}} \text{ (mm}^2 \text{ s}^{-1})
\]
where \( L \) is the thickness of the samples in mm and \( t_{1/2} \) is the half rise time in seconds.[17] The principle of the laser flash measurement method and a typical curve of temperature versus time are shown in Figure S6, Supporting Information.

**Conflict of Interest**

The authors declare no conflict of interest.

**Keywords**

artificial muscles, magnetosensitivity, nanocomposites, soft actuators

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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