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Insulating Composites Made from Sulfur, Canola Oil, and Wool

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Insulating composites made from sulfur, canola oil, and wool

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Introduction

Consideration of the feedstocks, manufacturing processes, and lifecycle of the materials in our built environment is an important aspect of sustainability.[1, 2] In this context, innovations in sustainable composites for construction and insulation are required.[3-5] Motivated by this need, we investigated a novel composite derived from sulfur, canola oil, and raw wool and evaluated its potential as thermal insulation (Figure 1). Sulfur is an attractive building block because it highly abundant geologically and nearly 80 million tonnes is produced each year as a byproduct of petroleum refining.[6] Elemental sulfur[7] and, more recently, polymers made with high sulfur content[8, 9] have shown promise as low-cost and sustainable thermal insulation. However, the mechanical strength and durability of these high-sulfur materials are not suitable for many applications in construction and transport. It has recently been established that judicious selection of fillers and crosslinkers can improve the mechanical performance of sulfur polymers and related composites.[10-14] We considered that preparing composites from sulfur polymers and natural and sustainable fibers such as wool[15, 16] might further improve the utility and sustainability profile of these materials. We were also motivated to explore the polymer made from sulfur and canola oil because it can be prepared on multi-kg scale[17] and it is derived from low-cost and sustainable building blocks.

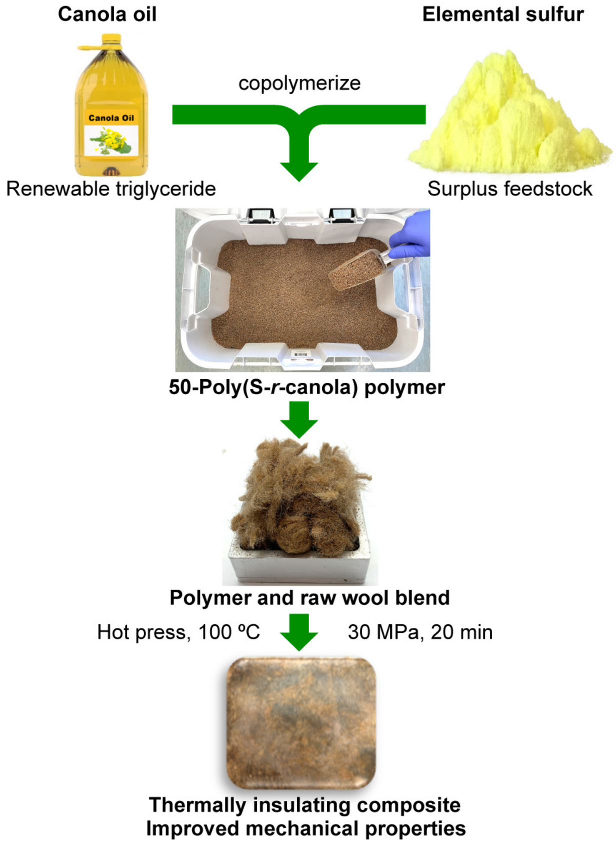

Figure 1. The aim of this study was to evaluate a composite made from sulfur, canola oil, and wool as thermal insulation. The material is prepared by hot pressing raw wool with a polymer made from sulfur and canola oil. The polymer binds together through an S-S metathesis mechanism we refer to as reactive compression molding.

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In pursuing this hypothesis, we took advantage of the unique properties of polymers made by inverse vulcanization—a copolymerization process between elemental sulfur and a polyene that results in versatile organic polysulfides.[18-22] The S-S bonds in these polysulfide polymers can be readily exchanged with mild heating (≤ 100 °C) and pressure (10 to 40 MPa) to force together the reactive interfaces and shape the polymer into a desired architecture—a process we refer to as *reactive compression molding*.[12] We have recently shown that reactive compression molding can be used as a process to embed filler materials into the polymer, providing straightforward access to diverse composites.[12] To reinforce the polymer, unprocessed wool was used directly in the composite synthesis.[15, 16] This strategy provided a material with greater tensile strength, reduced thermal conductivity, and greater flame resistance relative to the base polymer. The composite obtained is therefore a promising material for energy saving insulation and it is made from low-cost, abundant and sustainable feedstocks

Results and discussion

First, a polymer was prepared by the copolymerization of sulfur and canola oil, as previously described by our laboratory.[17, 23] Briefly, sulfur (450 g) and canola oil (450 g) were reacted directly at 175 °C in a 5 L stainless steel reactor with mixing from an overhead mechanical stirrer. The temperature of the reaction was monitored and controlled directly by inserting the temperature probe directly into the reaction mixture, with heat provided from a hotplate. Sodium chloride (2.10 kg) was added slowly to the reaction mixture, serving as a porogen to increase the surface area of the final polymer. Continued heating and mixing for 20 minutes after the addition of the sodium chloride led to a rapid increase in viscosity as the polymerization occurs. When the torque of the overhead stirrer registered 40 N•cm, the reaction was removed from the hotplate. The polymer was then removed from the reactor and ground into a powder. The sodium chloride was removed by repeated washing with water and the polymer powder (0.5-3.0 mm) was dried in a fume hood. The final polymer is a soft, compressible rubber powder. The thermal, mechanical, and spectroscopic properties of this polymer were consistent with our previous reports on this material.[17, 23, 24] This polymer is referred to as 50-poly(S-*r*-canola), with 50 indicating the mass percent of sulfur and *r* indicating that it is a random copolymerization.[25] Our working hypothesis of the mechanism of this reaction, and a general structure of the polymer is shown on page S4.

Next, wool from a Border Leicester sheep was obtained from a local farm, directly after shearing. The raw wool was not washed or pre-treated with any reagent in order to keep the composite synthesis as simple as possible and generate less waste. A sample of the wool fibers was analyzed using a scanning electron microscope (SEM), which revealed an average fiber diameter of 34 ± 3 µm.

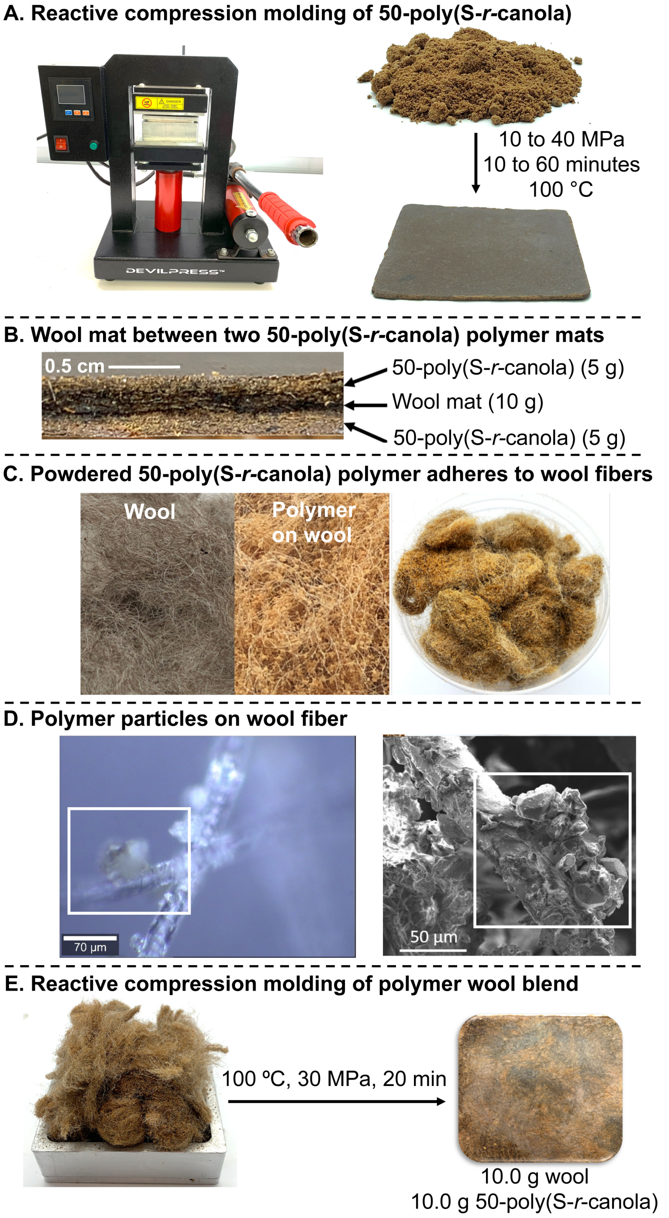


Figure 2. A. Reactive compression molding converts the 50-poly(S-*r*-canola) to a polymer mat. B. Hot pressing a mat of wool (10 g) between two mats of 50-poly(S-*r*-canola) polymer (5 g each) (100 °C, 40 MPa, 20 min) provides a composite mat (cross-section shown). C. Powdered 50-poly(S-*r*-canola) polymer adheres to the wool fibers, facilitating relatively even mixing of the polymer and wool. D. Images of polymer particles bound to the wool fiber. E. Reactive compression molding of the wool-polymer blend (100 °C, 30 MPa, 20 min) provides a composite mat.

With the polymer and wool in hand, composite synthesis by reactive compression molding was investigated. The samples were processed in a 10 × 10 cm aluminum mold, with controlled heating and compression provided by a press (Figure 2A). Poly(tetrafluoroethylene) (PTFE) sheets were used in the press to prevent the polymer from adhering to the metal mold. Based on our previous study in reactive compression molding,[12] applying a pressure between 10 and

40 MPa with heating at 100 °C for 10-60 minutes induces S-S metathesis reactions that convert the powdered 50-poly(S-*r*-canola) into a solid polymer mat or block. This process can also be used to prepare composites by applying pressure and heat to a mixture of the polymer and a filler material, such as wool.

First, the powdered 50-poly(S-*r*-canola) was converted to a 15.0 g polymer mat by reactive compression molding (100 °C, 20 MPa, 15 min). This polymer mat served as a control sample, but it could also be used as a substrate for additive compression molding in which multiple polymer mats can be reacted to form a thicker mat or composite (*vide infra*). The wool alone (without added polymer) was also processed in the hot press to determine if this process resulted in any wool to wool adhesion or fiber damage. After processing 10 g of the raw wool (100 °C, 40 MPa, 20 min) a mat of wool was obtained indicating some adhesion between the fibers. SEM micrographs revealed the fibers to be compressed and flattened to a width of 40 ± 4 µm (S8), but the fiber structure appeared intact. If two of these wool mats were re-subjected to the molding process, the mats did not adhere together (S9). In a control experiment using wool that was washed in organic solvent, the wool mat still formed in the hot press, indicating that the wool oils are not required for the wool fibers to bind together (S16).

To access composite materials made from 50-poly(S-*r*-canola) and the wool, several procedures were evaluated. PTFE sheets were used in all experiments to prevent the polymer or wool binding to the aluminum mold. In the first method, the pre-formed wool mat (10 g) was sandwiched between two of the pre-formed polymer mats (15 g each) and then subjected to reactive compression molding (100 °C, 40 MPa, 20 min). The result was a single mat made of three layers: two outer polymer layers and a middle layer of wool (Figure 2B). It appeared that at some regions in the composite, the polymer mats actually come into contact during the compression through the wool layer, causing all three layers to adhere. In a control experiment using wool that was washed with organic solvent, the polymer still adhered to the wool and formed the mat during reactive compression molding. This control experiment indicated that the oils on the surface of the wool, such as lanolin,[26] were not required for polymer binding. A cross-section of the composite clearly showed the fibrous structure of the wool layer (S11-S12).

In the second method of composite preparation, the powdered 50-poly(S-*r*-canola) (10.0 g) and the raw wool (10.0 g) were mixed together directly. Fortuitously, the powdered polymer readily holds static charge which facilitates its binding to the wool fiber. The polymer adhesion to the wool was observed by optical and Raman microscopy, as well as scanning electron microscopy (Figure 2C-D and S13-S14). We have previously observed similar electrostatic adhesion in blends of the polymer and activated carbon.[27] This phenomenon is useful in the composite synthesis context of this study because it helps create a relatively even distribution of polymer across the wool fiber surface. Subjecting the polymer-coated wool to reactive compression molding (100 °C, 30 MPa, 20 min) results in the formation of a composite mat in which the wool fibers are randomly distributed through the polymer binder (Figure 2E).

The third method of composite manufacture used aligned wool fibers compressed between two pre-formed mats of 50-poly(S-*r*-canola). The wool fibers (2.0 g, 11 cm) were combed straight and clipped in the mold so they were taut across the surface of the bottom polymer mat (5.0 g) (Figure 3A). The top polymer mat (5.0 g) was then added to the mold and processed with the hot press (100 °C, 40 MPa, 40 min), providing the composite mat with aligned wool fibers.

The fourth method of composite preparation featured two non-woven middle layers of aligned wool (2.0 g of wool each) sandwiched between to outer layers of polymer mats (5.0 g of polymer each). The two wool mats had their fibers aligned orthogonally to each other. To make this composite, wool (2.0 g) was stretched taut across the bottom pre-formed polymer mat (5.0 g) and processed in the hot press (100 °C, 30 MPa, 30 min), which embeds the wool fiber in the bottom polymer mat. The excess wool was trimmed off of the edges and then the second aligned wool sample (2.0 g) was placed orthogonally and on top of the bottom wool layer and clamped into place. The top pre-formed polymer mat was then added and then processed in the hot press (100 °C, 30 MPa, 30 min) to provide the final composite mat (Figure 3B and S19).

With the panel of composites in hand, tensile strength was assessed using a Dynamic Mechanical Analyzer (DMA). Each sample was prepared by cutting the polymer mat or composite into a rectangular sample (3.0 x 0.5 cm) with a scalpel. The test was carried out in tension mode of the DMA instrument with ramp displacement of 1000 mm/min. A stress-strain plot was obtained, from which the Young's modulus, the yield strength, and the percent elongation at fracture were determined (S20-S23). In this analysis, data was only used when the fracture occurred in the middle of the sample (fractured samples are shown on page S21). If the fracture occurred at the clamps, the data was not analyzed. The modulus of elasticity (Young's modulus) for each sample are compiled in Figure 4. As expected, the polymer alone had the lowest modulus of elasticity and the lowest yield strength. All samples with wool were superior in this regard to the base polymer. The highest modulus of elasticity was observed for the composite in which the wool fibres were aligned and the tensile force was applied along the length of the fibers. The tensile modulus for this sample was increased 10-fold over the base polymer. Even the mixed composite with random wool positioning or the composite made from the pre-formed polymer and wool mats were superior in yield strength and modulus compared to the polymer alone. This result indicates that improved material properties can be imparted to the base polymer by the simple process of reactive compression molding without any requirement to position the fibers in a specific orientation.

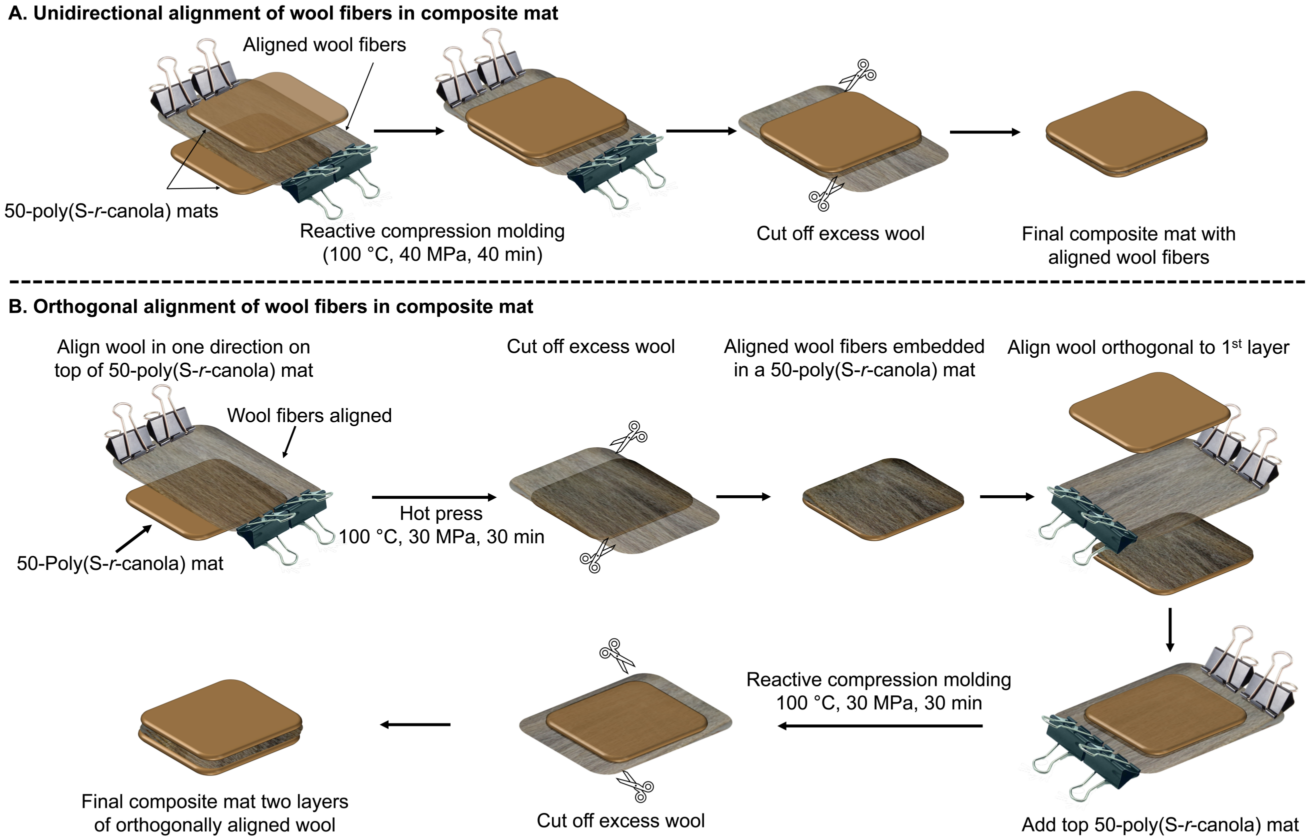
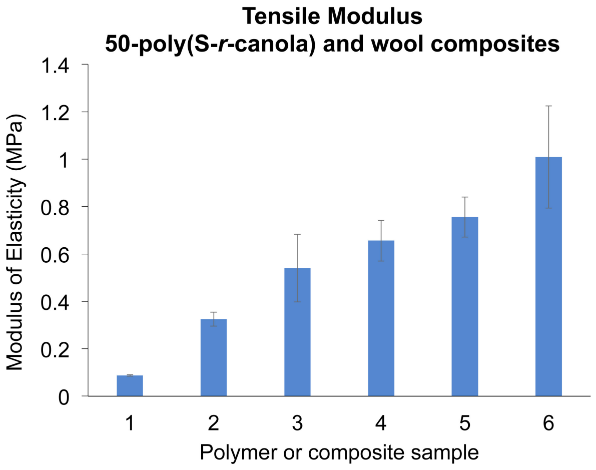


Figure 3. A. Preparation of a composite mat with wool fibers aligned in one direction. B. Preparation of composite mat in which two wool layers are aligned orthogonally.

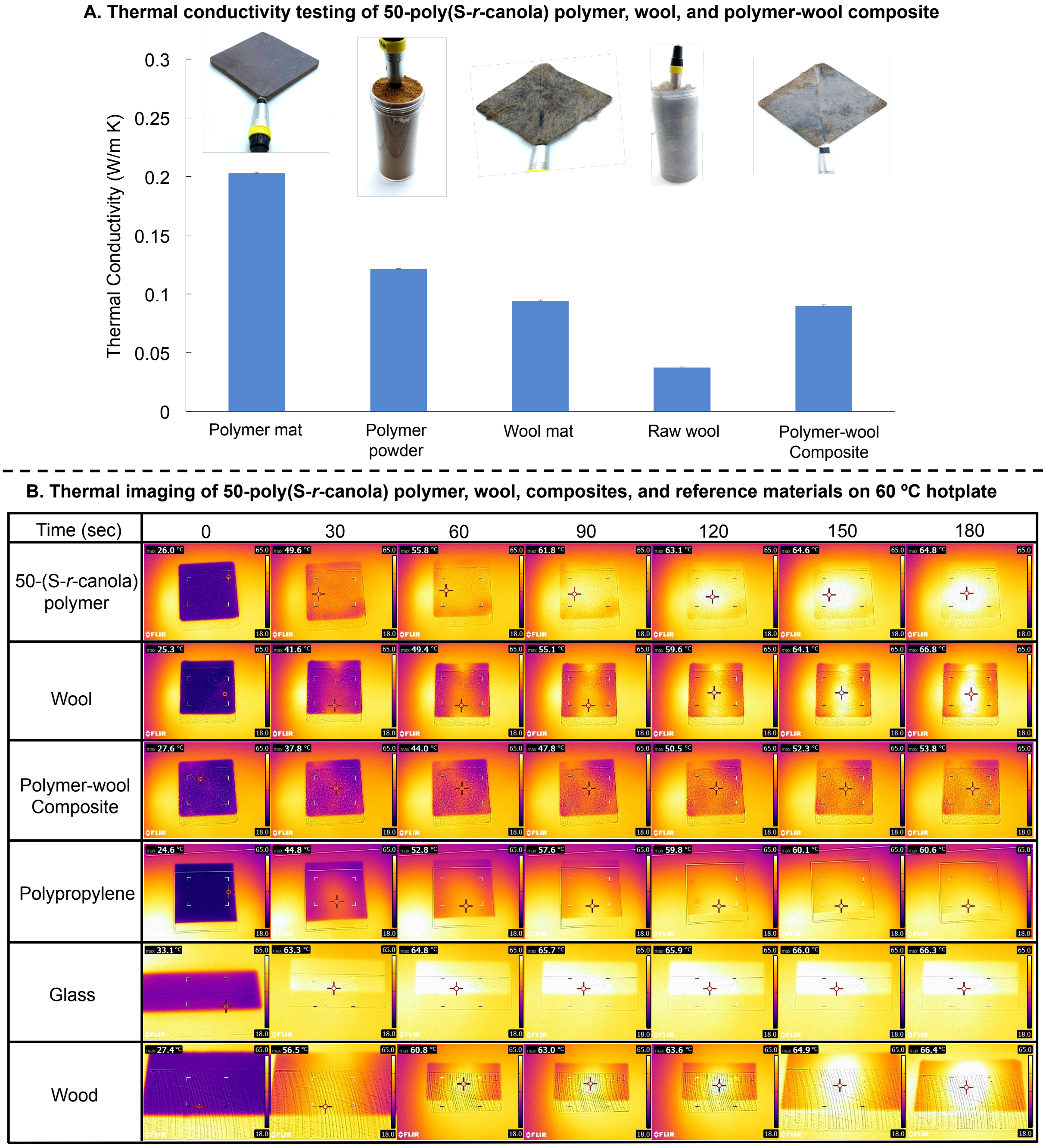
The thermal conductivity of the 50-poly(S-*r*-canola) polymer, wool, and composite material were evaluated next. To ensure a suitable thickness for the thermal conductivity probe, iterative reactive compression molding was used to make a 50-poly(S-*r*-canola) polymer mat. Accordingly, pre-formed polymer mats were molded around the thermal conductivity probe needle, which allowed the formation of a suitable cavity in the mat for the thermal conductivity testing (S24-S25). A similar process was used to make a wool mat, as well as the 50-poly(S-*r*-canola) and wool composite mat. The composite mat was made of equal massses of wool and 50-poly(S-*r*-canola) (10.0 g each). Thermal conductivity was measured using a Thermtest TLS-100 probe for all three mats, as well as the powdered polymer and the uncompressed wool (Figure 5A and S24-S29). The polymer mat had a measured conductivity of 0.20 $Wm^{-1}K^{-1}$, which is the same as elemental sulfur (~0.2 $Wm^{-1}K^{-1}$).[9, 28, 29] This result indicates the 50-poly(S-*r*-canola) polymer retains the insulating property of the monomer precursor, sulfur. The composite mat made from equal masses of wool and polymer had an even lower thermal conductivity (0.09 $Wm^{-1}K^{-1}$). And while this measured thermal conductivity of the composite made from wool and 50-poly(S-*r*-canola) is slightly higher than air and foamed materials (typically 0.02-0.04 $Wm^{-1}K^{-1}$), it is lower than lightweight concrete (0.1-0.3 $Wm^{-1}K^{-1}$), typical bulk polymers (0.1-0.5 $Wm^{-1}K^{-1}$),[30] and glass (~1 $Wm^{-1}K^{-1}$).[29]



1. Polymer control: 50-poly(S-*r*-canola)
2. Composite: aligned wool (tension orthogonal to axis of wool fibers)
3. Composite: random wool alignment
4. Composite: wool mat between two 50-poly(S-*r*-canola) mats
5. Composite: crosshatched wool
6. Composite: aligned wool (tension along axis of wool fibers)

Figure 4. Tensile testing of 50-poly(S-*r*-canola) and wool composites using a dynamic mechanical analyzer revealed an increase in modulus of elasticity of all polymer-wool composites compared to the polymer alone.

To visually illustrate the insulative properties of the composite mat made from 50-poly(S-*r*-canola) and wool (equal masses and randomly mixed before reactive compression molding), an infrared camera was used to image the composite mat after placing it on a metal sheet heated to 60 °C. All materials tested had a thickness of 1.3 mm. The camera was set to scan and record the hottest point of the mat every 30 seconds over a period of 3 minutes. The composite mat heated up more slowly over this time than the polymer alone, a wool mat, polypropylene, glass, and wood (Figure 5B). The composite also displayed the highest heat retention during cool down (S31). This slower dissipation of heat is consistent with its low thermal conductivity. This is a promising lead in the preparation of composite materials for construction and insulation.



A. Thermal conductivity testing of 50-poly(S-*r*-canola) polymer, wool, and polymer-wool composite

B. Thermal imaging of 50-poly(S-*r*-canola) polymer, wool, composites, and reference materials on 60 ºC hotplate

Figure 5. A. The thermal conductivity of a 50-poly(S-*r*-canola) polymer mat and powder, wool mat and free wool, and the polymer-wool composite (random wool alignment, equal masses of wool and 50-poly(S-*r*-canola)) was measured using a Thermtest TLS-100 probe. B. Thermal imaging of 50-poly(S-*r*-canola) polymer mat, wool mat, polymer-wool composite (random wool alignment, equal masses of wool and 50-poly(S-*r*-canola)), and reference materials was carried out after placing 1.3 mm sample on a hotplate preheated to 60 °C. The polymer-wool composite heated up the most slowly before thermal breakthrough, demonstrating its insulating properties.

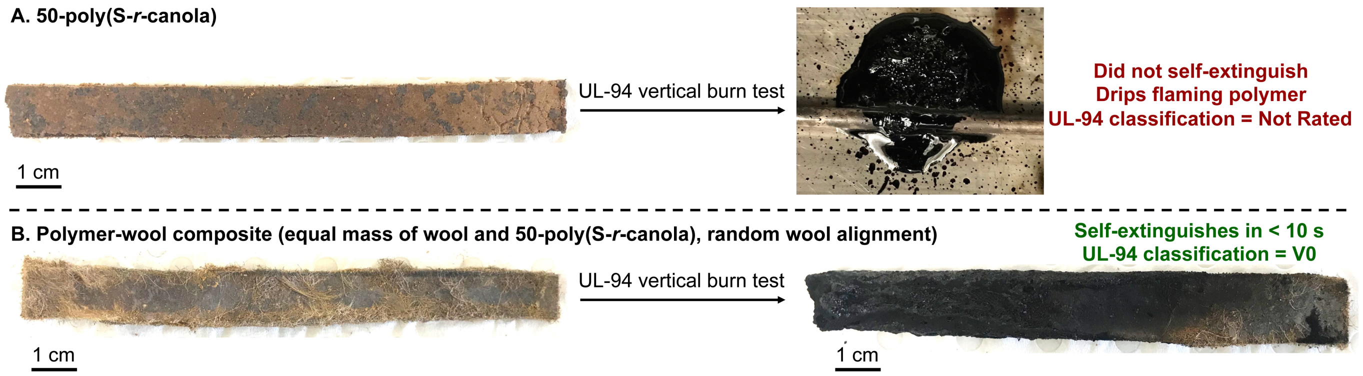


Figure 6. A. Strip of 50-poly(S-*r*-canola) polymer before and after the UL-94 vertical burn test. The polymer specimen did not self-extinguish and dripped flaming polymer. The black residue shown is the product that drips from the burning polymer. B. The polymer-wool composite before and after the UL-94 vertical burn test. The composite self-extinguished in less than 10 seconds in all five tests, resulting in a V0 classification.

For materials under consideration for insulation or construction, it is important to evaluate flammability. Accordingly, UL-94 vertical burn tests were conducted to evaluate the flammability of the 50-poly(S-*r*-canola) polymer mat and the corresponding wool-polymer mat according to ASTM D3801-19. The sample dimensions used in this test were 125 x 13 x 3.0 mm. The mats were cut into the dimensions using scissors and 5 tests were performed for each sample (an average of the 5 tests is represented in Table 1). The UL-94 vertical burn test evaluates the self-extinguishing performance of samples. The samples are each ignited for 10 seconds, the after flame is then recorded (the time required for samples to self-extinguish). This process is completed a total of three times. These times are denoted as t_1, t_2, and t_3.

In this testing method there are three flammability classifications, V0-V2, and a fourth 'Not Rated' classification, indicating that the sample does not possess any self-extinguishing properties. To achieve a V0 rating, both t_1 and t_2 are required to be below 10 seconds. The UL-94 rating for all samples is presented in Table 1. For the neat polymer the poorest classification of 'no rating' was found as the first flame does not extinguish and the sample burns to the clamp (Figure 6 and Video S1). The sample also dripped flaming polymer, suggesting that the material, once ignited, would likely spread fire. In contrast, the addition of wool to the polymer imparted significant flame retardant properties, with a V0 classification achieved in all 5 samples. It is important to note that none of the samples displayed any dripping (Video S2). The polymer-wool composite samples after UL-94 evaluation retained a significant amount of structural integrity (Figure 6). The rapid formation of a char at the material's surface is obvious, and likely a contributing factor to the suppression of flammability. Wool is known to be flame resistant due to its high ignition temperature, which is attributed to high moisture regain and high nitrogen content of the wool fibers.[31] But, while wool is known to be flame resistant, it was not clear at the outset of this study that wool's flame resistance could be conferred to the polymer-wool composite. The results in Figure 6 confirm that imparting flame resistance to the sulfur polymer is indeed possible.

Table 1. UL-94 ratings of 50-poly(S-r-canola) and wool-polymer composite samples. T_1 and T_2 refer to the average time to self-extinguish after the first and second ignition. BC = burn to the clamp. NR = no rating.

Samples	T_1 (s)	T_2 (s)	UL-94 rating	Dripping
50-poly(S-r-canola)	BC	-	NR	Yes
Wool-polymer composite	2.4	1.4	V0	No

To corroborate the observations made in the UL-94 test, thermogravimetric analysis (TGA) was carried out in both oxidative and non-oxidative environments. For air atmospheres, a flow rate 60.0 mL min^{-1} was used, and for those performed in nitrogen, a flow rate of 40.0 mL min^{-1} was employed. Samples of ~5–10 mg were heated from 20 °C to 1000 °C at a constant heating rate of 10 °C min^{-1}. TGA was used to explore the decomposition of each of the samples. The comparison of TGA curves in nitrogen (Figure 7A) reveals an increase in char yield (15%) for the wool-polymer sample and interestingly an extended elongation of the char yield (450-800 °C) for the wool-polymer composite. This demonstrates that the introduction of wool to the polymer composite is able to impart a greater flame resistance, improving the composites ability to withstand higher temperatures. A similar trend is observed for the TGA curves in air (Figure 7B), with the wool composite samples showing a char yield in air (at ~500 °C) of approximately 35%, corresponding to the char of the wool present in the sample. This product is then oxidized to completion as the temperature is increased.

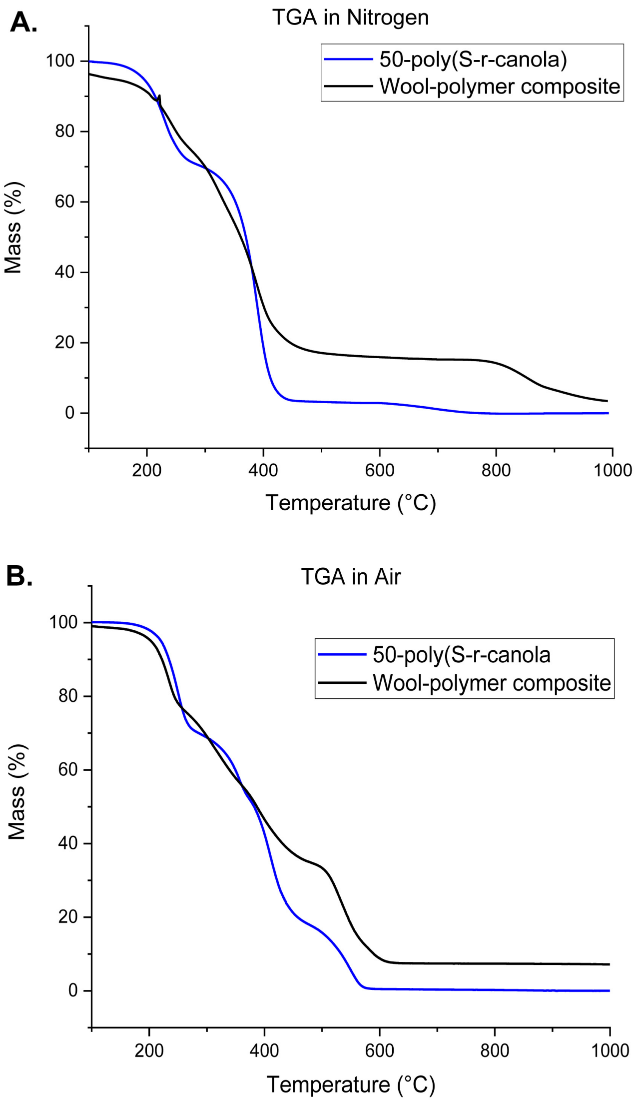


Figure 7. A. TGA of neat polymer and composite samples in inert (N_2) atmosphere. B. TGA of polymer and composite samples in air.

Conclusions and outlook

Composite materials were prepared using reactive compression molding of a mixture of renewable wool fibers and a polymer made by inverse vulcanization. The key polymer material was made from canola oil (a renewable feedstock) and sulfur (a highly abundant material and a byproduct of petroleum refining). The polymer synthesis is atom economical and the sulfur serves multiple roles as monomer, solvent, and initiator. There is also minimal waste generated in the composite preparation. The powdered polymer conveniently holds static charge, which facilitates its binding and coating of the wool fibers and the formation of the composite mat after hot pressing. The wool filler imparted tensile strength to the composite, with tensile modulus improved 10-fold for some samples. The composite was found to be an effective thermal insulator and wool conferred excellent flame resistance to the composite. The promising mechanical and insulation properties of this composite bodes well for further exploration in energy saving insulation in our built environment. Another important consideration with respect to green chemistry and sustainability is the design of a full life cycle for new materials. To that end, we are currently evaluating the long-term (bio)degradation of these materials. Our goal is to provide useful energy-saving insulation from sustainable feedstocks, and a process for degrading these materials in a safe and responsible fashion at the end of their life.

Conflicts of interest

J.M.C. is an inventor on patent applications covering the synthesis and processing of sulfur polymers, including the polysulfide featured in this study (WO2017181217 and AU2020901215). These patents are assigned to Clean Earth Technologies.

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Insulating composites made from sulfur, canola oil, and wool

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General considerations

Wool: Wool was obtained directly after shearing a 1.5 year old male Border Leicester sheep. The property of the farm is managed to organic standards.

Hot press: All reactive compression molding experiments were performed in the S15 Devil Press 10-ton hydraulic heated press.

Optical microscopy: Optical microscope images were acquired using a Witec alpha300R microscope with a 20X magnification objective. The working distance and numerical aperture of the objective used are 4 mm and 0.4 respectively.

SEM and EDX: All Scanning Electron Microscopy (SEM) images were obtained using a FEI F50 Inspect system. The corresponding EDS spectra were obtained using an EDAX Octane Pro detector. All samples were coated with 5 nm of platinum before imaging.

Raman Spectroscopy: Raman spectra were acquired using a Witec alpha300R Raman microscope at an excitation laser wavelength of 532 nm with a 20X objective (numerical aperture 0.40). Typical integration times for the Raman spectra were between 5-30 s for 2-3 accumulations.

Dynamic Mechanical Analysis (DMA): All tensile strength measurements performed using the tension clamps on the Dynamic Mechanical Analyser Q800. Measurements were carried out in triplicate at 20 °C, with the ramp displacement of 1000.00 μm/min.

Thermal Conductivity: All thermal conductivity measurements are performed using a TLS-100 thermal conductivity meter. They ensure complete contact with the material; the premium ceramic polysynthetic thermal compound was added to the probe before measurement, in accordance to the manufacture’s operating guidelines.

Infrared imaging: All thermal imaging was performed using a FLIR Ex series camera with the temperature scale range set to 18 – 65 °C in Thermal imaging mode. Auto Hot Spot mode was used to ensure the hottest area on the substrate was recorded.

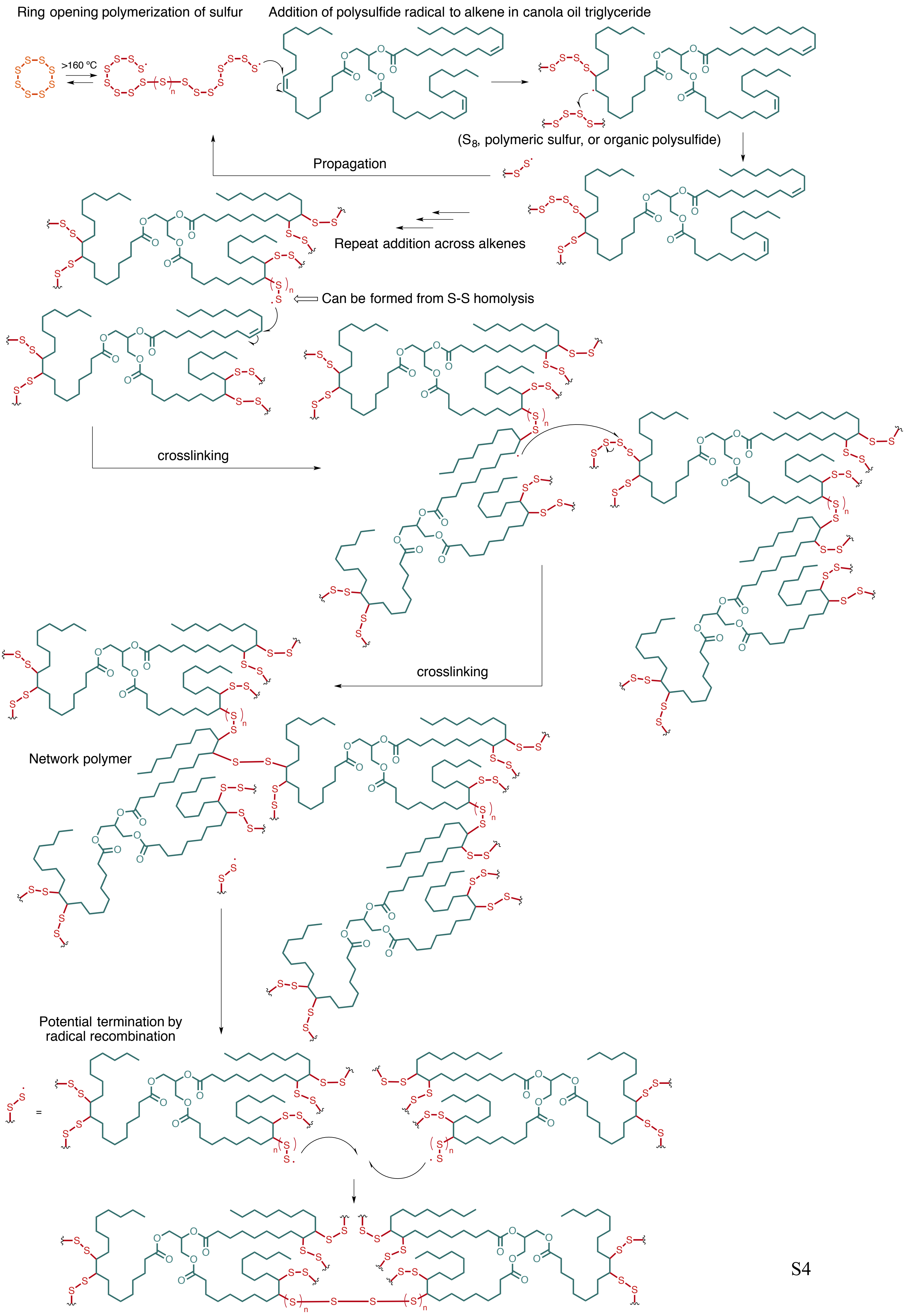
Hydraulic press large scales

Reactive compression moulding experiments for the large scales were performed in JL-15A Ezylif 15-ton hydraulic heated press and DK 16 Digital Knight press.

50-poly(S-*r*-canola) synthesis

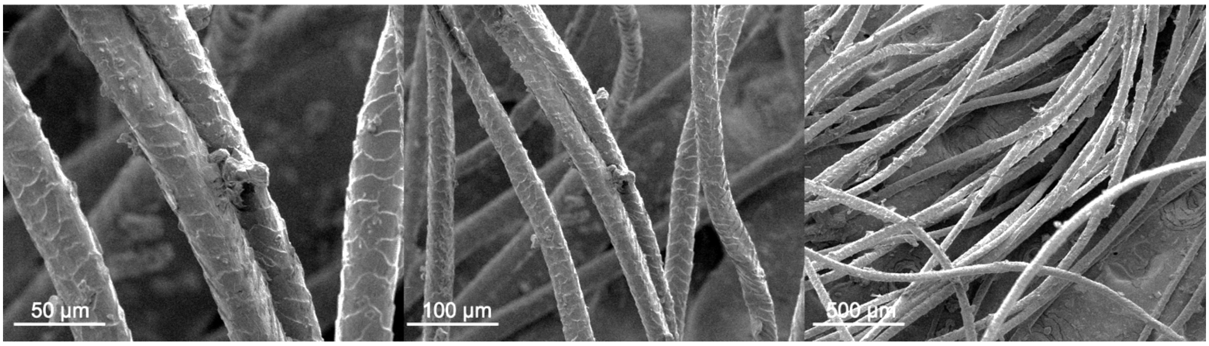
As shown in a previous study by our lab, inverse vulcanisation of sulfur and canola oil (50% sulfur by mass) results in a rubber-like material. The synthesis process used in this study is based on the protocol reported by Worthington et al.[1] First, in a fume hood, pristine canola oil was heated in a stainless-steel vessel to 175 °C. The temperature of the oil will be monitored and controlled directly with a temperature probe. Mixing was controlled and maintained at 90 rpm using an overhead stirrer. Next, 450 g of sulfur was added carefully through a funnel at such a rate that the internal temperature did not fall below 159 °C. This process took approximately 10-15 minutes. When the sulfur was added, the reaction mixture became two transparent liquid phases. The bottom layer (dark orange to red) was molten sulfur and sulfur pre-polymer formed by ring opening polymerisation, while the top layer (light yellow) was the canola oil. The mixture continues to react until it becomes opaque and the two layers converge to an apparent single phase. At this time, sodium chloride (2100 g) was then added through the funnel. The sodium chloride serves as a porogen to increase the surface area and lower the density of the polymer. The addition of the sodium chloride was added such that the reaction temperature remained above 159 °C. After the addition of salt, the mixture was typically an orange and relatively free-flowing slurry, and it thickend and darkened to brown colour. When the viscosity increases and the overhead stirrer registers a torque of 40 N•cm, the reaction was stopped. The reaction vessel was then removed from the hotplate and allowed to cool to room temperature. The polymer was removed using using a metal spatula and processed in a mechanical grinder to a particles size of 0.5 to 3 mm in size. To remove the salt, the polymer was washed thoroughly with 17 L of deionised water in a 20 L bucket with overhead stirring (30 minutes). Finally, the polymer was isolated by gravity filtration through a sieve (0.5 mm). This process was repeated three times to ensure removal of salt. The polymer was then dried in a fume hood. All spectroscopic (^{1}H NMR, IR) and thermal analysis (TGA and DSC) were consistent with those previously reported from our lab.[1]

Elemental sulfur (left), canola Oil (middle) and 50-poly(S-*r*-canola)

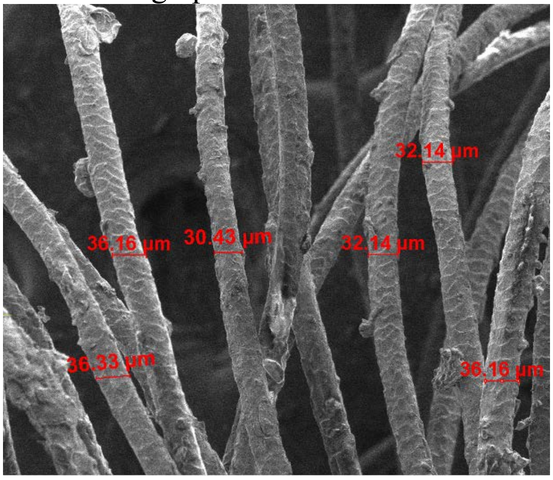

Ring opening polymerization of sulfur
Addition of polysulfide radical to alkene in canola oil triglyceride
>160 °C
(S8, polymeric sulfur, or organic polysulfide)
Propagation
Repeat addition across alkenes
Can be formed from S-S homolysis
crosslinking
crosslinking
Network polymer
Potential termination by radical recombination

Images of raw wool

SEM



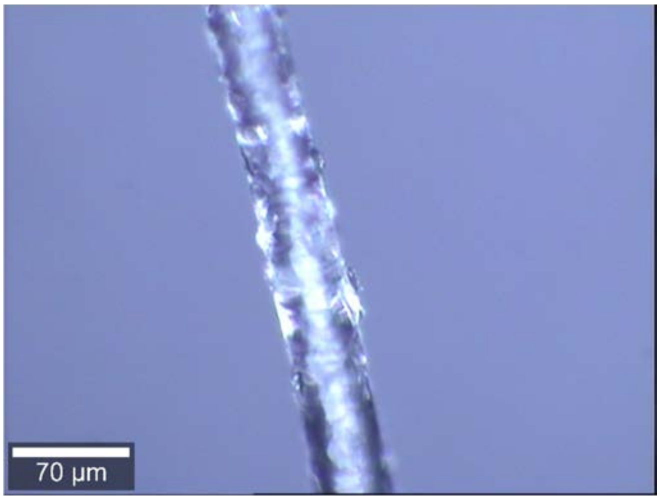
SEM micrographs of raw wool fibers at three different magnifications



SEM analysis measuring the thickness of raw wool fibers.

The average wool fiber thickness was determined by SEM to be 34 ± 3 µm.

Optical microscopic image of wool fiber used in the study:



Reactive compression molding of 50-poly(S-*r*-canola)

The 50-poly(S-*r*-canola) (15.0 g) was placed between two 10 × 10 cm poly(tetrafluoroethylene) (PTFE) sheets in an aluminum mold, as shown in the figure below. The mold lid was added and placed in the press, which was pre-heated to 100 °C. The heated plates of the press were brought into contact with the mold and the system was left to heat and equilibrate at 100 °C. Once the temperature reached 100 °C, the pressure was increased 20 MPa using the hydraulic jack. This pressure was applied for 15 minutes after which the mold was removed from the press and the resulting 50-poly(S-*r*-canola) mat was separated from the Teflon sheets. The final product after reactive compression molding is shown below.

Aluminum mold (10 × 10 cm) used in all reactive compression molding experiments

Assembled mold in S15 Devil Press

Image showing 50-poly(S-*r*-canola) powder before (left) and after (right) undergoing reactive compression molding to form a mat

SEM and EDX analysis of mat formed by reactive compression molding of 50-poly(S-*r*-canola)

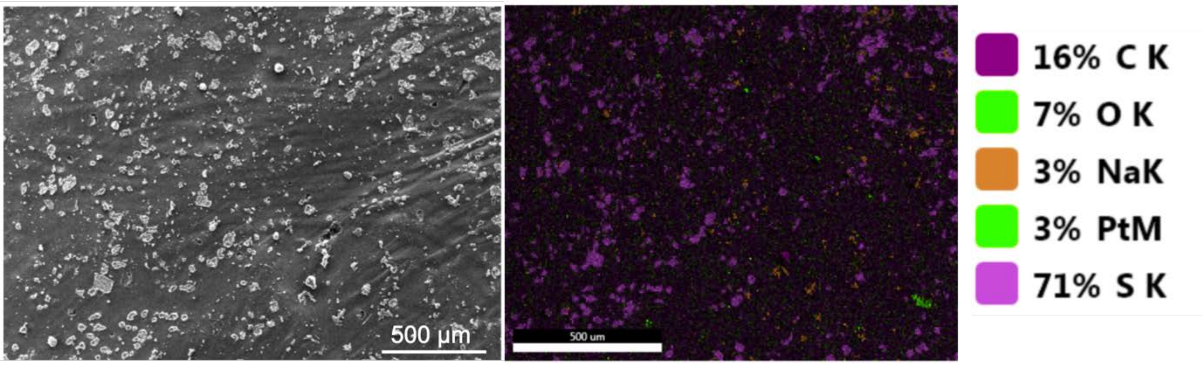


Image showing SEM of 50-poly(S-*r*-canola) mat (left) and EDX (right). The particles are free elemental sulfur extruded to the surface of the polymer mat, as previously described in our study of reactive compression molding of 50-poly(S-*r*-canola).[2]

Hot pressing raw wool

A 10 × 10 cm sheet of PTFE was placed into the aluminum mold, 10.0 g of raw wool was added on top of the PTFE sheet. The wool length was ~ 11 cm. A second PTFE sheet was placed above the wool, the lid was added, and the mold was placed in the press, which was preheated to 100 °C. The heat plates were pressed until they were touching each other and the system was left for 2 minutes to equilibrate at 100 °C before the pressure was increased to 40 MPa. This pressure was applied for 20 minutes. After this time, the pressure was released, and the mold was removed from the press. The resulting wool mat was removed from the mold and separated from the PTFE sheets. The final product is shown below.

Image showing the raw wool before (left) and after (right) hot pressing (100 °C, 40 MPa, 20 minutes).

SEM/EDX analysis of mat formed by hot pressing wool

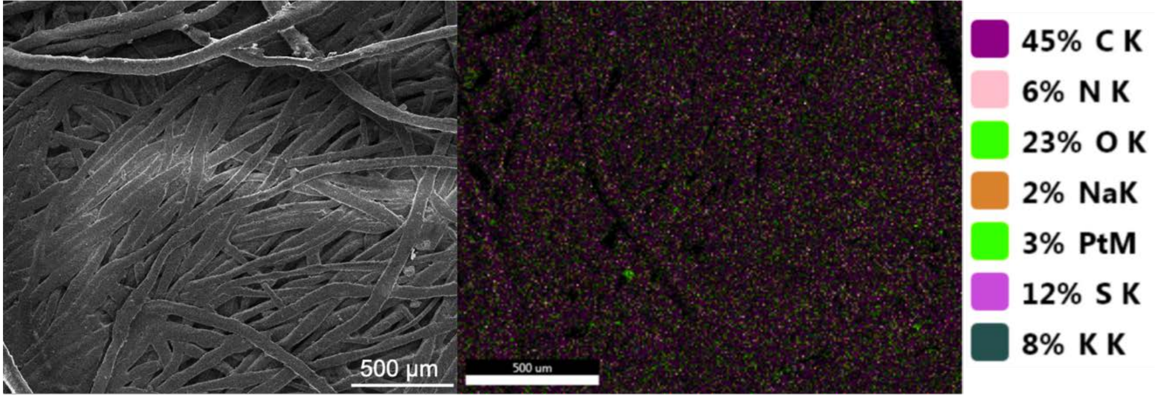
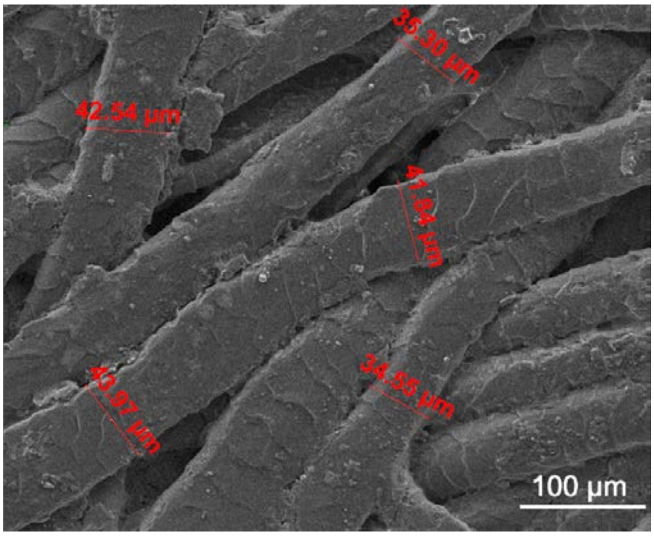


Image showing SEM micrograph (left), and EDX image (right) of wool fibers after hot pressing.



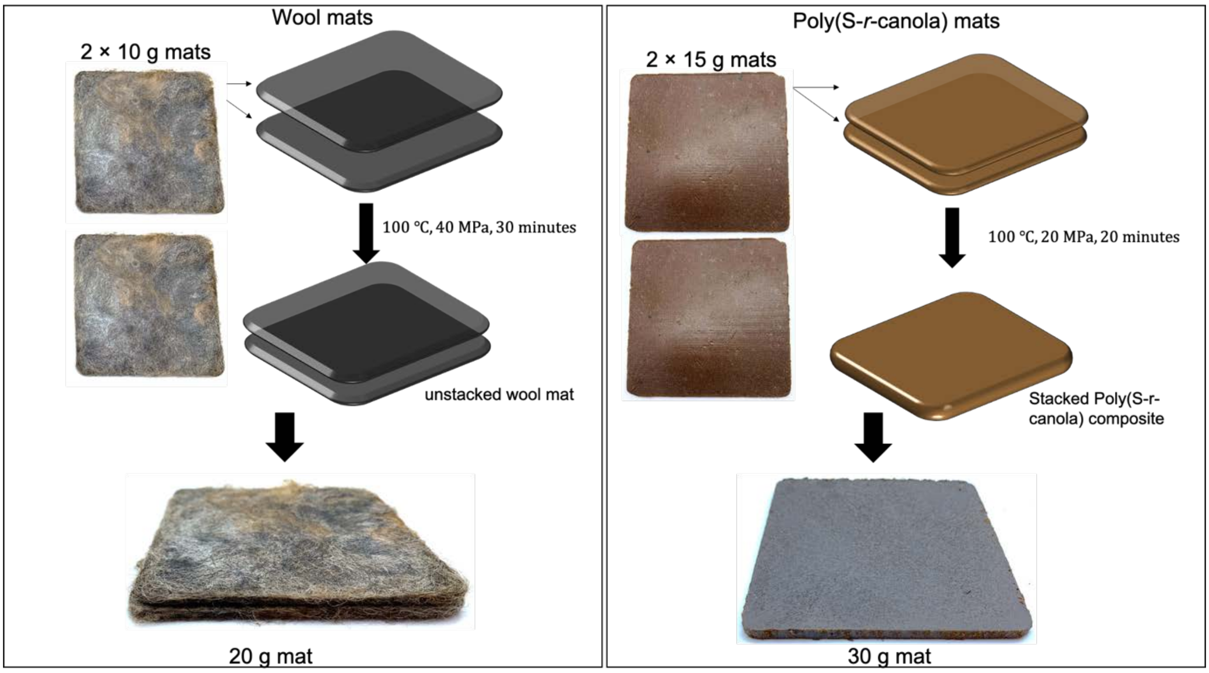
SEM micrograph of wool mat showing the effect of hot pressing on the width of the fibers. This micrograph also shows that the fibers are still intact after hot pressing, but flattened to a width of 40 ± 4 µm, based on the sample of widths indicated in the micrograph above.

Additive compression molding of preformed wool and 50-poly(S-*r*-canola) mats

A 10 × 10 cm PTFE sheet was placed in the aluminum mold and 10.00 g of raw wool was added. The second PTFE sheet was placed above the wool. The mold lid was added, and the mold was placed in the press, which was heated to 100 °C. The heated plates were brought in contact with the mold to preheat the system to 100 ºC. When the temperature was stable at 100 °C, the pressure was increased to 40 MPa. This pressure was applied for 20 minutes. After 20 minutes the pressure was released, and the mold was removed from the press. The resulting wool mat was removed and separated from the PTFE sheets. This process was repeated to form a second wool mat. Next a 10 × 10 cm PTFE sheet was placed in the aluminum mold, followed by both wool mats. A PTFE sheet was then placed on top of the two preformed wool mats, the lid was added, and the mold was placed in the press and processed at 100 °C and 40 MPa for 30 minutes. After this time the pressure was released, and the mold removed from the press. The resulting wool mats were removed and were not bound together (see figure below).

This entire process was repeated under with the preformed 50-poly(S-*r*-canola) mats (15 g for each preformed polymer mat). The press was used to bond the two mats together by reactive compression molding (100 °C, 20 MPa, 20 minutes). The two preformed 50-poly(S-*r*-canola) mats bonded to form a single mat after processing under the same conditions (see image below).

Additive manufacture

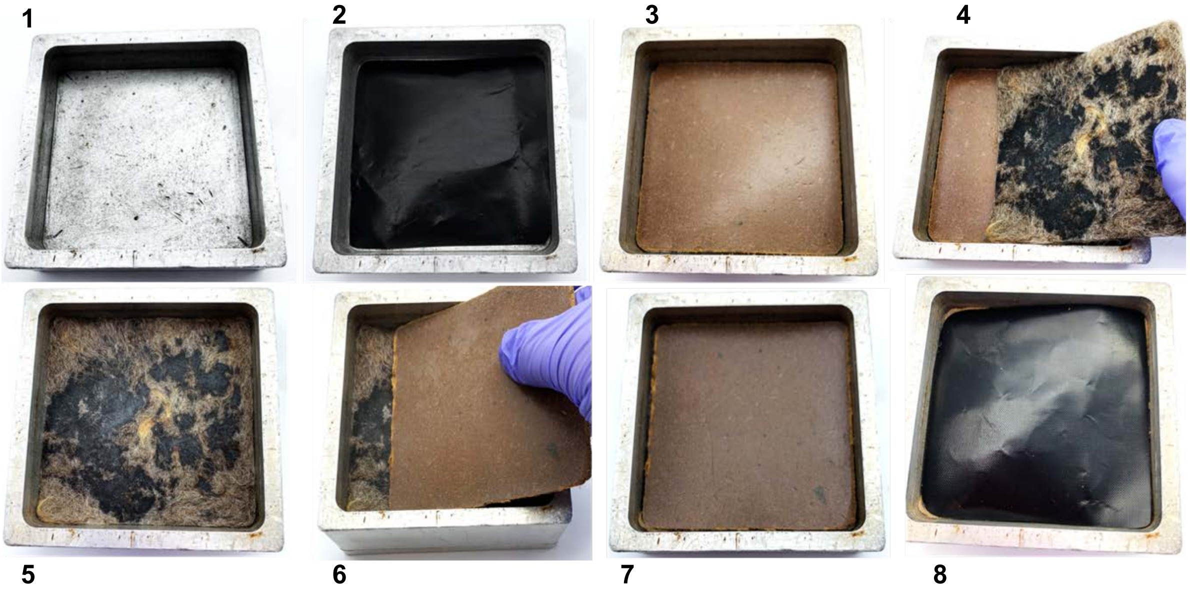


Reactive compression molding of wool mat between 50-poly(S-*r*-canola) mats

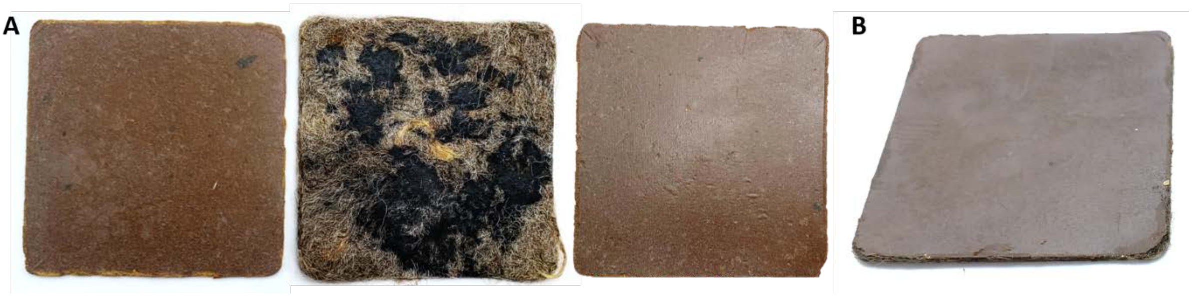
50-poly(S-*r*-canola) mats: A 10 × 10 cm sheet of PTFE sheet was placed in the aluminum mold, followed by 5.0 g of 50-poly(S-*r*-canola). Then the second PTFE sheet was placed above the polymer. The mold lid was added, and the mold was placed in the press and heated to 100 °C. Once the mold was equilibrated to 100 °C, the pressure was increased to 30 MPa and applied for 15 minutes. After this time, the pressure was released, and the resulting 50-poly(S-*r*-canola) mat from removed from the mold and PTFE sheets. This procedure was repeated to form a second 50-poly(S-*r*-canola) mat.

Wool mat: Raw wool (10.0 g) was added between two 10 × 10 cm PTFE sheets placed in the aluminum mold. The length of the used wool was ~11 cm. The mold lid was added, and the mold was placed in the press and heated to 100 °C. Then, the pressure was increased to 40 MPa and applied for 20 minutes. After this time the pressure was released, and the resulting wool mat was removed from the mold and separated from the PTFE sheets.

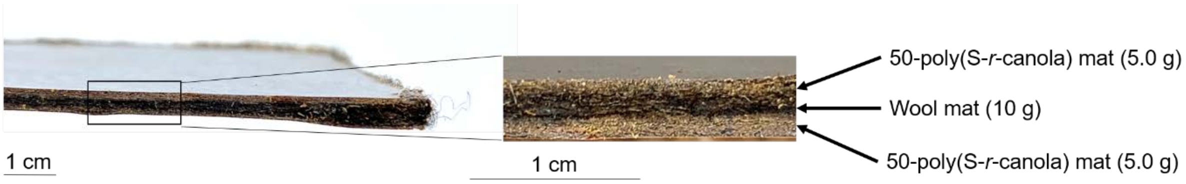
Composite mat: First, a 10 × 10 cm of PTFE sheet was placed in the aluminum mold, followed by the first 50-poly(S-*r*-canola) mat, the wool mat, the second 50-poly(S-*r*-canola) mat, and another PTFE sheet (see image below). The lid of the mold was added, and the mold was pre-heated to 100 ºC in the press. After reaching 100 ºC, the pressure was increased to 40 MPa for 30 minutes. After this time, the composite mat was removed from the mold and separated from the PTFE sheets (see next page).



1. Bottom of mold 2. Bottom PTFE sheet 3. Bottom polymer mat 4. Wool mat added 5. Wool mat in place 6. Top polymer mat added 7. Top polymer mat in place 8. Top PTFE sheet

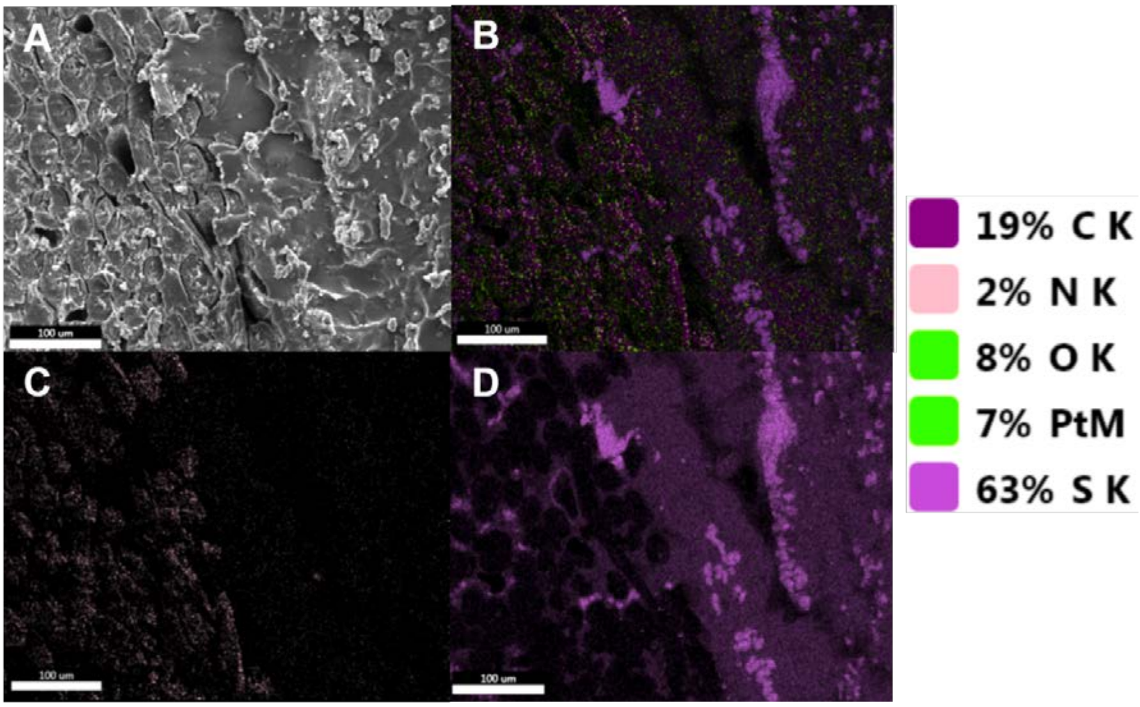


A) Two 50-poly(S-*r*-canola) mats and one wool mat used in the preparation of the composite mat, **B)** Composite mat with one wool mat fixed between two 50-poly(S-*r*-canola) mats using reactive compression molding.

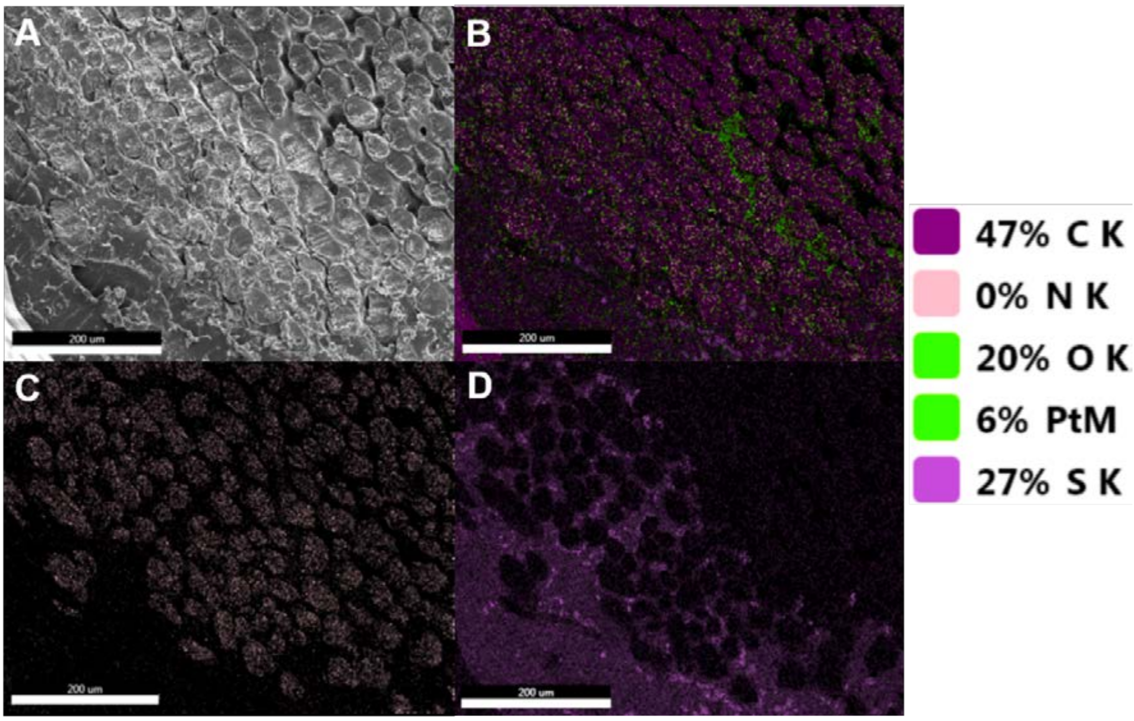


The above process was repeated under the same conditions using the same mass of raw wool that hadn't previously been pressed into a mat. The raw wool fibre was sandwiched in between two poly(S-*r*-canola) mats forming a composite mat (see below and following page).

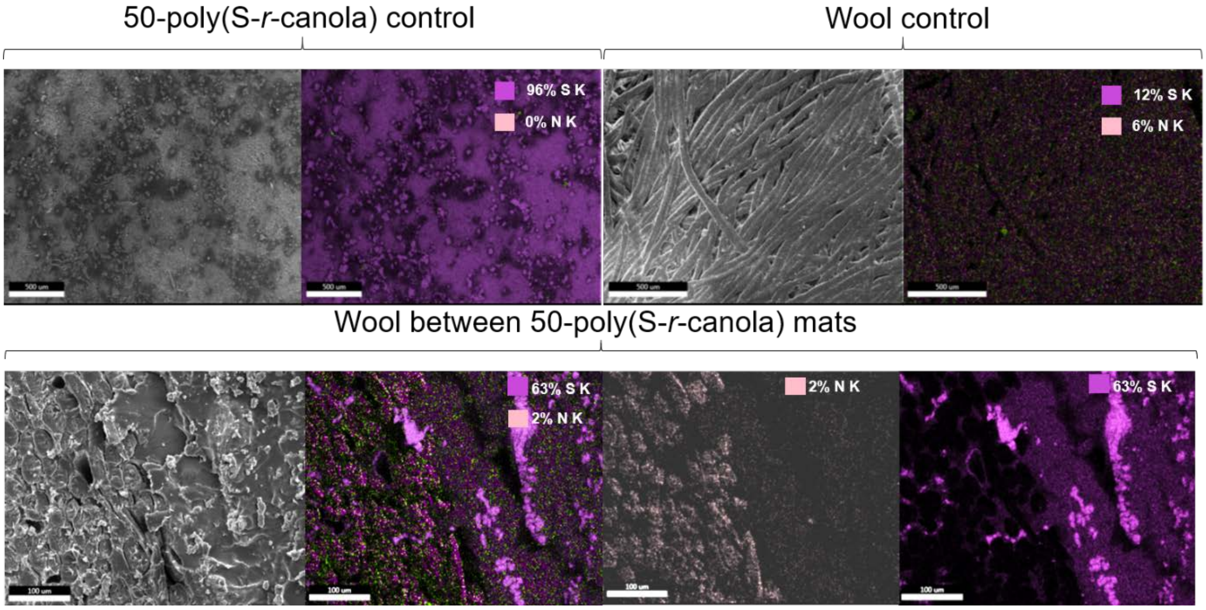
SEM/EDX analysis of mat formed by additive reactive compression moulding of wool sheet in between two preformed poly(S-*r*-canola) sheets and free raw wool in between two preformed poly(S-*r*-canola) sheets respectively



SEM image of area mapped using EDX. **B)** EDX mapping of image A with Carbon, Nitrogen, Oxygen, Platinum and Sulfur highlighted. **C)** EDX mapping of Nitrogen over surface of sample in A. **D)** EDX mapping of sulfur over surface of sample in A.



A) SEM image of area mapped using EDX. **B)** EDX mapping of image A with Carbon, Nitrogen, Oxygen, Platinum and Sulfur highlighted. **C)** EDX mapping of Nitrogen over surface of sample in A. **D)** EDX mapping of sulfur over surface of sample in A.

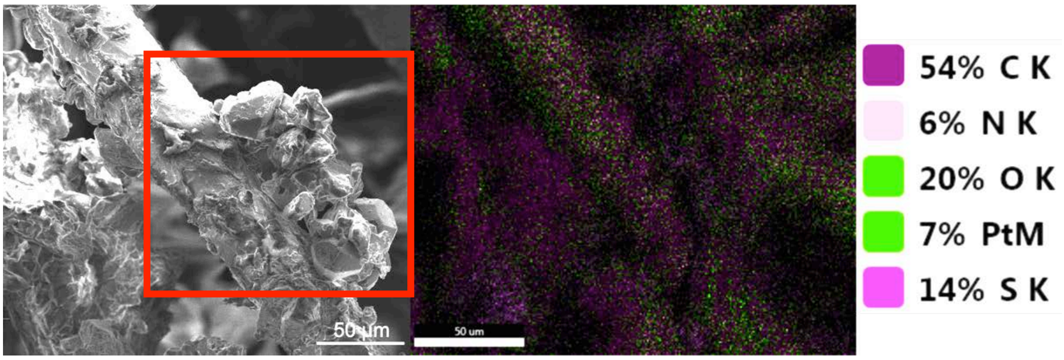


cross section EDX showing polymer encasing the wool

Powdered 50-poly(S-*r*-canola) adheres to wool fibers

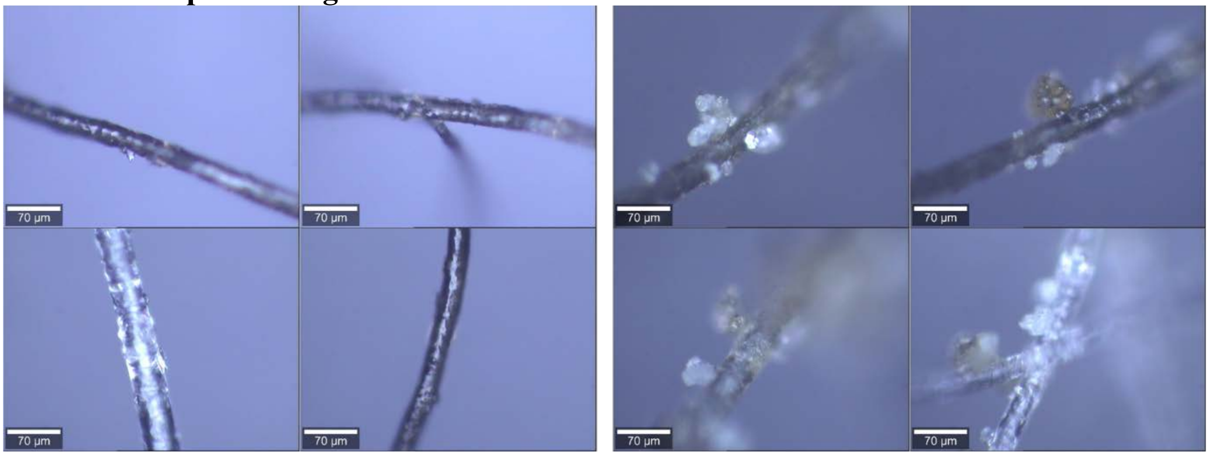
10.0 g of raw wool was mixed with 10.0 g of 50-poly(S-*r*-canola) were placed in a plastic container and then mixed by hand or by inverting the closed container several times. The static charge causes the polymer to adhere to the wool. This feature helps make a relatively uniform coating of the polymer on the wool.

A) 50-Poly(S-*r*-canola), **B)** Raw wool, **C)** Raw wool and 50-poly(S-*r*-canola) mixture

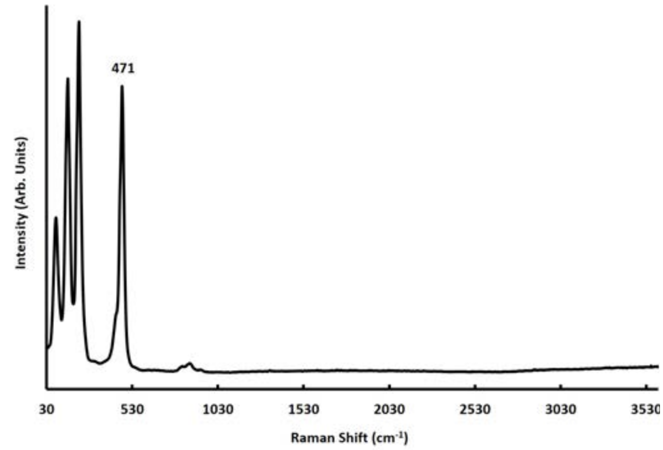


SEM micrograph (left) and EDX image (right) of the mixed 50-poly(S-*r*-canola)/wool mixture. The red box shows a polymer particle adhering to the wool fiber.

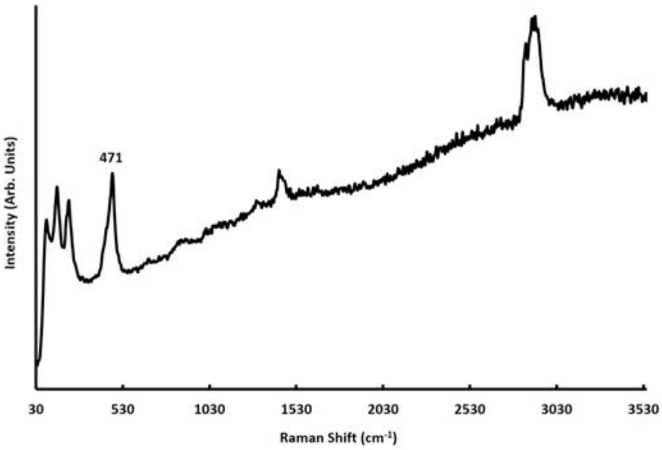
Raman and optical images here



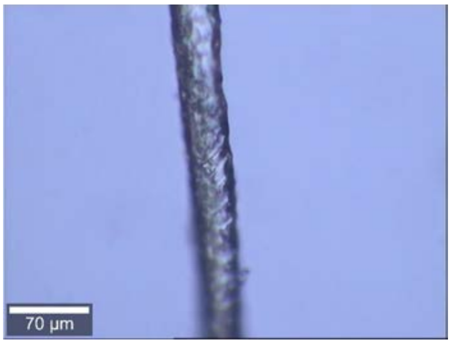
Raman spectra of S_8:

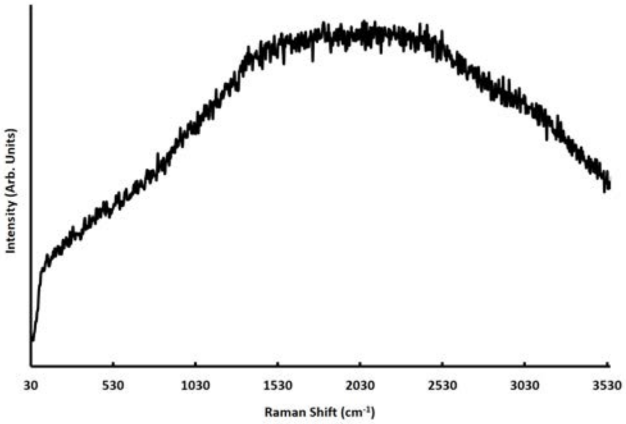


Raman spectra of 50-poly(S-*r*-canola):

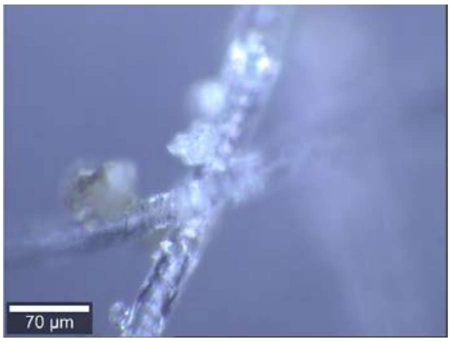


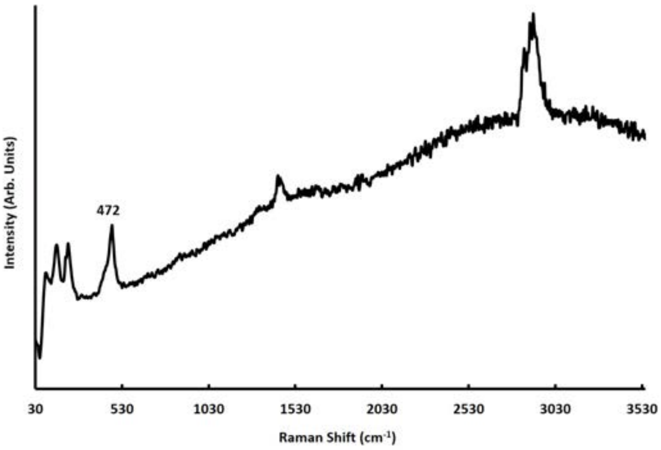
Optical image and Raman spectra of wool:





Optical image and Raman spectra of polymer on wool:



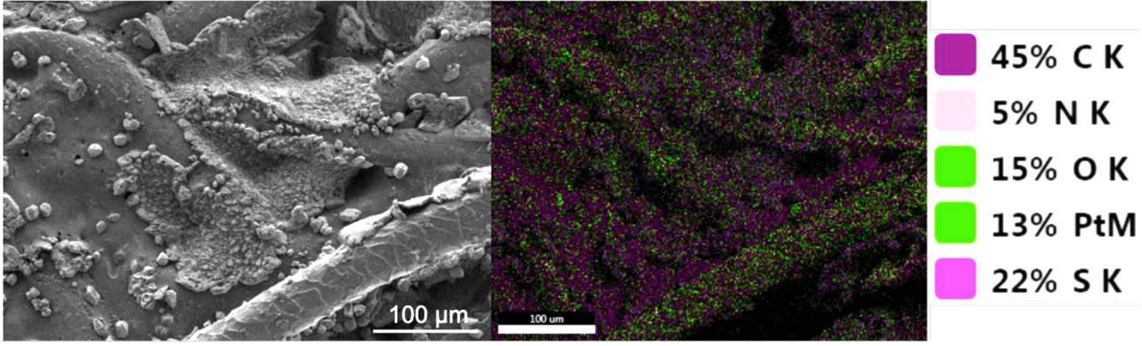


Reactive compression molding of 50-poly(S-*r*-canola) and wool mixture

Raw wool (10.0 g) was mixed with 50-poly(S-*r*-canola) (10.0 g) as described above to provide a mixture in which the polymer adheres to the wool fibers. The wool fiber length was ~ 11 cm. A 10 × 10 cm of PTFE sheet was placed in the aluminum mold, followed by the polymer and wool mixture. The second PTFE sheet was placed above the mixture and the mold lid was added. The mold was placed in the heated press and heated to 100 °C. When the temperature of the system reached 100 °C, the pressure was increased to 30 MPa and applied for 20 minutes. After this time, the pressure was released and the mold was removed from the press. The resulting composite mat was then removed from the mold. Images of the process are shown below:

Images demonstrating the process used to prepare the mixed 50-poly(S-*r*-canola) / wool composite mat. The image on the far right shows the final mat formed using this process.

SEM and EDX analysis of polymer-wool mat



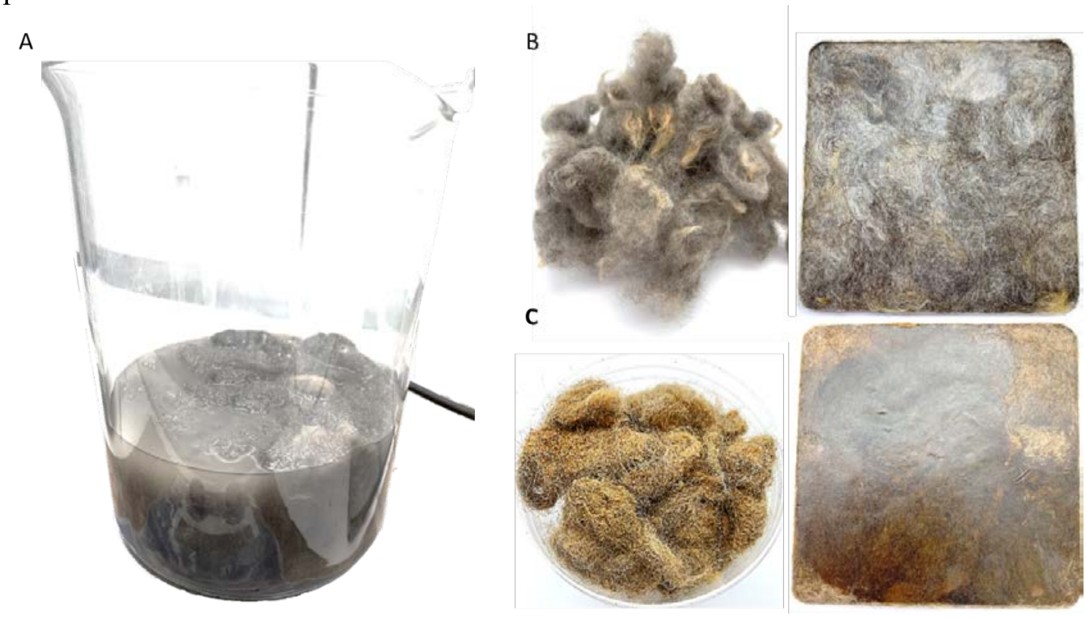
SEM (left) and EDX (right) micrographs of the polymer-wool composites. The wool fibers are embedded in the polymer matrix.

Reactive compression molding using washed wool

The wool was washed using dichloromethane to remove oils such as lanolin. In a 1 L beaker, 800 mL of dichloromethane was added, then 35.5 g of the raw wool added to the dichloromethane. A glass rod was used to stir the mixture every 5 minutes for a 30-minute wash period. After 30 minutes, the wool was then filtered through a Buchner funnel. This process was repeated using 600 mL of dichloromethane for a further hour. The wool was then filtered and left in a fume hood for 24 hours to dry. Wool weight recorded after drying to be 28.0 grams. This means the oily substance present made up 21% of the total raw wool weight.

Washed wool mat: A 10 × 10 cm sheet of PTFE was placed into the aluminum mold, followed by 10.0 g of the washed wool. A second PTFE sheet was placed on top of the wool, the lid was added, and the mold was placed in the press and heated to 100 °C. When the temperature reached 100 °C the pressure was increased to 40 MPa. This pressure was applied for 30 minutes. After this time, the pressure was released, and the resulting wool mat was removed from the mold and separated from the PTFE sheets. The wool mat still formed, indicating that the oils are not required for the wool to be hot pressed into a persistent shape. The final product is shown below.

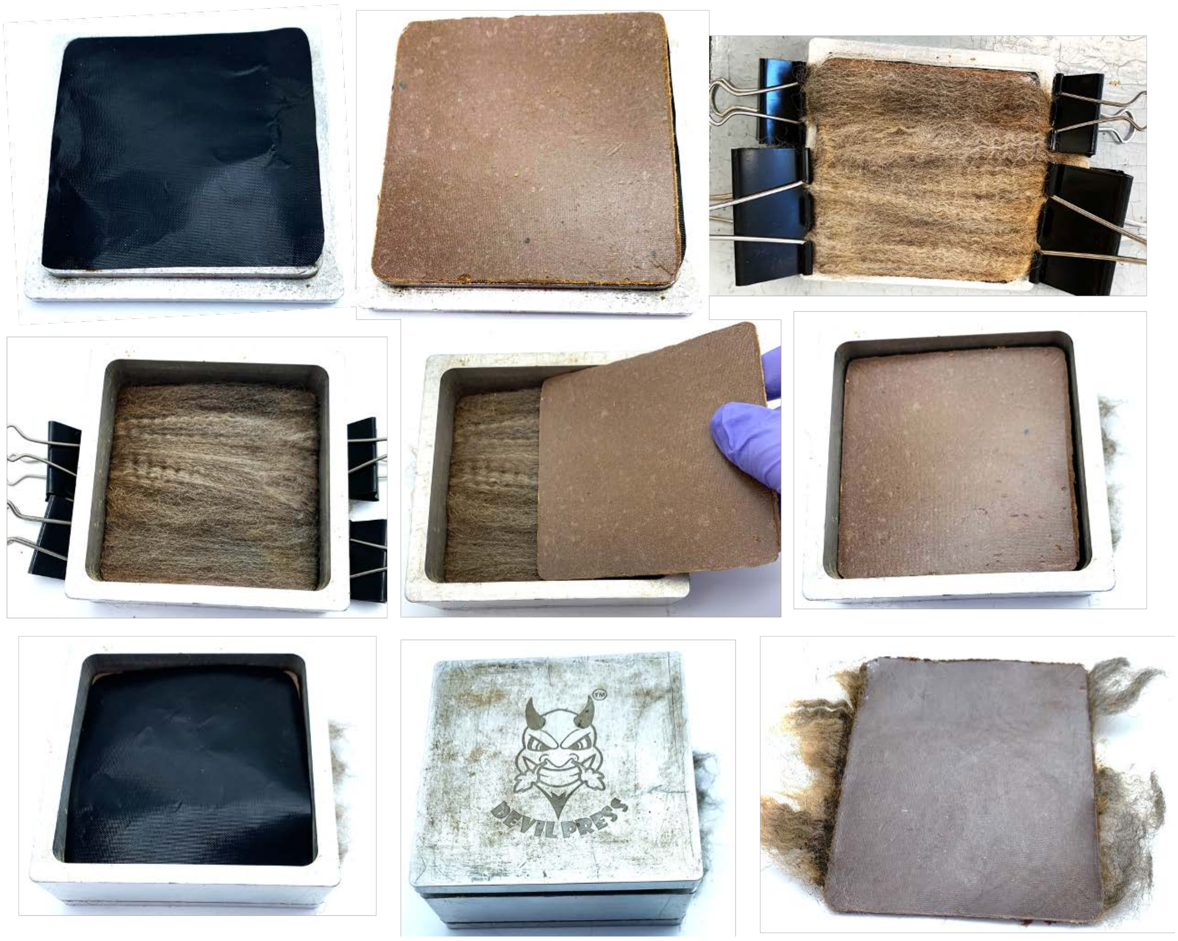
Composite mat made using 50-poly(S-*r*-canola) and washed wool: A 10 × 10 cm sheet of PTFE was placed into the aluminum mold followed by a mixture of 10.0 g of the washed wool and 10.0 g of 50-poly(S-*r*-canola) powder. A second PTFE sheet was placed above the mixture, the lid was added, and the mold was placed in the press and heated to 100 °C. When the temperature reached 100 °C the pressure was increased to 40 MPa. This pressure was applied for 30 minutes. After this time, the pressure was released, and the mold was removed from the press. The resulting composite mat was removed from the mold and separated from the PTFE sheets. The reactive compression molding proceeded as previously used with the raw wool, indicating that the wool oils are not required for composite formation. The final product is shown below.



A) Raw wool being washed in DCM, **B)** Washed and dried raw wool before (left) and after (right) hot pressing, **C)** Washed and dried raw wool mixed with 50-poly(S-*r*-canola) before (left) and after (right) reactive compression molding

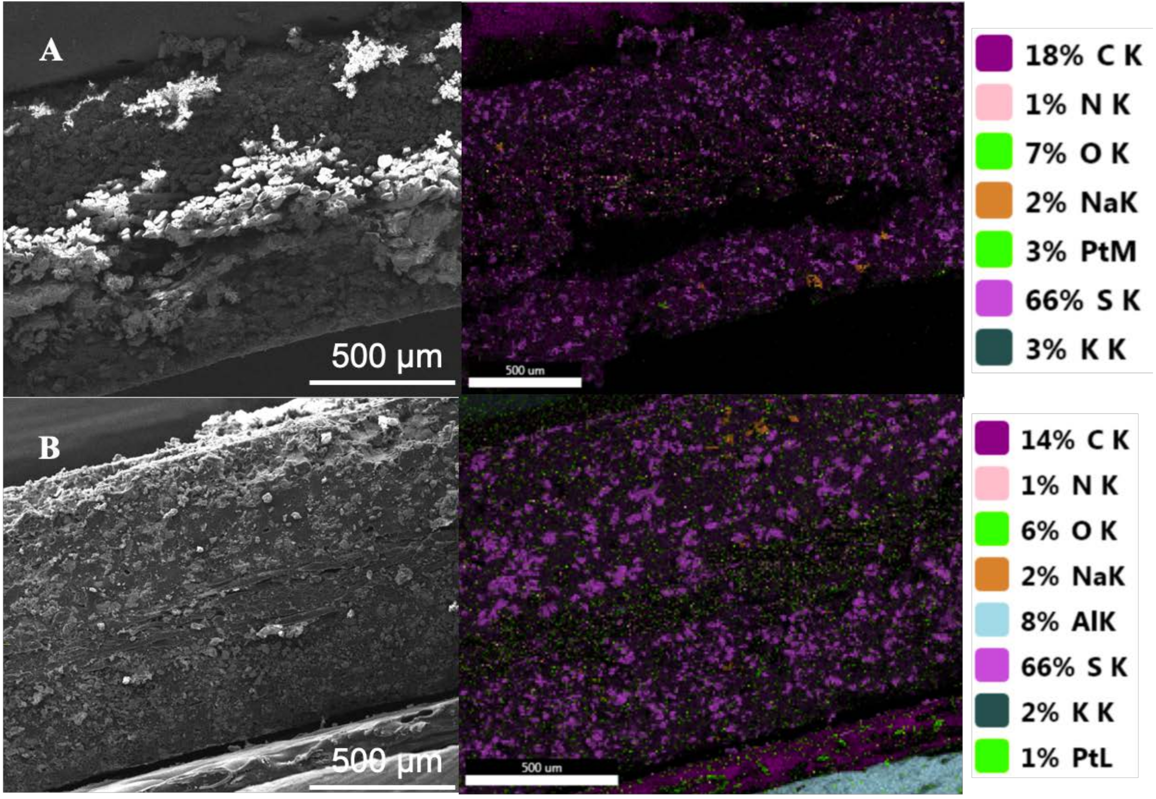
Reactive compression molding of 50-poly(S-*r*-canola) and raw wool with unidirectional fiber alignment

A 10 × 10 cm of PTFE sheet was placed on the base of the aluminum mold. Then a preformed 50-poly(S-*r*-canola) mat (5.0 g) was placed on the PTFE sheet. The wool (2.0 g, 11 cm) was aligned and clamped taut across the polymer mat. The second 50-poly(S-*r*-canola) mat was placed above the aligned wool followed by the second PTFE sheet. The lid of the mold was added before the entire mold was placed in the press and heated to 100 °C. When the temperature reached 100 °C, the pressure was increased to 40 MPa. This pressure was applied for 40 minutes. After this time the pressure was released, and the mold was removed from the press. The resulting composite with aligned wool fibers was removed from the mold and separated from the PTFE sheets. The entire process is depicted below.

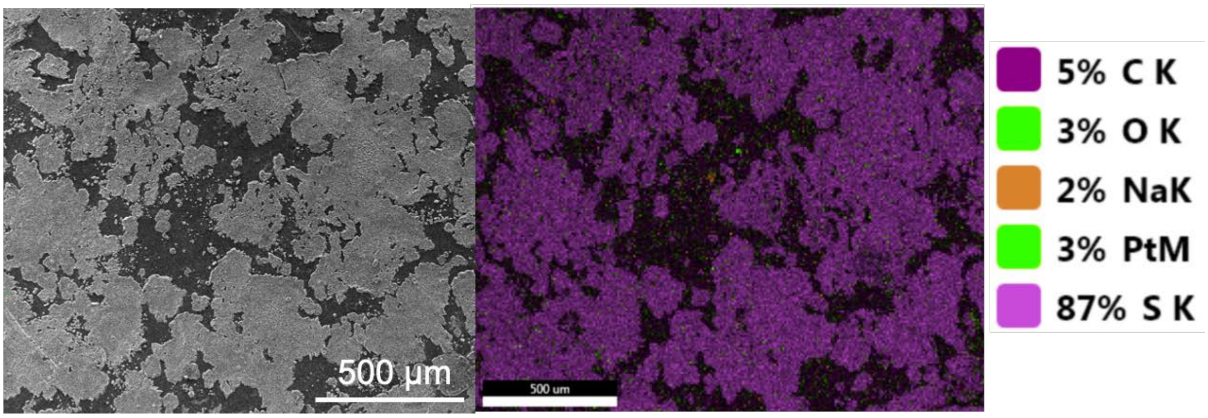


From left to right and top to bottom, images demonstrating the process used to prepare the composite mat with aligned wool fibers.

SEM and EDX analysis of composite mat with aligned wool fibers



A) SEM (left) and EDX (right) micrographs of a cross section of the composite mat with aligned wool fibers. The fibers are going into the page. **B)** SEM (left) and EDX (right) micrographs of a cross section of the composite mat with aligned wool fibers. The fibers are going across the page.



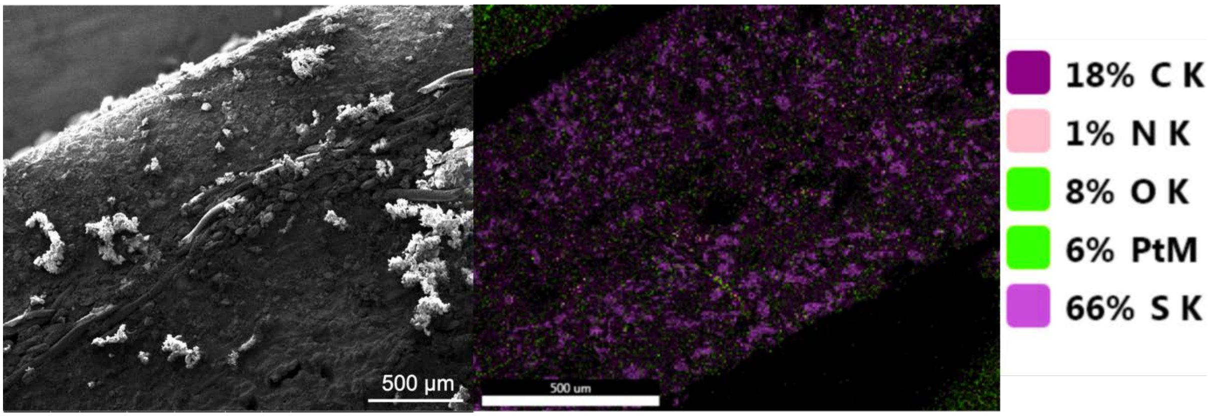
SEM (left) and EDX (right) micrographs of the surface of the 50-poly(S-*r*-canola) and wool composite with aligned fibers. Free sulfur is predominant on the surface of the mat.

Reactive compression molding of 50-poly(S-*r*-canola) and raw wool with dual fiber mats of orthogonal alignment

A 10 × 10 cm PTFE sheet was placed in the aluminum mold followed by one of the preformed 50-poly(S-*r*-canola) polymer mats (5.0 g prepared at 100 °C, 40 MPa, 20 minutes). Then 2.0 g of wool (11 cm length) was aligned and clamped taut across the polymer mat. A second PTFE sheet was added and the mold was closed and heated to 100 °C in the hot press. Once the temperature reached 100 °C, the pressure was increased to 30 MPa for 30 minutes. After this time the pressure was released, and the mat was removed to trim the excess fibers. The mat was returned to the mold and second batch of wool (2.0 g, 11 cm) was clamped taut across the mat, with fibers aligned orthogonal to the first batch of wool. A second 50-poly(S-*r*-canola) mat (5.0 g prepared at 100 °C, 40 MPa, 20 minutes) was added on top of the second lot of fibers followed by the second PTFE sheet. The mold lid was added then the mold was heated to 100 °C in the hot press. When the temperature reached 100 °C, the pressure was increased to 30 MPa. This pressure was applied for 30 minutes. After this time the pressure was released, and the resulting composite mat was removed from the mold. The entire process is depicted below.

From left to right and top to bottom, images demonstrating the process used to prepare the composite mat with dual wool mats with orthogonal fiber alignment

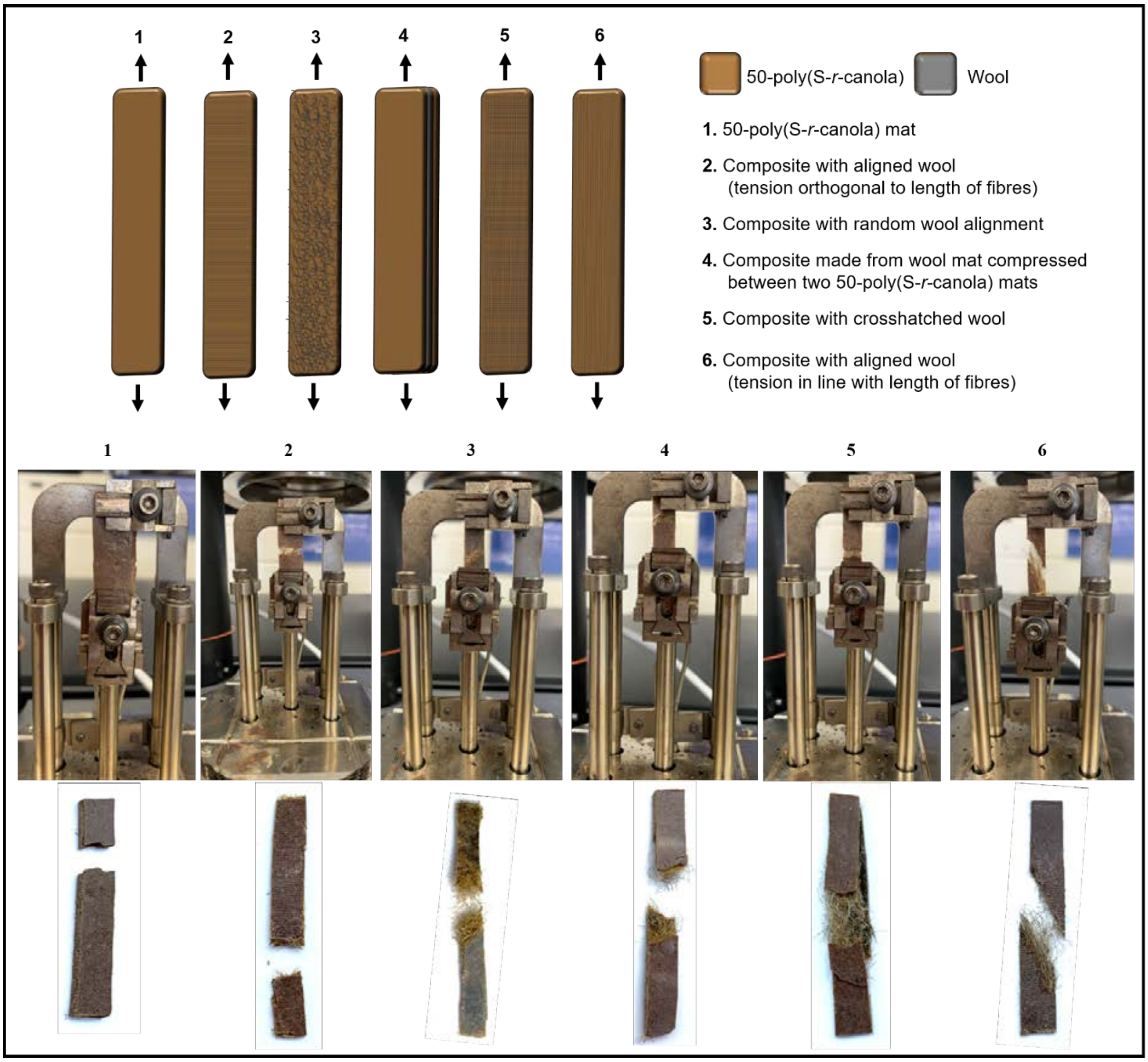
SEM and EDX of 50-poly(S-*r*-canola) and wool composite with dual wool mats of orthogonal fiber alignment



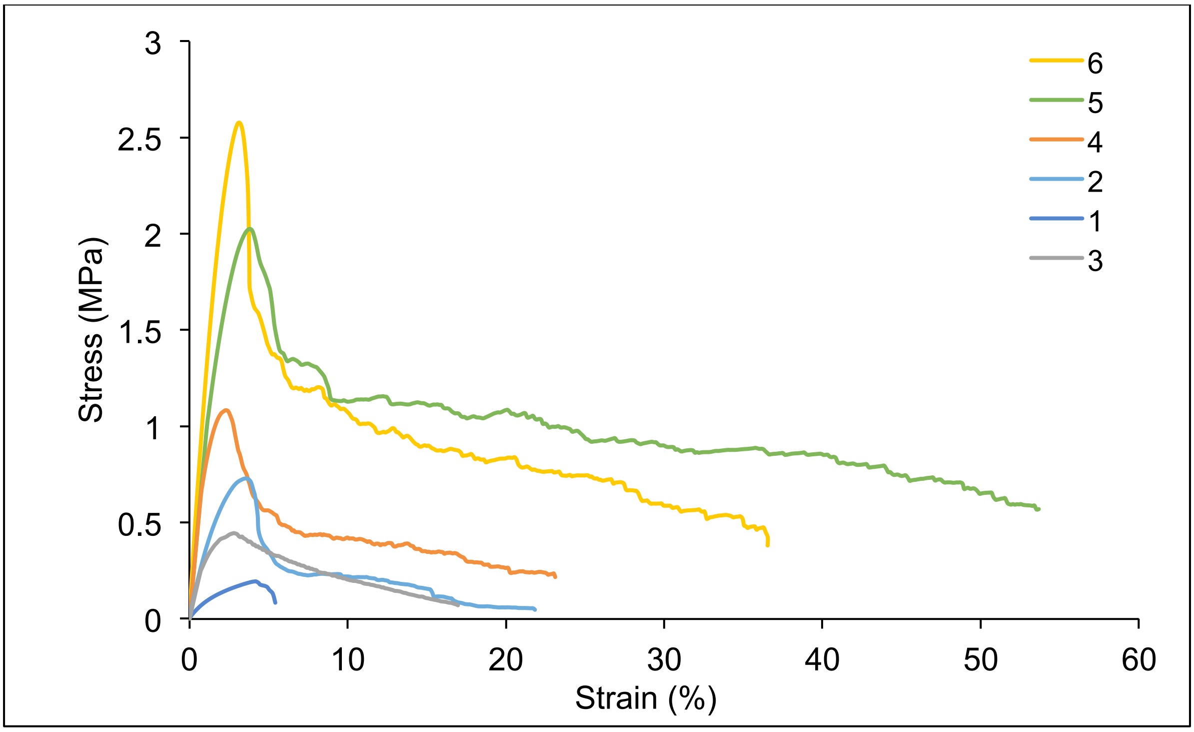
SEM (left) and EDX (right) micrographs of composite with dual wool mats of orthogonal fiber alignment

Tensile strength of composite samples using dynamic mechanical analysis

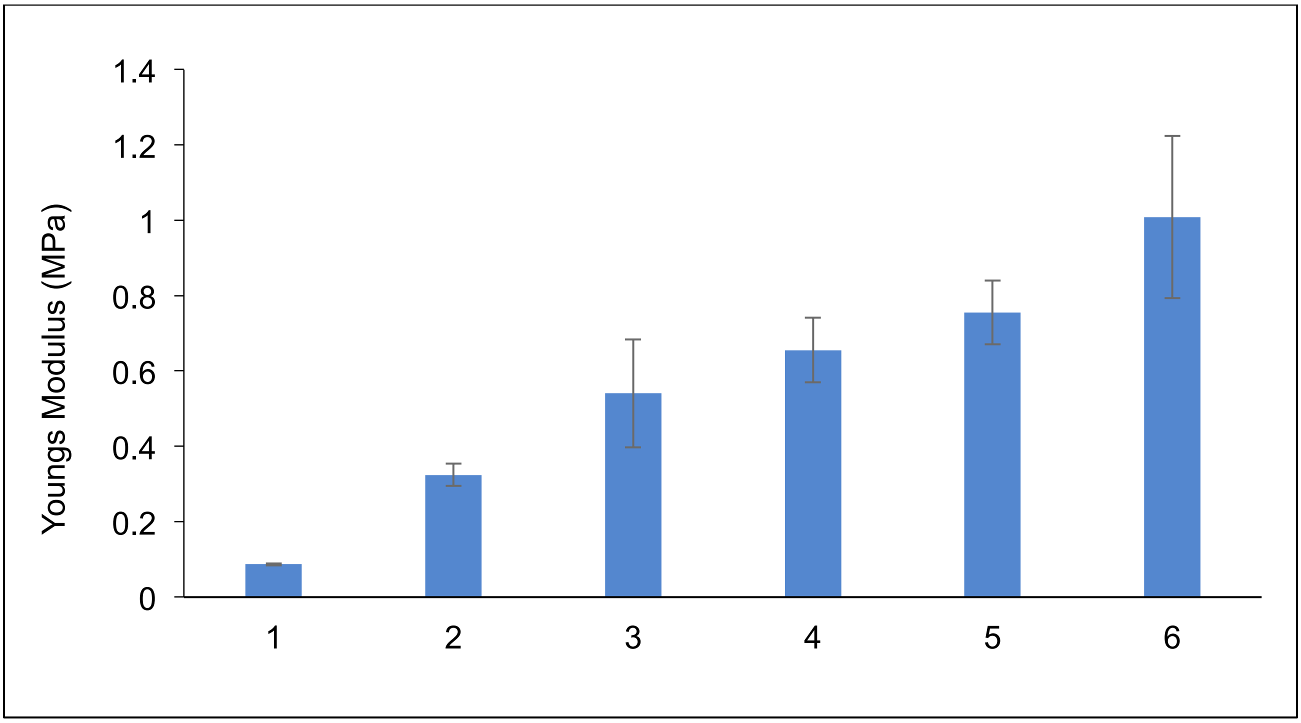
All samples were cut into rectangles (3.0 × 0.5 cm) using a scalpel. Triplicate samples were prepared for all tests. For the composite mats with the unidirectional fiber alignment, three samples were prepared in which the 3.0 cm length of the rectangle aligned with the fibers and three samples were prepared in which the 3.0 cm length of the rectangle was orthogonal to the fibers. All tensile tests were performed on a TA Q800 dynamic mechanical analyzer using the tension clamp. The measurement was accomplished in triplicate at room temperature (20 °C) with the ramp displacement 1000.0 µm/min. From these measurements a plot of stress (MPa) versus strain (%) was produced. From this plot the yield strength, the fracture point, and the Young’s modulus were determined. Note that for the data to be valid, the fracture must occur in the middle of the sample and not at the clamps. Only data obtained with fracture in the middle of the sample was used in the modulus analysis.



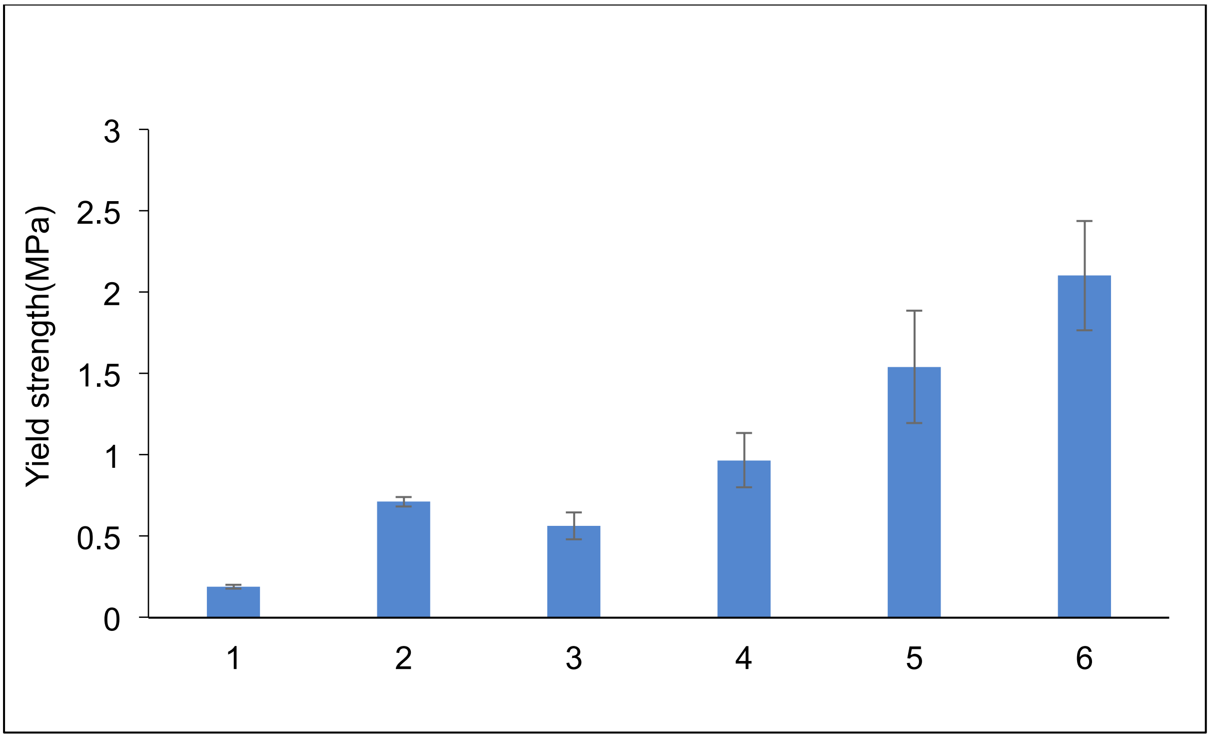
Polymer and composite samples at failure during tensile testing



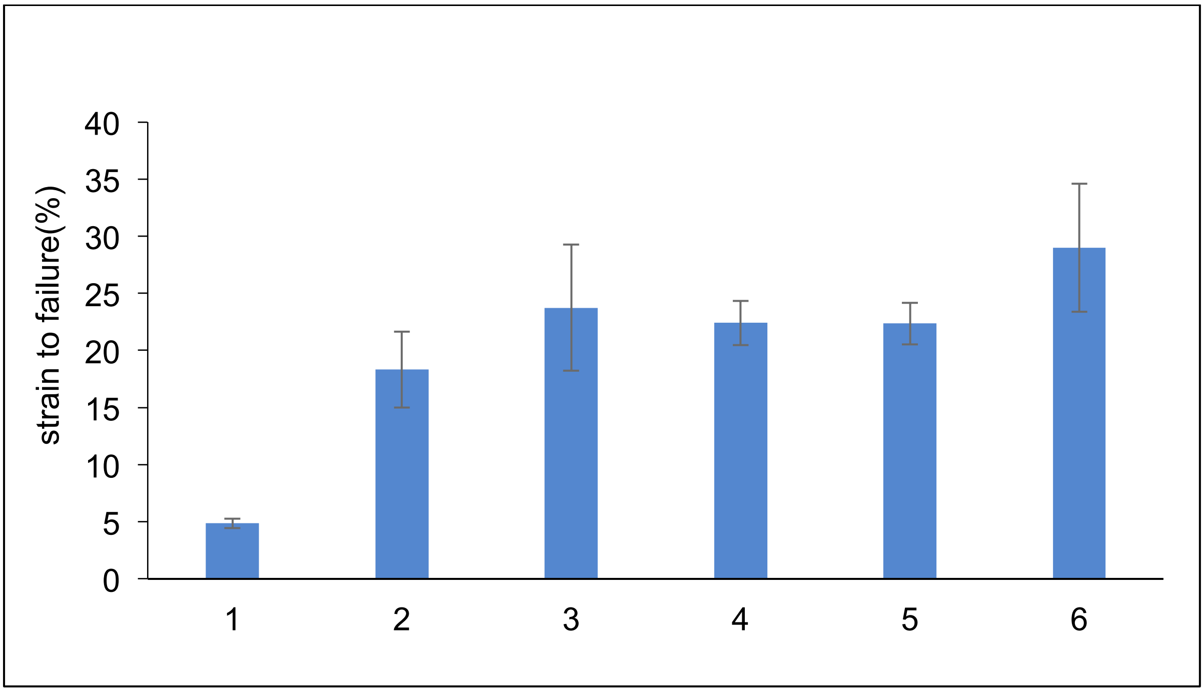
Stress strain curve for all samples produced during tensile strength measurements on the DMA. The sample numbering is the same as specified on the previous page.



Young's modulus (MPa) for all samples determined from the slope of the stress strain curve produced during tensile strength measurements on the DMA.



Yield strength (MPa) for all samples determined from the slope of the stress strain curve produced during tensile strength measurements on the DMA.



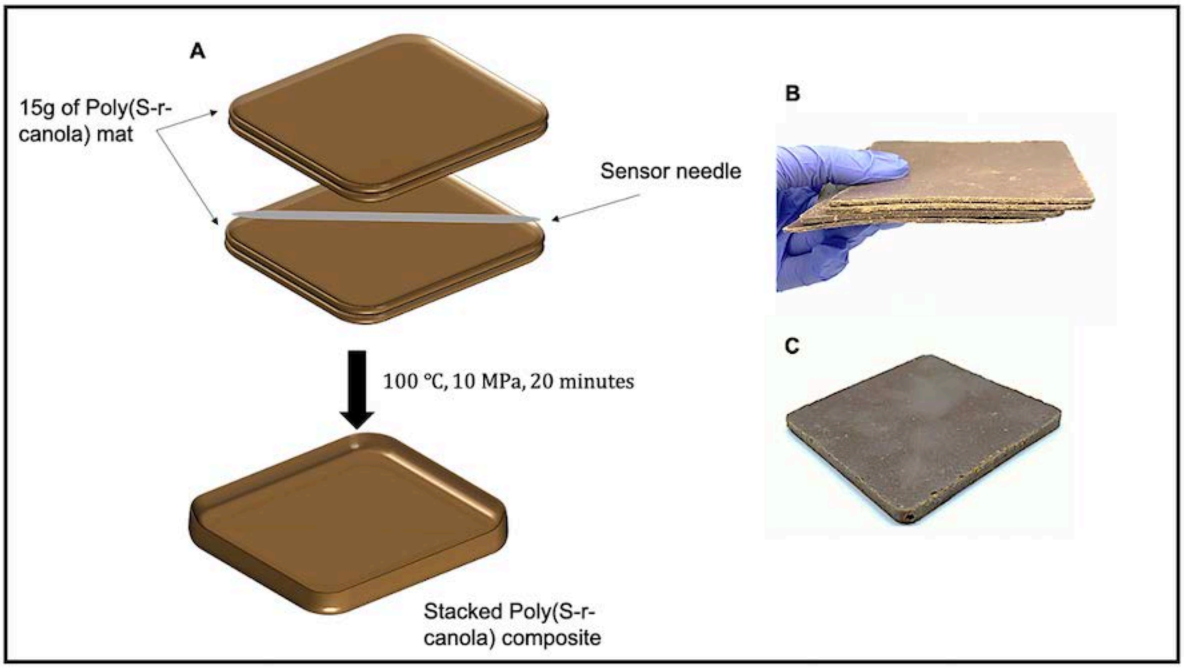
Fracture point (%) for all samples determined from the slope of the stress strain curve produced during tensile strength measurements on the DMA.

50-Poly(S-*r*-canola) mat preparation for conductivity tests

A 10 × 10 cm PTFE sheet was added to the aluminum mold. On top of the PTFE sheet, 15 g of 50-poly(S-*r*-canola) was added, followed by a second PTFE sheet. The lid for the mold was added, and the whole system was placed in hydraulic heated press, which was preheated to 100 °C. The heated plates were pressed until they were in contact with the mold without adding pressure. Once the temperature of the mold equilibrated at 100 °C the pressure was increased to 30 MPa for 15 minutes. After this time the pressure was released, and the mold was removed from the press. The resulting mat was removed from the mold and separated from the PTFE paper. The above process was repeated three times to prepare four 50-poly(S-*r*-canola) mats. The four 50-poly(S-*r*-canola) mats were used to make a thicker mat for the thermal conductivity test using additive reactive compression molding.

First a 10 ×10 cm PTFE sheet was added to the aluminum mold. On top of the PTFE sheet, two of the 50-poly(S-*r*-canola) mats were added, followed by a second PTFE sheet. The lid for the mold was added, and the whole system was placed in the hydraulic heated press pre-heated to 100 °C. The heat plates were pressed until they were in contact with the mold without applying any pressure. Once the temperature stabilised at 100 °C, the pressure was raised to 20 MPa. This pressure was maintained for 15 minutes before being released, and the resulting ~30 gram polysulfide mat was then removed from the mold. This process was repeated with the remaining two 50-poly(S-*r*-canola) mats to form a second ~30 gram polysulfide mat.

Next a 10 × 10 cm PTFE sheet was added to the aluminum mold followed by one of the 30-gram mats. A steel cover for the thermal conductivity meter was placed on top of the first mat so that the opening was direction in the corner of the mat. The second 30-gram mat was added on top before adding the second PTFE sheet. The lid to the mold was added and the entire system was placed in the press that was pre heated at 100 °C. The mold just fit into the press with the heated plates already being in direct contact with the mold. This was due to that larger volume of the two 30-gram mats inside the mold. No additional pressure was added in this case as the system was already under pressure due to the high volume of polymer in the mold. No added pressure was added to also ensure that the stainless-steel thermal conductivity probe cover wasn't damaged during the molding process. The system was left in the press for 20 minutes. After this time the mold was removed. The resulting mat was removed from the mold and separated from the Teflon sheets. A figure demonstrating this process is shown on the following page.



A) Process used to prepare the stacked 50-poly(S-*r*-canola) composite mat, **B)** Image of the four 50-poly(S-*r*-canola) preformed mats used, and **C)** Final product formed through additive manufacturing of 50-poly(S-*r*-canola) pre-formed mats.

Wool mat preparation for conductivity tests

A 10 × 10 cm PTFE sheet was added to the aluminum mold. The stainless-steel cover for the thermal conductivity meter was inserted between 13 g of randomly placed wool before being placed into the mold on top of the PFTE sheet. The lid for the mold was added, and the whole system was placed in the hydraulic heated press pre-heated to 100 °C. The heat plates were pressed until they were in contact with the mold without applying any pressure. Once the temperature stabilised at 100 °C, the pressure was raised to 20 MPa. This pressure was maintained for 30 minutes before being released, and the resulting composite mat then removed from the mold and separated from the PTFE sheets.

From left to right and top to bottom, images demonstrating the process used to prepare the wool composite mat for thermal conductivity.

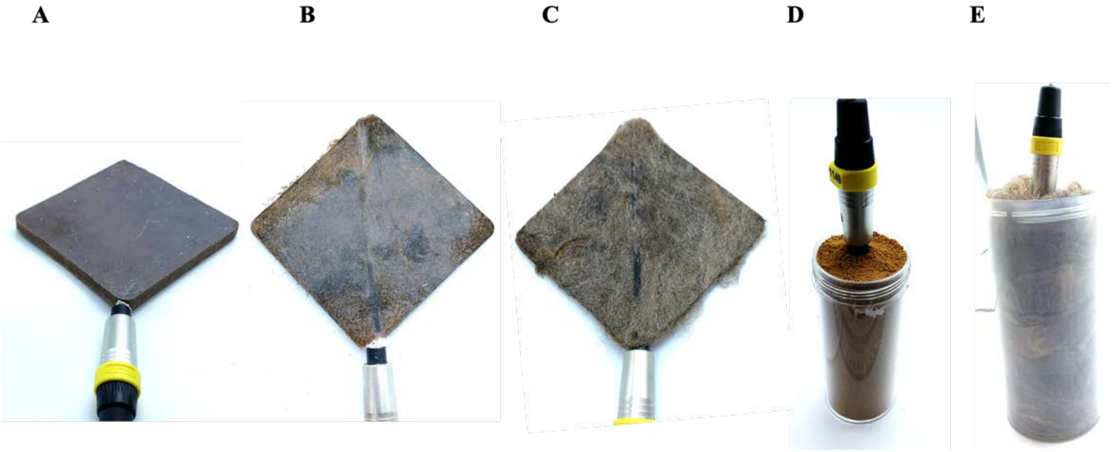
50-Poly(S-*r*-canola) / wool composite mat preparation for conductivity tests

A 10 × 10 cm PTFE sheet was placed in the aluminum mold. Then 10.0 g of raw wool was mixed with 10.0 g of polymer. The wool length was ~ 11 cm. The polymer particles adhered to the wool fibers, as previously described. The stainless-steel cover for the thermal conductivity meter was placed between the mixture so that it sat roughly in the middle. This was then added into the mold on top of the initial PTFE sheet. The second PTFE sheet was placed on top of the mixture. The mold lid was added, and the mold was placed in the press, which was preheated to 100 °C. The heat plates were pressed until they were in contact with the mold without adding pressure. When the temperature reached 100 °C, the pressure was increased to 30 MPa. This pressure was applied for 20 minutes. After this time the pressure was released, and the mold was removed from the press. The resulting composite was then removed from the mold and separated from the PTFE sheet.

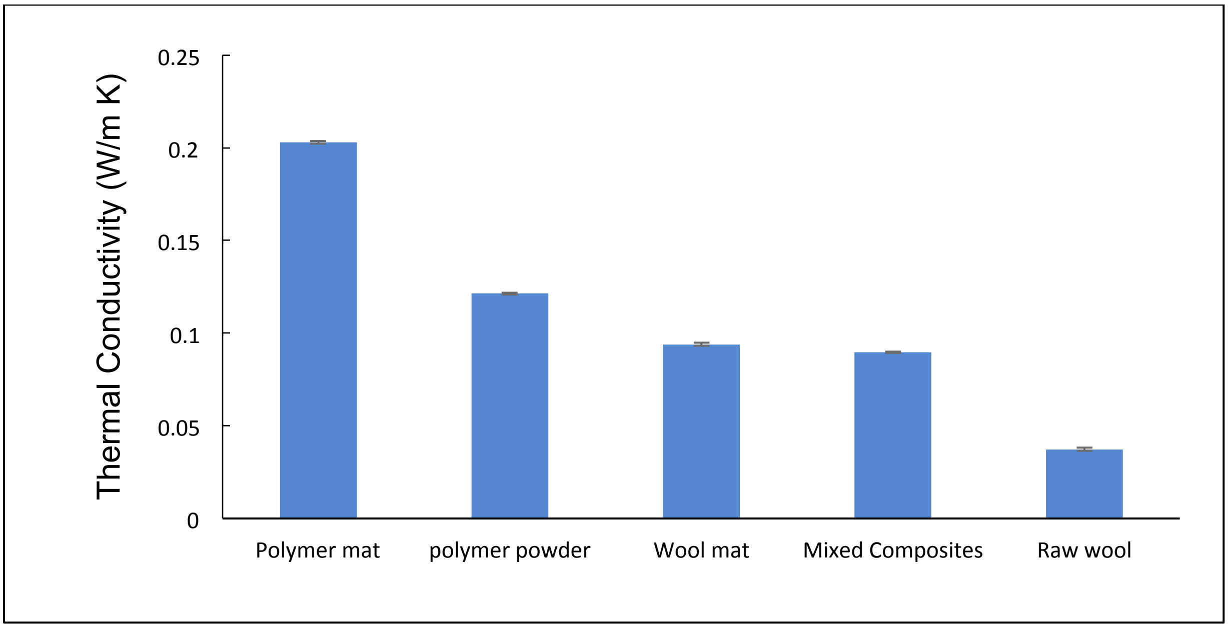
Thermal conductivity measurements

All thermal conductivity measurements were performed using a TLS-100 thermal conductivity meter, pictured below. A ceramic thermal compound paste (Arctic Silver Arctic Alumina) was added to the needle sensor before analysis to ensure complete contact with the material being tested. The sensor needle was left for 15 minutes after inserting in the sample before starting the measurement to allow the system to equilibrate. Initially the meter was calibrated by placing the sensor needle into a reference standard provided with the conductivity meter. For the polymer mat, the sensor needle was inserted in the 50-poly(S-*r*-canola) mat with the stainless-steel cover used during the mat preparation. The thermal paste was coated on the sensor needle to ensure that the needle was in direct contact with the cover to ensure optimal heat transfer to and from the sample. The system was left for 15 minutes to equilibrate before measurement. After this time the thermal conductivity and resistivity were recorded using the meter. This process was repeated two more times to get triplicate measurements for each sample. This entire process was then repeated for the wool mat and composite mat samples, each in triplicate. The results are shown on the following page.

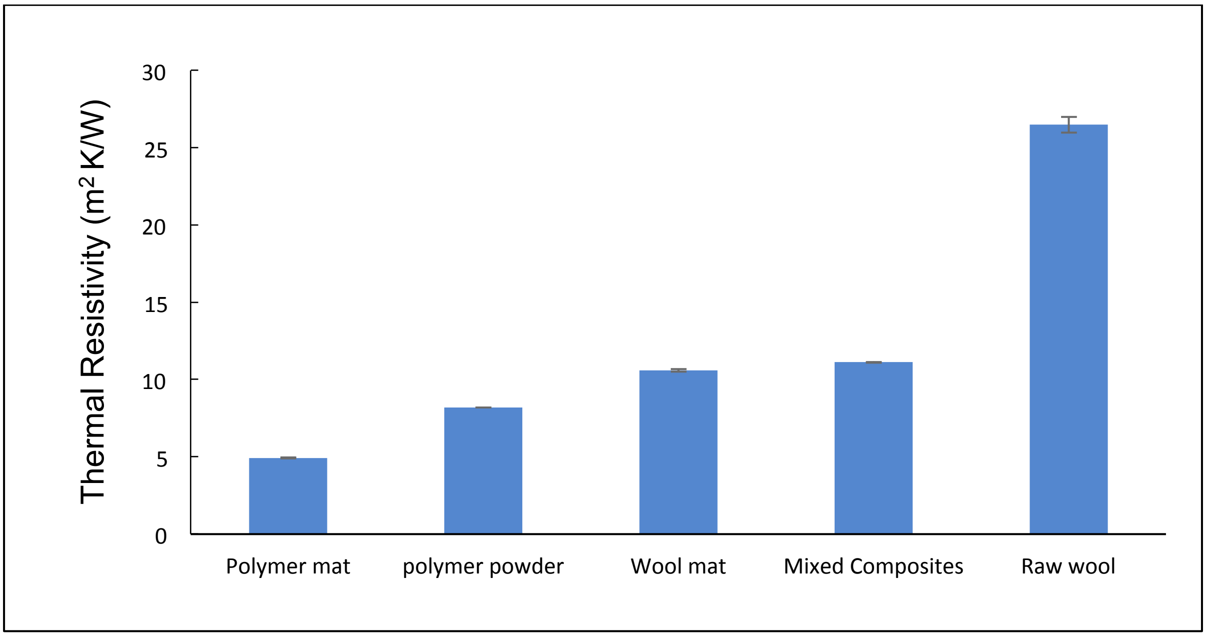
TLS-100 thermal conductivity meter

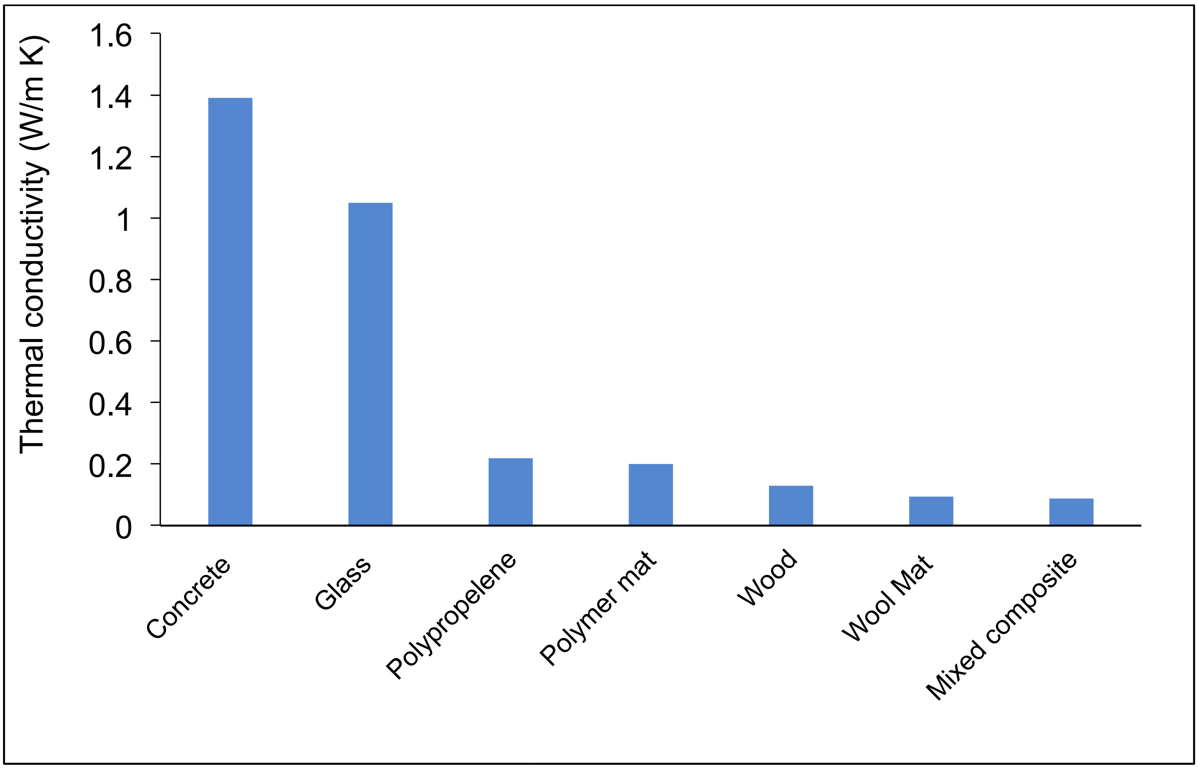


Thermal conductivity testing using the TLS-100 Thermal conductivity meter. **A)** 50-Poly(S-*r*-canola) mat, **B)** Mixed composite mat made from 50-poly(S-*r*-canola) polymer and wool, **C)** Wool mat, **D)** 50-Poly(S-*r*-canola) powder, and **E)** Raw wool. The thermal conductivity is plotted below.



[The "mixed composite' sample refers to the mat made from equal masses of 50-poly(S-*r*-canola) and wool, with the wool randomly aligned]



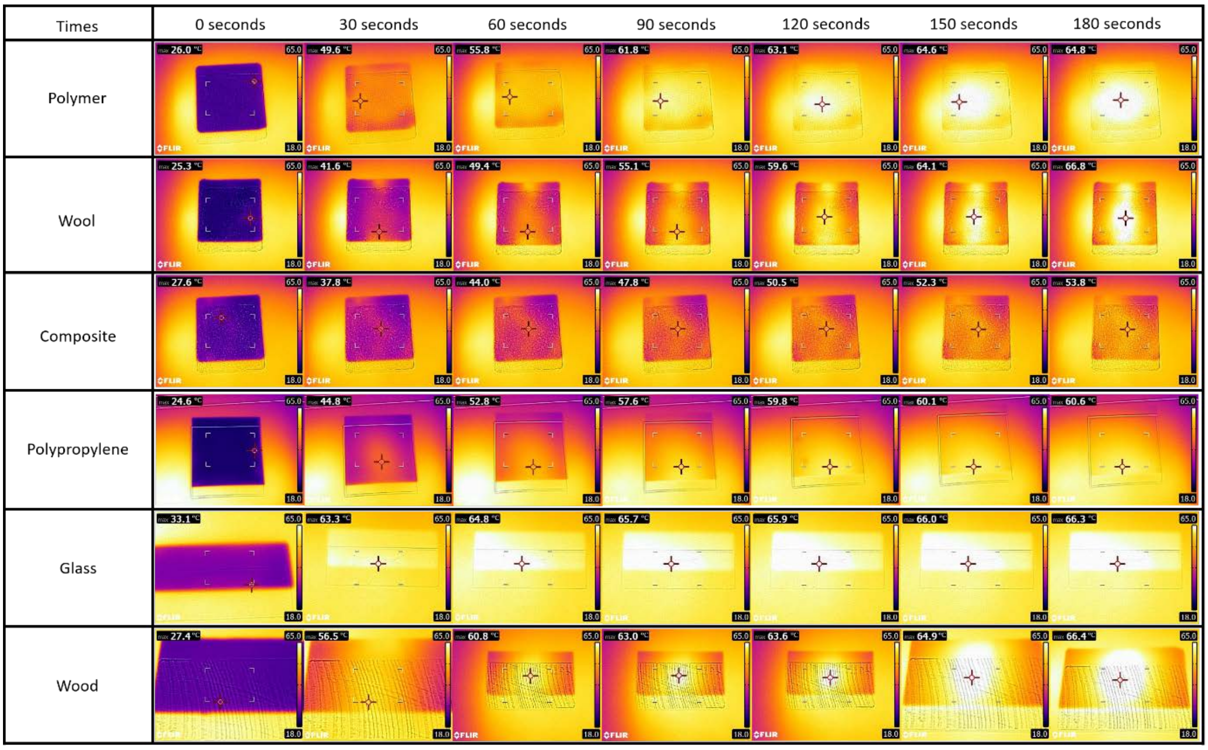


Thermal conductivity of concrete[3], wood[4], glass and polypropylene[5] for comparison

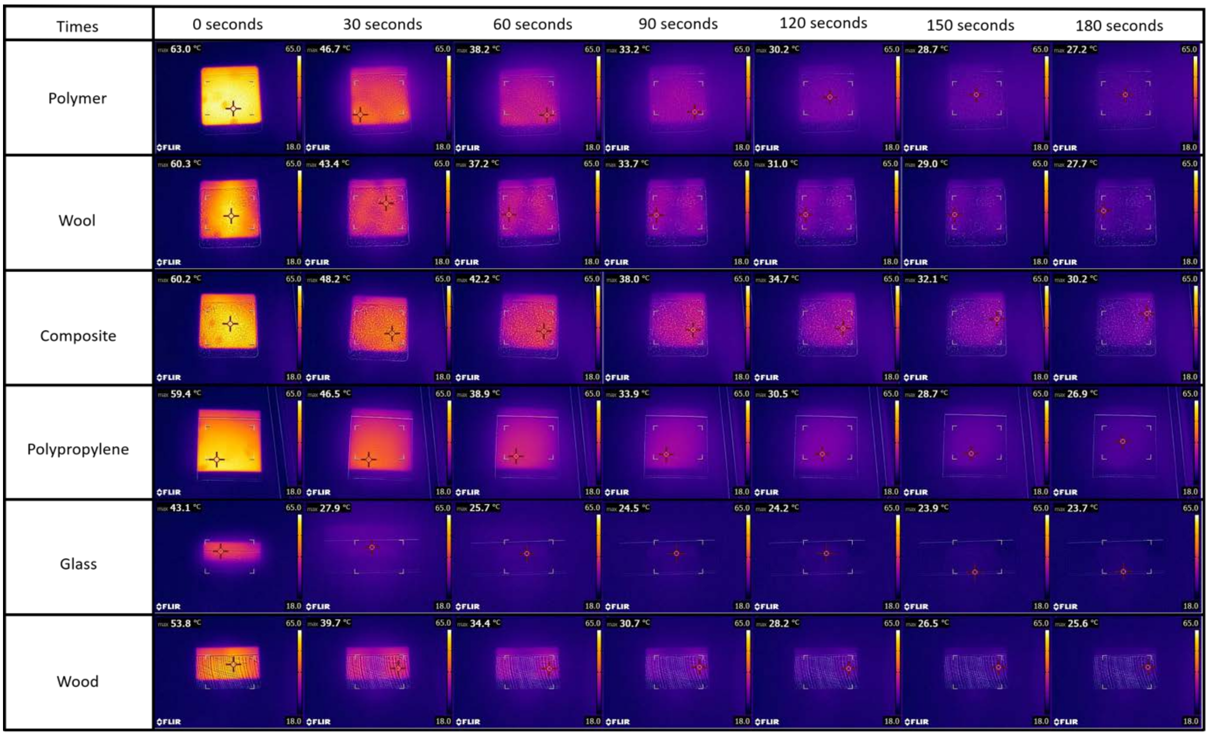
[The "mixed composite' sample refers to the mat made from equal masses of 50-poly(S-*r*-canola) and wool, with the wool randomly aligned]

FLIR imaging

All FLIR imaging was performed using a FLIR Ex series camera with the temperature scale range set to 18-65 °C in Thermal imaging mode. Auto Hot Spot mode was used to ensure the hottest area on the substrate was recorded. A hotplate was pre-heated to approximately 60 °C. An aluminum sheet was placed on top of the hotplate and left to equilibrate to 60 °C. The aluminum sheet ensure even heating across the surface on which the sample was placed. The 100% polymer control mat (1.3 mm thickness) was placed directly in the center of the tray on the hotplate and thermal imaging was started. Images of the sample were recorded every 10 seconds for 3 minutes or until the sample had reached 60 °C. Once it reached this temperature, the sample was removed from the hotplate and transferred directly to a second aluminum sheet to cool down. Images were recorded every 30 seconds over 3 minutes furing the cool down. This process was repeated for samples of wool, the polymer-wool composite (equal masses of polymer and wool, randomly mixed before reactive compression molding), polypropylene, glass and wood. All samples were prepared at a thickness of 1.3 mm. The results are shown below.



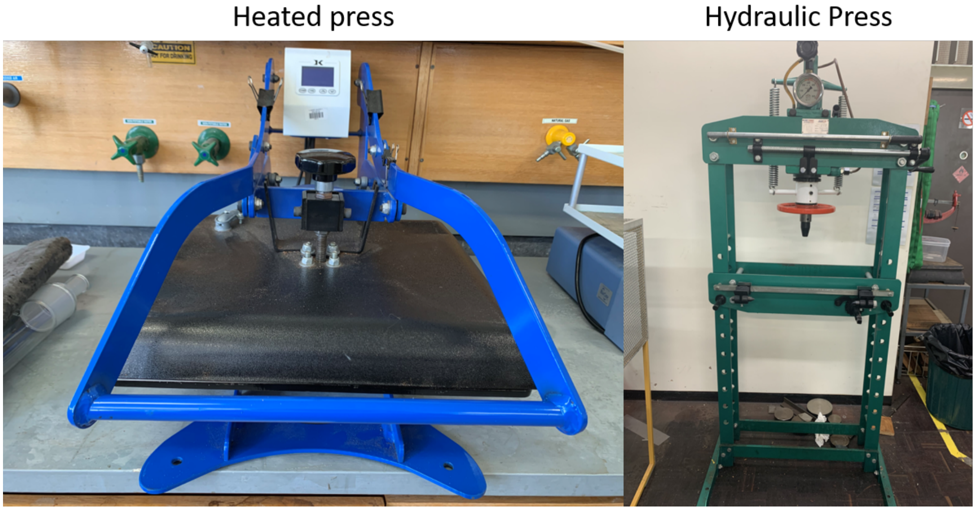
Images showing sample heating over three minutes



Images showing sample cooling over 3 minutes

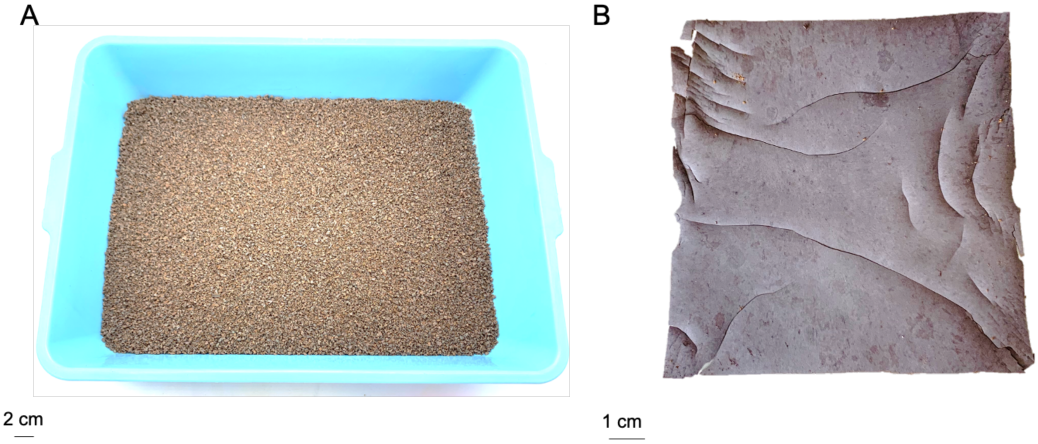
Large scale 50-poly(S-*r*-canola) mat preparation (250 g)

Two aluminum plates and two steel plates were pre-heated in an oven for 1 hour at 150 °C. Baking paper was placed in an aluminum tray (40 × 50 cm) followed by 250 g of 50-poly(S-*r*-canola), and the second baking paper was added above followed by the second tray. The baking paper prevented the polymer from sticking to the aluminum. The whole system was placed into the DK 16 Digital Knight press, shown in the figure below, for 5 minutes at 120 °C. After that time, the trays were flipped and heated for an extra 5 minutes. The whole system was removed from the hot press. The first steel and aluminium plates were removed from the oven. Directly after removal, the steel plate was placed on the JL-15A Ezylif 15-ton hydraulic press followed by the aluminium plate. The JL-15A Ezylif 15-ton hydraulic press is shown below.



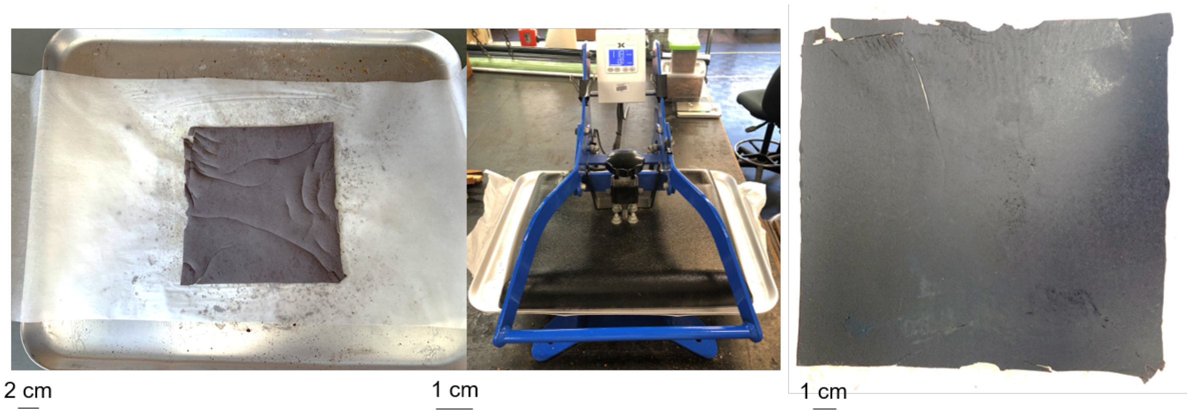
Heated press (left) and Hydraulic press (right)

The 50-poly(S-*r*-canola) polysulfide, sandwiched in between the two aluminium trays with baking paper, was added on top of the first preheated aluminium plate. The second aluminium plate was added directly on top of the aluminium tray followed by the second pre-heated steel plate. The pressure on the JL-15A Ezylif 15-ton hydraulic heated press was then increased to 300 kg/cm^2. This pressure was maintained for 10 minutes. After that time the pressure was released, and the polymer mat was removed. The sample is shown below.



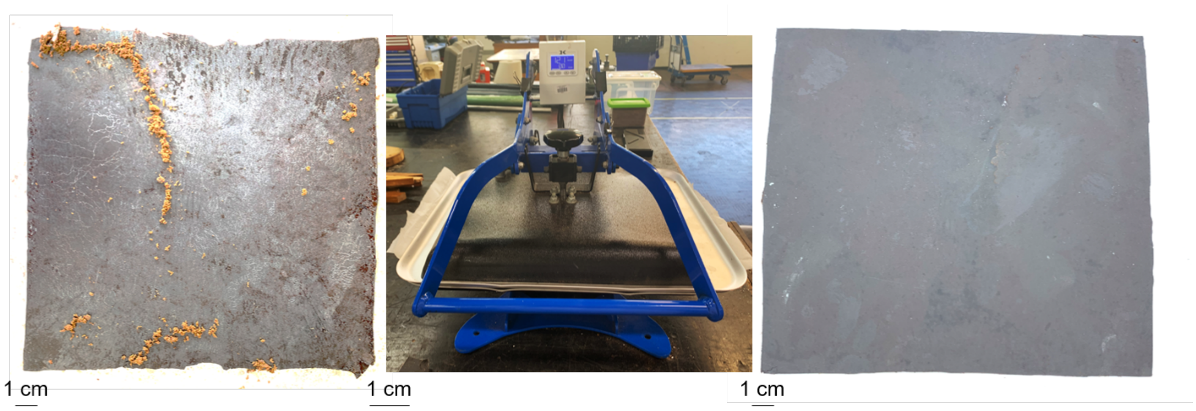
A) 50-poly(S-*r*-canola), **B)** 50-poly(S-*r*-canola) after molding

To make the polymer mat more uniform and seal the cracks in the sample, it was placed in between two aluminum trays with baking paper separating the sample from the trays. The system was then placed back into the DK 16 Digital Knight press at 120 °C for 2 hours. After 2 hours most of the cracks had healed leaving a few remaining. This can be observed in figure below.



50-poly(S-*r*-canola) mat contains significant cracking (left), 50-poly(S-*r*-canola) mat in the DK 16 Digital Knight press (middle) and resulting 50-poly(S-*r*-canola) mat after 2 hours heated (right).

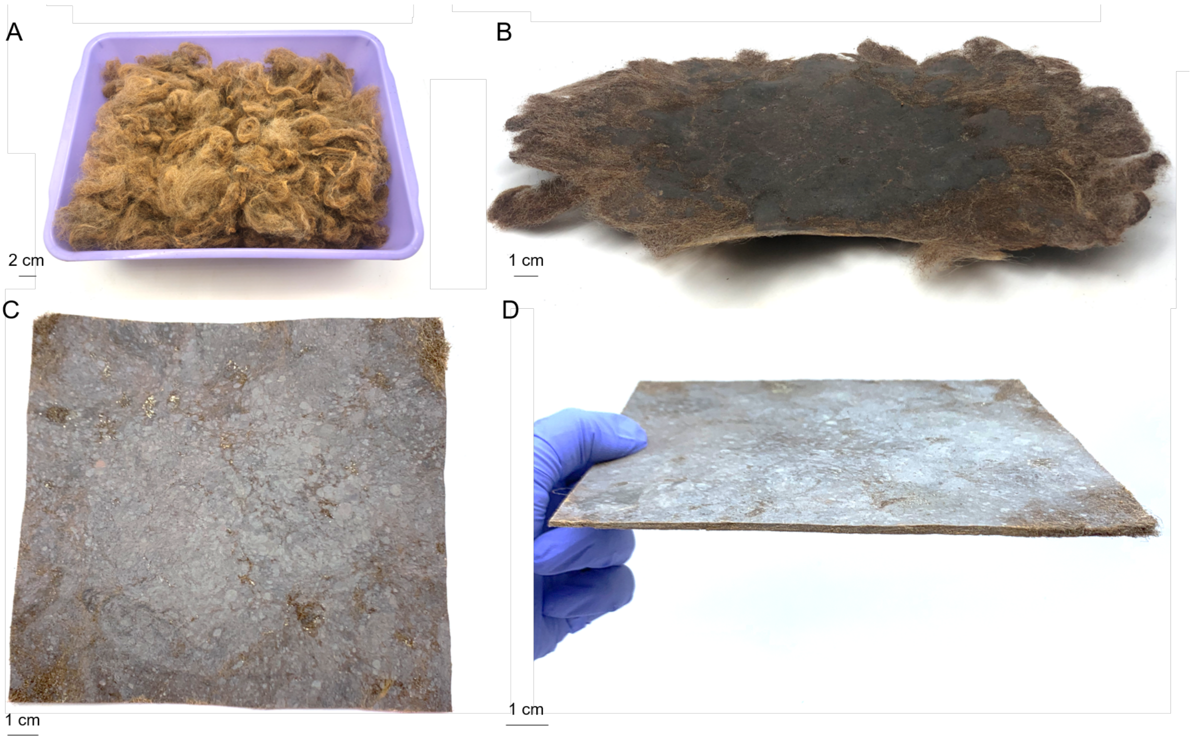
To repair the remaining cracks 0.2 grams of 50-poly(S-*r*-canola) powder was placed over the damaged araa. The whole system was placed back into the hot press for a further 2 hours. After that time, the polymer mat was flipped and pressed for extra 2 hours. The final 250 g mat was removed and was uniform with no cracks.



50-poly(S-*r*-canola) mat with 50-poly(S-*r*-canola) powder sprinkled over the cracks (left), 50-poly(S-*r*-canola) mat in the DK 16 Digital Knight press (middle) and resulting 50-poly(S-*r*-canola) mat (right).

Large scale of 50-poly(S-*r*-canola) / wool composite preparation (200 grams)

Two aluminum plates and two steel plates were pre-heated in an oven for 1 hour at 150 °C. 100 grams of raw sheep wool with length of ~11 cm was mixed with 100 grams of 50-poly(S-*r*-canola) by hand. Baking paper was placed in aluminum tray (40 × 50 cm) followed by the polymer-wool mixture. A second sheet of baking paper was placed on top before adding the second aluminum tray. The whole system was placed into DK 16 Digital Knight press for 5 minutes at 120 °C. After that time the sample was flipped and heated for an extra 5 minutes. Then the whole system was removed from the hot press and transferred to the JL-15A Ezylif 15-ton hydraulic heated press. The first steel and aluminium plates were removed from the oven. Directly after removal, the steel plate was placed on the JL-15A Ezylif 15-ton hydraulic press followed by the aluminium plate. The wool-50-poly(S-*r*-canola) mixture, sandwiched between two aluminium sheets, was added on top of the first preheated aluminium plate. The second aluminium plate was added directly on top of the aluminium tray followed by the second pre-heated steel plate. The pressure on the JL-15A Ezylif 15-ton hydraulic heated press was then increased to 400 kg/cm2. This pressure was maintained for 10 minutes. After that time the pressure was released, and the composite mat was removed.



A) Raw wool mixed with 50-poly(S-*r*-canola), **B)** Mixed composite mat after reactive compression molding, **C, D)** Mixed composite mat after cut from the center

Flammability testing

UL-94 vertical burn tests were carried out as described in the main text and carried out according to ASTM D3801-19.

Video S1 and Video S2 show representative tests for the 50-poly(S-*r*-canola) and the polymer wool composite, respectively. These video files are available to view as Electronic Supplementary Information.

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Other files