

## Magnetic responsive composites made from a sulfur-rich polymer

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A composite was prepared by hot-pressing a polysulfide polymer in the presence of  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> nanoparticles. The polymer was prepared by the direct copolymerisation of elemental sulfur and canola oil and the hot-pressing embedded the magnetic particles in the polymer. The resulting composite was used as a mercury sorbent capable of magnetic retrieval, and the magnetic particles allowed rapid microwave heating for curing into a consolidated object that could be remotely manipulated with a magnet. The multi-functional nature of the magnetic-responsive composite portends new frontiers for sulfur-rich polymers in environmental protection, recyclable materials, and magnetically activated machine components.

### Introduction

Magnetic responsive materials have found increasing use in biomedical devices,<sup>1, 2</sup> sensors,<sup>3</sup> sorbents for environmental remediation,<sup>2, 4</sup> and actuators for soft robotics.<sup>5, 6</sup> Among these functional materials, composites made from polymers with embedded magnetic particles have proven especially versatile.<sup>1, 3, 7</sup> Indeed, a wide range of commodity polymers can be melt casted, extruded, moulded or synthesised in the presence of inorganic fillers such as particles of Fe<sub>3</sub>O<sub>4</sub>,  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub>, FeN, FePd or Ni to provide these stimuli responsive materials.<sup>1, 3, 7</sup> Application of magnetic fields enable remote manipulation of the polymer composite, provoking responses such as shape changes, movement, or thermal actuation via induction.<sup>1, 3, 7</sup> As these composites become increasingly integrated into devices, there will be a need to develop reproducible and scalable syntheses, as well as methods to process and recycle these materials.<sup>2</sup> Accordingly, there is a need to develop novel polymer systems that impart these multifunctional capabilities.

In a step toward that goal, this report features a multi-functional, sulfur-rich polymer embedded with  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> nanoparticles. The dynamic S-S bonds in the polymer carrier allow highly versatile processing, novel moulding techniques, and mechanical and chemical recycling of the composite. The embedded nanoparticles impart the composite with magnetic-responsive properties that allow retrieval of the material from complex mixtures. This ability, coupled with the mercury affinity of the sulfur-rich polymer, was useful in water purification. The

$\gamma$ -Fe<sub>2</sub>O<sub>3</sub> particles also enable rapid heating of the composite with microwave irradiation, which was used as a technique to thermally fuse composite particles into desired shapes. This novel curing process was used to make a solenoid valve, which reduced the mass 10-fold in comparison to the commercial all-metal counterpart. Such advances are important for next-generation composite materials for a variety of electrical and mechanical systems as lighter weight can be important for fuel efficiency for mobile devices. The recyclability of the magnetic composite was an important feature for future integration into recyclable machine components.

The polymer matrix for the magnetic responsive composite was first made by the direct copolymerisation of equal masses of an unsaturated triglyceride (canola oil) and elemental sulfur, as previously described by our lab<sup>8, 9</sup> and related developments from the Theato laboratory.<sup>10</sup> This reaction and material was inspired by the development of *inverse vulcanisation* reported by Pyun and collaborators in which elemental sulfur is copolymerised with an unsaturated organic molecule, typically an alkene, diene, or polyene.<sup>11</sup> Inverse vulcanisation and related reactions provide organic polysulfides with sulfur content commonly ranging from 30–80 wt%. These unique polymers, rich in dynamic S-S bonds, have found use in a range of applications including cathode materials for Li-S batteries, novel materials for infrared optics, sorbents for environmental remediation, self-healing and repairable materials, thermal insulation, slow-release fertilisers, antimicrobial surfaces, adhesives, and many other applications—as chronicled in several recent reviews.<sup>12–18</sup>

Here, the featured sulfur-rich polymer, referred to as 50-poly(S-*r*-canola) to indicate it is a random copolymer made from 50 wt% sulfur and 50 wt% canola oil, was used as a binder to make a magnetic responsive composite in which the magnetic nanoparticles are embedded into the polymer after hot-pressing. Notably, sulfur polymers prepared by inverse vulcanisation have previously been investigated as components of polymer-metal nanocomposites. For instance, Pyun, Char and co-workers elegantly demonstrated that oleylamine and

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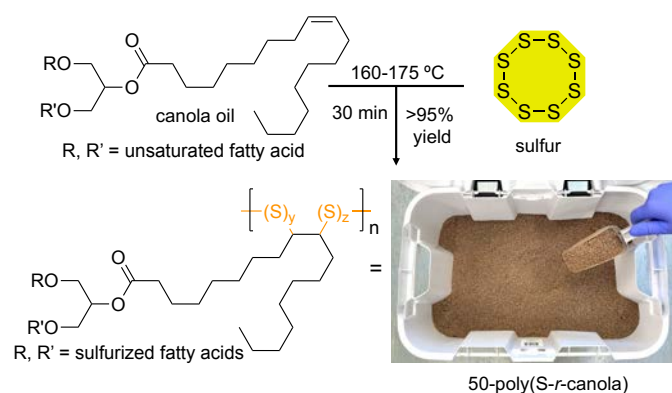
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sulfur could react and form the polymer carrier, with byproduct H<sub>2</sub>S serving as a reducing agent for in situ formation of PbS nanoparticles from PbCl<sub>2</sub>.<sup>19</sup> To complement this approach, we wanted to test if a preformed sulfur polymer could be embedded with metal nanoparticles in a simple, homogenous fashion and then serve as a magnetic responsive composite that is recyclable and useful in a number of applications. This goal would also expand how fillers and additives can enhance the physical properties and applications of sulfur-rich polymers.<sup>20-25</sup>

## Results and discussion

### Polymer and composite synthesis

The polymer 50-poly(*S-r*-canola) was prepared as previously described (Figure 1 and S3-S7).<sup>9, 26</sup> Briefly, canola oil (375 g) was heated to 170 °C and mixed with an overhead stirrer in a 5L stainless steel reactor. Elemental sulfur (375 g) was added next at a rate to maintain a temperature of 160 °C or greater. Next, sodium chloride (1.75 kg) was added, serving as a porogen to increase the surface area of the polymer, which facilitates grinding the final polymer into a powder. After the sodium chloride was added (over 25 minutes), taking care to maintain a temperature > 160 °C, the slurry thickened and darkened to a brown colour. As the polymerisation proceeds, the viscosity increased. When the overhead stirrer registered a torque of 40 N•cm, the impeller was stopped and the reactor was removed from the heat source to cool. The product was removed from the vessel and then ground with a pelletiser into particles approximately 0.5 to 3 mm in diameter. The sodium chloride porogen was then removed by repeated washing with water. The polymer was isolated with a sieve and dried in a tray in a fume hood to provide the final polymer (typically >95% yield). Thermochemical and spectroscopic characterization data (<sup>1</sup>H NMR, TGA, and IR) were consistent with this previously described method of synthesis (S8-S11).<sup>26</sup> The soft, friable rubber had a *T*<sub>g</sub> = -26 °C and combustible analysis was consistent with high atom economy and the expected 50 wt% sulfur (S11). SEM and EDX analysis revealed a relatively uniform distribution of sulfur throughout the bulk polymer (S10).



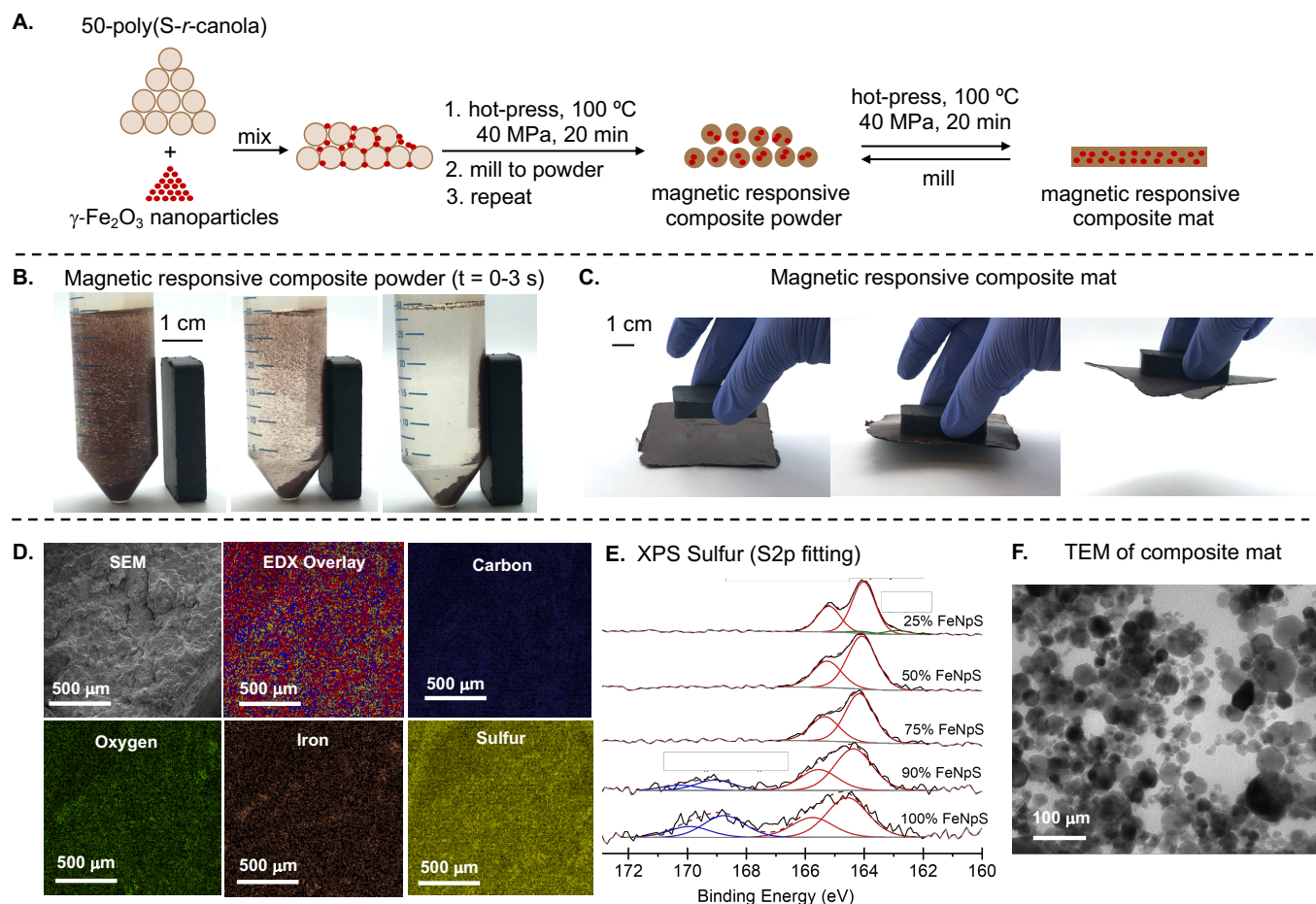
**Figure 1.** Equal masses of canola oil (an unsaturated triglyceride) and sulfur were reacted directly to form sulfur-rich copolymer. This synthesis was carried out as previously described.<sup>26</sup>

On this reaction scale, efficient fume extraction is required to prevent exposure to potentially harmful off-gases such as H<sub>2</sub>S, SO<sub>2</sub> and gaseous elemental sulfur.<sup>10, 27</sup> With that said, lead acetate paper suspended in the head space of the reaction vessel indicated minimal H<sub>2</sub>S production, which means this protocol provides relatively good control of the reaction exotherm and prevents polymer decomposition. In contrast, a positive indication of H<sub>2</sub>S was clearly observed during intentional combustion of a 0.5 g sample of 50-poly(*S-r*-canola) (S5-S7).

The 50-poly(*S-r*-canola) was chosen as the polymer binder for the proposed magnetic responsive composites for several reasons. First, it is a soft, friable rubber that can easily consolidate around fillers when hot pressed.<sup>21, 24</sup> Second, hundreds of kilograms of this material can be made industrially so it is readily available for integration into new technologies.<sup>16</sup> Third, the polysulfide is known to have affinity for iron, which was hypothesised to facilitate binding to the magnetic iron particles.<sup>28</sup>

As a first test, 1.0 g of  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> nanoparticles were physically mixed with 6.5 g of 50-poly(*S-r*-canola) and then hot-pressed at 100 °C and 40 MPa for 10 minutes (S12). Encouragingly, the resulting mat was shape persistent and attracted to a hand-held magnet (Figure 2). Next, the mass ratio of the nanoparticles to polymer binder was varied to determine the maximum amount of particles that could be incorporated (for increased responsiveness to an external magnet), while also maintaining physical integrity of the mat. It was found up to 75% of the mass of the composite could be  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> nanoparticles, with higher values resulting in a fragile mat (S13-S14). It was also discovered that milling the mat back to a powder and then re-pressing at 100 °C and 40 MPa for 20 minutes resulted in more uniform consolidation of the polymer around the  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> particles.

Scanning electron microscopy (SEM) was used to confirm that repeated milling and hot-pressing resulted in more uniform distribution of the particles throughout the composite (Figure 2D). Interestingly, when the composite was milled back into a powder, the  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> particles did not appear to physically separate from the polymer and so the magnetic responsiveness of the composite was maintained (Figure 2B). It was thought that the sulfur content of the polymer may bond to the iron particles, so this hypothesis was tested using X-ray photoelectron spectroscopy (XPS). However, no evidence for Fe-S bonds were detected, which would otherwise be expected at 162.5 eV (Figure 2E).<sup>29, 30</sup> For this reason, we tentatively concluded that the iron particles are simply embedded in the polymer matrix rather than bonded to the sulfur in the polymer. X-ray diffraction (XRD) revealed the crystalline structure of the iron particles remained consistent throughout the processing (S18). TEM also showed that the iron particles remained crystalline and distributed through the polymer matrix and did not aggregate extensively (Figure 2F, S19-S23). This distribution of  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> particles through the polymer matrix was sufficient for consistent response to external magnets such that even the powdered material exhibited good mass balance in magnetic retrieval (Figure 2B).



**Figure 2.** **A)** Repeatedly hot-pressing and milling a mixture of 50-poly(*S-r-canola*) and  $\gamma$ - $\text{Fe}_2\text{O}_3$  nanoparticles results in a magnetic responsive composite. The composite can be fashioned into a powder or a mat. **B)** Demonstration of the magnetic responsive composite powder in water (25 wt%  $\gamma$ - $\text{Fe}_2\text{O}_3$  nanoparticles and 75 wt% 50-poly(*S-r-canola*)). Magnetic immobilization was achieved in 3 seconds. **C)** Demonstration of the magnetic responsive composite mat. (25 wt%  $\gamma$ - $\text{Fe}_2\text{O}_3$  nanoparticles and 75 wt% 50-poly(*S-r-canola*)). **D)** Scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy of a cross-section of the 25 wt%  $\gamma$ - $\text{Fe}_2\text{O}_3$  composite mat revealed a relatively uniform distribution of iron in the polymer matrix. **E)** X-ray photoelectron spectroscopy was used to determine if the sulfur in the polymer binder forms a bond to iron. No evidence of an Fe-S bond was found, which would be expected at 162.5 eV. **F)** Transmission electron microscopy (TEM) of the magnetic responsive composite mat showed the crystalline  $\gamma$ - $\text{Fe}_2\text{O}_3$  particles (25 wt% of the composite) distributed through the polymer matrix. The  $\gamma$ - $\text{Fe}_2\text{O}_3$  particle sizes ranged from approximately 5-250 nm in size and did not aggregate during hot-pressing.

The magnetic responsive composite was also prepared by direct addition of the  $\gamma$ - $\text{Fe}_2\text{O}_3$  particles during the synthesis of the 50-poly(*S-r-canola*) polymer (S24-S27). However, this protocol slowed the polymerization, which took more than 1 hour to vitrify even on a small scale (20 g). The resulting composite was still responsive to an external magnet, but because of the potential for the iron particles to interfere in the polymerization, this protocol was not pursued further.

These initial experiments provided a simple route to magnetic responsive polymer-metal composites. And while polymer-metal composites have been prepared previously by hot-pressing a mixture of powdered polymers and metals, these approaches typically rely on thermoplastic properties of polymers such as polymethylmethacrylate that soften and flow during melt processing.<sup>31</sup> In contrast, the 50-poly(*S-r-canola*)

polymer does not liquify at the processing temperature due to its crosslinked nature. Rather, the particles are thought to bind together and consolidate through S-S metathesis reactions at the polymer interfaces, resulting in the magnetic particles becoming embedded in the polymer.<sup>21</sup> With ready access to the magnetic responsive composite, its applications were investigated next.

#### Mercury remediation

A composite mat was made as described above using a 3:1 mass ratio of 50-poly(*S-r-canola*) to  $\gamma$ - $\text{Fe}_2\text{O}_3$  nanoparticles. The hypothesis was at this composition there will be enough polymer to be an effective mercury sorbent and enough nanoparticles to enable magnetic retrieval of the sorbent. This mat was then milled into a powder and hot-pressed again for 20

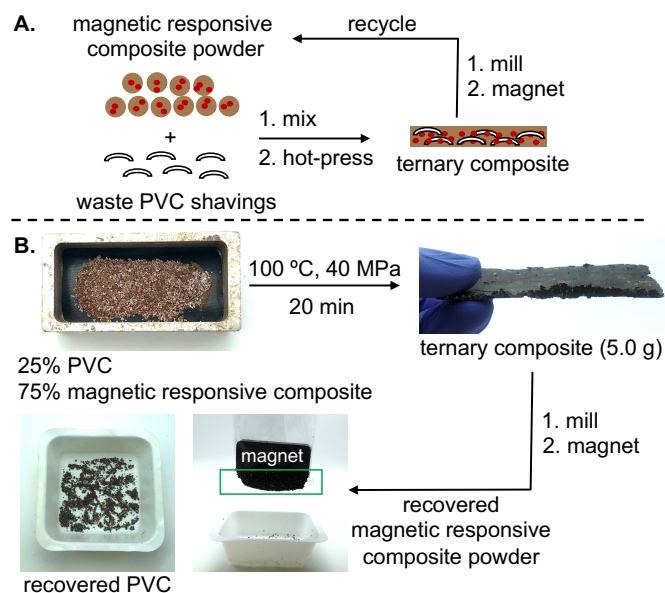
minutes at 100 °C and 40 MPa, to complete the consolidation process. A final milling provided the composite as a powder for investigation in mercury remediation. It has previously been shown that 50-poly(*S-r*-canola) has a high affinity for a variety of mercury species.<sup>9,32</sup> However, the retrieval of the 50-poly(*S-r*-canola) sorbent in these studies was done by simple filtration. And while such a retrieval method is fine for homogenous liquid samples contaminated with mercury, the polymer sorbent is much more difficult to isolate in complex mixtures of liquids and solids. The magnetic responsive composite, in contrast, was anticipated to overcome these limitations.

To demonstrate magnetic retrieval after mercury remediation, 1.0 gram of the magnetic composite was added to a mixture containing HgCl<sub>2</sub> (10 ppm), 3.0 g of solid mine tailings, and 20 mL of water. The mixture was rotated end-over-end for 24 hours. After this time a magnet was used to immobilise the magnetic particles, which allowed the non-magnetic tailings and water to be decanted from the tube. Good mass balance of the magnetic sorbent was observed, with 1.0 g recovered in triplicate runs (S29). The solid tailings and water were then separated by simple filtration and the mercury content of the water was measured using cold vapour atomic absorption (CVAA) spectroscopy. The mercury concentration in the water was reduced to < 7 ppb, which is a 99.9% reduction in mercury from the original mixture. And while sulfur-rich magnetic materials for mercury sorption have been reported previously by Hasell and collaborators,<sup>33</sup> the composite reported here is easier to prepare and demonstrated on a more complex field sample from a mine. Finally, it was demonstrated that the iron could be separated from the mercury and recovered using a thermal reduction-desorption process adapted from the analogous process for carbon-based sorbents (S30-S32).<sup>34</sup> While this process resulted in a phase change from  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> to  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>, the latter still retains magnetic properties and therefore facilitates recycling of the iron.

### Ternary composite preparation and recycling

In the same way the magnetic composite was prepared by hot-pressing 50-poly(*S-r*-canola) and  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> nanoparticles, other composites with additional components can be made. For example, we have previously shown that 50-poly(*S-r*-canola) can bind together composites containing waste plastic, sand, carbon fibre, or natural fibres such as wool.<sup>21, 23, 24</sup> And while re-milling these composites does allow, in some cases, for the polymer to be recovered based on differences in density,<sup>24</sup> there is still a need to introduce more efficient recycling methods. Therefore, the magnetic responsive composite made from a 3:1 mass ratio of 50-poly(*S-r*-canola) to  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> nanoparticles was evaluated as a recoverable binder. In this case, waste polyvinyl chloride (PVC) shavings (1.0 g) were mixed with the magnetic composite powder (4.0 g) and then hot-pressed into a 3-component composite mat for 20 minutes at 100 °C and 40 MPa (Figure 3). It has been previously proposed that this method of composite preparation could be used to re-purpose plastic waste into mats with prospective use in tiling, bumpers, walkways or construction materials.<sup>21</sup> The novel feature in this preparation, however, is that the mat can be re-

milled, and the magnetic composite separated from the PVC using a magnet. This demonstration (Figure 3) is another example of how the magnetic responsive component can be separated from complex mixtures. In this application, the original components can be recovered which makes recycling of these composites more controlled.



**Figure 3.** **A)** Concept for recycling and separating a 3-component composite back into its original components. **B)** Implementation of the composite recycling process for a composite made from waste PVC and the magnetic responsive powder. The recovered magnetic responsive powder is shown in the green box. The magnet was contained in a plastic bag to simplify the recovery of the magnetic material.

Additionally, mechanical testing of the ternary materials made from waste PVC and the magnetic-responsive composites was undertaken (S33-S35). The addition of  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> resulted in an increase of 25% in tensile modulus and over 90% in flexural modulus in comparison to a sample made from only hot-pressed polymer. When the ternary composite with PVC was produced, the compression and flexural moduli increased significantly while the tensile modulus decreased. The composites with PVC lost much of their flexibility, becoming more rigid and brittle. These observations indicate that  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> improves the tensile properties of the composite while PVC is acting as an inflexible particle which dominates the compressive properties. Overall, these results indicate that the  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> not only imparts magnetic responsiveness, but also can be used to modify mechanical properties of the composites.

### Microwave curing

The  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> component of the magnetic responsive composite rapidly increases temperature when exposed to microwave irradiation.<sup>35</sup> It was thought that this property could be useful in curing the composite into a desired shape. It has previously been shown that 50-poly(*S-r*-canola) and related sulfur-rich

polymers are excellent thermal insulators,<sup>23, 36</sup> so efficient heating and thermal curing of this polymer is not trivial and the  $\gamma\text{-Fe}_2\text{O}_3$  particles were therefore evaluated as microwave absorbing susceptors to mediate heating of the composite for curing.

First, 1100 W microwave irradiation was used to assess how rapidly each component changes temperature. Over 40 seconds of irradiation, 50-poly(*S-r*-canola) did not increase in temperature, nor did the PTFE sheet on which it was placed, as measured with an infrared thermometer. However, the  $\gamma\text{-Fe}_2\text{O}_3$  nanoparticles and the magnetic responsive composite increased in temperature linearly over this time (Figure 4A). It was also found that the temperature increased more rapidly for the composite with increasing amounts of  $\gamma\text{-Fe}_2\text{O}_3$  nanoparticles, with heating rate directly proportional to the mass % of  $\gamma\text{-Fe}_2\text{O}_3$  (S37-S41). Importantly, this information helped determine how long the composite should be irradiated with microwaves to induce the S-S metathesis and consolidation. To demonstrate this capability in curing, 800 mg of the powdered composite (25 wt%  $\gamma\text{-Fe}_2\text{O}_3$  nanoparticles and 75 wt% 50-poly(*S-r*-canola) was placed in the barrel of a 20 mL plastic syringe and compressed using the plunger. Next, the composite-loaded syringe was irradiated with microwaves (1100 W) for 3 × 30 seconds to provoke S-S metathesis and polymer consolidation around the  $\gamma\text{-Fe}_2\text{O}_3$  nanoparticles. This process provided a composite disc, demonstrating that rapid microwave heating can be used to bind the polymer together around the magnetic particles (Figure 4B and S41-S42).

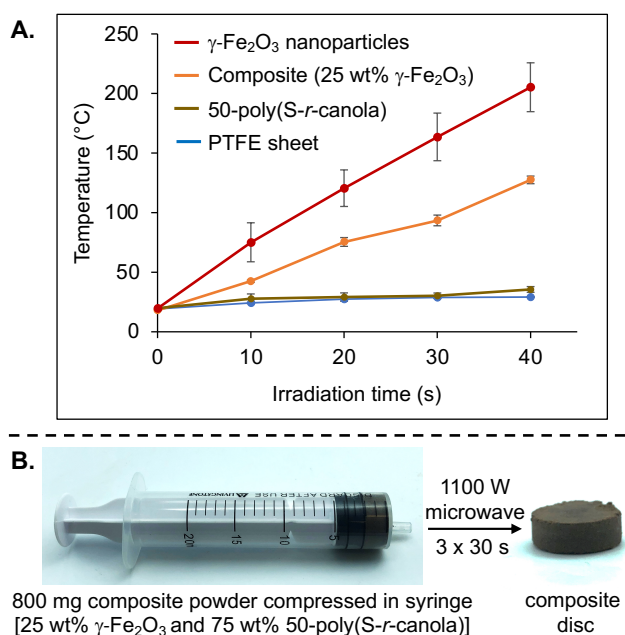
responsive composite powder into a solid disc. The consolidation is thought to occur by thermally induced S-S metathesis of the 50-poly(*S-r*-canola) polymer that binds the powder together.

#### Use of magnetic-responsive composite as machine component

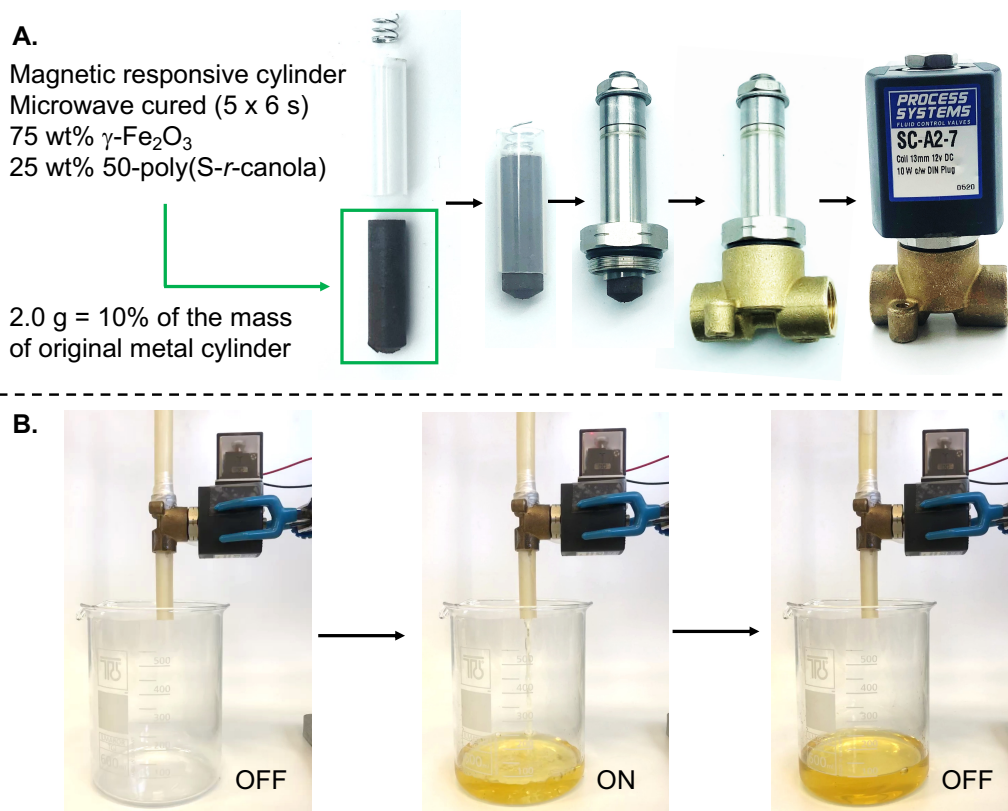
Next, the microwave curing was used to prepare a magnetic responsive machine component. Accordingly, the composite powder was prepared with 75 wt%  $\gamma\text{-Fe}_2\text{O}_3$  nanoparticles and 25 wt% 50-poly(*S-r*-canola). The high  $\text{Fe}_2\text{O}_3$  content was required to ensure sufficient magnetic response for its use in a solenoid valve. Additionally, the syringe barrel used in the curing process provided a composite cylinder with a diameter that could fit directly into the valve (Figure 5 and S43-S44). In the microwave curing process, 2.0 g of the composite powder was loaded into the syringe barrel and compressed by hand. The system was then heated in a 1100 W microwave. Shorter irradiations (5 × 6 s) were required to cure the sample because of the higher iron content. The consolidated composite was removed from the syringe and was then shown to operate effectively in the solenoid: when electrical current was passed through the solenoid coil, the magnetic force generated caused the composite cylinder to move and open the valve. Turning off the current removes the magnetic force, and a spring forces the cylinder back in the path of the fluid, closing the valve. The composite cylinder was 10% of the mass of the original all-metal component and was equally effective in the valve operation. It is also worth noting that the rubber-like and hydrophobic nature of the sulfur polymer in the composite is useful for creating a tight seal in the valve. This is a demonstration of how polymer-based composites can be lightweight replacements of machine components that are traditionally composed entirely of metal. Such capabilities are important in soft robotics as well as machines that require lighter weight for more efficient transport.

#### Conclusions

A multifunctional composite was made by hot-pressing a sulfur-rich polymer with  $\gamma\text{-Fe}_2\text{O}_3$  nanoparticles. This process induces S-S metathesis reactions that enable the polymer to encapsulate the nanoparticles. The resulting material was demonstrated as an effective sorbent for mercury, with magnetic retrieval of the sorbent facilitating the remediation of complex mixtures. The composite could also be used as a recyclable binder for the preparation of higher order bulk composites. The composite was also amenable to moulding into various shapes under microwave irradiation, with the heating rate proportional to the  $\gamma\text{-Fe}_2\text{O}_3$  content. This novel curing method was used to manufacture a lightweight solenoid valve. Future applications of these materials and methods will include integration into recyclable machines, electronics, and soft robotics.



**Figure 4.** **A)** Over a 40 second period, microwave irradiation (1100 W) of  $\gamma\text{-Fe}_2\text{O}_3$  nanoparticles or composites containing  $\gamma\text{-Fe}_2\text{O}_3$  nanoparticles results in a linear increase in temperature. **B)** Microwave irradiation was used to mold the magnetic



**Figure 5.** A magnetic responsive composite cylinder was prepared by curing 2.0 g of the magnetic composite powder in a syringe. The magnetic responsive cylinder (green box) was then used to replace a metal valve in solenoid. **B.** The solenoid in operation. The magnetic field generated when the power is on is sufficient to move the cylinder against the spring and open the valve. When the power is turned off, the cylinder no longer experiences a magnetic force, the spring forces the cylinder back into a position where the flow of the liquid is stopped.

### Conflicts of interest

There are no conflicts to declare.

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