Synthesis and gas permeability of highly elastic poly(dimethylsiloxane)/graphene oxide composite elastomers using telechelic polymers

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Abstract
This study illustrates that amine functional groups on the ends of telechelic poly(dimethylsiloxane) (PDMS) can undergo post-processing reactions with surface epoxy groups on graphene oxide (GO) to form a robust elastomer during simple heating. In these materials, GO acts both as a nanofiller which reinforces the mechanical properties and participates as a multifunctional crosslinker, thereby promoting elastic properties. Experiments indicate that the telechelic PDMS/GO elastomer is highly crosslinked (e.g., more than 75 wt % is a non-dissolving crosslinked gel) but highly flexible such that it can be stretched up to 300% of its original length. Finally, the PDMS/GO elastomer was tested as a single gas barrier membrane and gas permeability was decreased ~45% by incorporating 1 wt % (0.43 vol %) of GO, thereby highlighting its potential use in practical applications.

1. Introduction

In recent years, flexible polymers have become increasingly important for many applications that impact daily life, including wearable devices [1–3], flexible displays [4–6], and devices for monitoring physiological signals [7–9], to name a few. These high-end applications typically require the use of an elastomer or rubber, such as crosslinked poly(dimethylsiloxane) (PDMS), to impart flexibility and mechanical robustness in addition to comfort; all are important features for interfacing with the human body. While there exist commercial choices for PDMS elastomer precursors, such PDMS precursors are generally high in viscosity, limited to several grades and the recipe for preparing the final elastomer can be complex [10–12]. Therefore, the development of new methods for forming elastomers with only a few simple components which exhibit unique combinations of properties (e.g., electrically conductive but flexible and mechanically robust, etc.) could be very useful.

Herein, we report a simple process for creating a strong and flexible elastomer from a reactive mixture of graphene oxide (GO) and functional telechelic (i.e., both ends functionalized) PDMS. The resulting composite has excellent mechanical properties but also serves as an effective and potentially economical gas barrier membrane for industrial use. For context, materials presenting a high barrier to oxygen permeation are typically very important for the applications listed above because flexible devices often contain organic components to promote flexibility that can be particularly susceptible to oxidative degradation.

For a number of reasons, the telechelic PDMS/GO composite elastomer described in this study can be considered to be unique when compared alongside other crosslinked PDMS materials. For instance, a distinguishing characteristic of the present elastomer is that it not only possesses the traditional properties of crosslinked PDMS materials, such as flexibility and good mechanical integrity, but it can also be made by simple and scalable methods using two low-cost materials: graphite and telechelic polymer. In contrast, conventionally crosslinked PDMS elastomers are typically synthesized by one of three methods: platinum-catalyzed addition, vinyl-peroxide, or condensation curing [13]. All of these methods require a catalyst to perform the reaction, accompanied with a range of several different chemical additives, sometimes including fillers such as silica [11,12]. Many prior studies have investigated the tuning of physical properties, including mechanical, electrical, and
dielectric properties, of PDMS materials by adjusting the crosslink density [14], adding fillers such as silica [15], clays [16], carbon nanotubes [17], graphene derivatives [18] or other polymers [19]. Generally, these additional additives are most commonly physically intermixed with PDMS with no chemical bonding between the PDMS and additives. In contrast, the preparation of telechelic PDMS/GO composite elastomers in this study incorporates a GO filler that also participates as a multi-functional crosslinker, which facilitates formation of the final covalently bonded macromolecular network.

While neat PDMS is highly gas permeable, the addition of impermeable platelet-like fillers such as GO can dramatically inhibit gas permeation. Recent studies of gas barrier membranes using different graphene derivatives have focused on coating the surface of flexible substrates (such as PDMS films or sheets) with a thin layer of graphene or GO that is typically weakly bound by noncovalent adhesion forces [6,20,21]. Graphene coated membranes could be susceptible to sections of the coating delaminating from the substrate after prolonged repeated stress, bending or upstream/downstream pressure cycles [6,20,22]. Other potential sources of damage from environmental exposure of the active graphene or GO coating layer, such as irreversible adsorption of undesired contaminations during prolonged use, could be possible. In contrast to this coating approach, the elastomer described herein forms a covalent macromolecular network between the flexible component (PDMS) and the gas impermeable component (GO) producing a dense and homogeneous barrier membrane. In summary, from a membrane engineering perspective, we believe this composite provides for the possibility of a cost-effective, high performance gas barrier membrane.

2. Experimental section

2.1. Materials

In order to synthesize PDMS/GO elastomers, three components were combined: telechelic PDMS as the base material, GO as both a filler and multifunctional crosslinker, and tetrahydrofuran (THF; Fisher Chemical) as a solvent. Aminopropyl terminated telechelic PDMS with a viscosity of 1000 cSt (Mn = 25,000 g/mol, DMS-A31, henceforward referred to as N1000) and 2000 cSt (Mn = 30,000 g/mol, DMS-A32, henceforward referred to as N2000) were purchased from Gelest and used without any further purification. For comparison, silanol terminated PDMS with a viscosity of 1000 cSt (Mn = 26,000 g/mol, DMS-S31) and epoxypropoxypolypropyl terminated PDMS with a viscosity of 15 cSt (Mn = 550 g/mol, DMS-E111) were also purchased from Gelest and used as received. GOS were synthesized by the modified Hummer’s method as described elsewhere [23], where sulfuric acid, potassium permanganate, hydrogen peroxide, and hydrochloric acid were purchased from Sigma–Aldrich and used as received. Pre-oxidized graphite from Bay Carbon Inc. (SP-1) was used as the starting material to synthesize GOS. The nominal number average particle size of the SP-1 was used as the starting material to synthesize GOs. The nominal number average particle size of SP-1 was used as the starting material to synthesize GOs.

2.2. Fabrication of PDMS/GO elastomers

First, a known amount of telechelic PDMS (for 1 wt % GO composites, 990 mg) was dissolved in THF (6 ml) while stirring to ensure complete dissolution of the polymers. Then, dried GO (10 mg) was suspended in the solution with vigorous stirring, and a homogeneous dispersion of GO in this solution was promoted by sonication using a 400 W probe sonicator (Branson Digital Sonifier 450) with 10% amplitude for 10 min (24 kJ) in an ice bath. It is noteworthy to point out that, although THF is not a good solvent for GOS, amine terminated telechelic PDMS can effectively act as a surfactant to assist in dispersing GOS homogeneously in THF through hydrogen bonding between amine ends and oxygen containing groups on GO surfaces. A good dispersion after sonication was confirmed by examining the residual liquid film in the vial while rolling the vial containing the solution and after pouring the majority of the solution into a Teflon dish for solution casting. In either case, the liquid film in the vial was essentially transparent (although brownish-black from the GO content) for all solution concentrations and no aggregates could be identified with the naked eye. This solution was immediately poured into a Teflon dish and covered in order to slowly evaporate the solvent thereby solution casting a film at room temperature. Solvent was evaporated for at least 2 days and then the sample was vacuum dried at room temperature for an additional day to make a homogeneous PDMS/GO uncrosslinked liquid sol. In order to form a crosslinked elastomer, the sol precursor was heated to 160 °C in a vacuum oven for 24 h and cooled slowly to room temperature before use.

For one of the gas permeation tests, a reference sample (neat PDMS elastomer) was synthesized by reacting aminopropyl terminated telechelic PDMS and epoxypropoxypolypropyl terminated telechelic PDMS. A detailed explanation of how this freestanding, neat PDMS elastomer was synthesized is summarized in the supporting information.

2.3. Characterization of PDMS/GO elastomers

The chemical crosslinking reaction between PDMS and GOS was investigated using Fourier transform infrared spectroscopy (FTIR; Thermo Nicolett, 6700) with a scan size (resolution) of 2 cm⁻¹ collected over 256 scans per sample. Solvent uptake and gel content of each composite elastomer were calculated using Equations (1) and (2) below where ‘w’ indicates weight, according to ASTM D2765 test method C. THF was used as the solvent to dissolve away unreacted soluble polymers.

\[
\text{Solvent Uptake (\%)} = \frac{W_{\text{after swell}} - W_{\text{before swell}}}{W_{\text{before swell}}} \times 100 \quad (1)
\]

\[
\text{Gel Content (\%)} = \frac{W_{\text{after swell and dried in vacuum}}}{W_{\text{before swell}}} \times 100 \quad (2)
\]

All rheological experiments were conducted on a shear rheometer (TA Instruments, AR-2000EX) using an 8 mm parallel upper plate and a Peltier lower plate fixture for temperature control with a gap of 500 μm. First, a strain sweep was conducted to identify the linear viscoelastic regime at 25 °C in the range of 0.01–10% strain at a frequency of 1 Hz. A frequency sweep was then conducted at 25 °C in the range of 0.01–10 Hz with 0.1% strain. Temperature sweeps were conducted from 25 °C to 100 °C with 5 °C step changes using both 0.1% strain and a frequency of 1 Hz. Stress relaxation tests were conducted by subjecting the sample to an initial strain of 10% and 20%. In order to measure the mechanical properties of the composite elastomers, microtensile specimens were prepared by solution casting on a mold (i.e., to produce a dog bone sample with a gauge length of 22 mm, width of 4.8 mm and thickness of about 0.7 mm) satisfying ASTM D1708–13 standards. An Instron (model 5966) equipped with a 1 kN load cell was used with a strain rate of 10 mm/min (i.e., at our gauge length this is a nominal strain rate equivalent to 50 mm/min for ASTM D638 standard samples, which is the recommended speed for rubber samples) and samples were tested in triplicate.
Thermal properties of the elastomers were characterized by differential scanning calorimetry (DSC; Mettler Toledo DSC1). Heating and cooling rates of 10 °C/min were used for all experiments, and the second heating curve was used for the glass transition temperature (T_g) analysis to erase thermal history. GOs were further characterized using X-ray photoelectron spectroscopy (XPS; Kratos) to compare the composition of the GOs before and after annealing. CasaXPS software (v 2.3.16) was used to perform curve fitting in which a Shirley background was assumed. The dispersion of GOs in the composite material was determined by optical microscopy (OM; Olympus BX60) using a micromotated sample on a 100 mesh Cu grid. Sectioning was performed by a benchtop microtome (RMC Products, PT-XL Power Tome) with a diamond knife where the temperature of the diamond knife and the sample holder were both set to –150 °C. According to the images (supporting information), all of the composite materials considered in this study showed good dispersion of GOs without any noticeable large aggregates.

2.4. Single gas permeation measurements

Single gas, including H_2, O_2, N_2, CH_4 and CO_2, transport properties were measured using the constant volume, variable pressure method at 35 °C with ultra-high purity grade gases from Airgas. A 1000 psig pressure transducer (Honeywell Sensotec, Model STJE) was used to measure the upstream pressure in the system. A 10 Torr capacitance manometer (MKS, Baratron 626 A) was used to measure the downstream pressure, and the downstream pressure was kept below 10 Torr using a vacuum pump. All data were recorded using National Instruments Lab-VIEW software. Permeability, which is an intrinsic property of a material to a specific permeate, was expressed in barrer units, where 1 barrer equals to 10^{-10} cm^3(STP) cm/cm^2 s cmHg. The average thickness of the sample was measured using a dial gauge (Mitutoyo, Absolute digimatic indicator) by sandwiching a sample in between two thin metal plates (each metal plate thickness = 0.05 mm). Metal plates were used to prevent samples from being compressed by the tip of the dial gauge during measurement. Thicknesses for the neat PDMS elastomer and telechelic PDMS N1000/1 wt % GO elastomer were 0.998 ± 0.021 mm and 0.354 ± 0.010 mm, respectively.

3. Results and discussion

To fully understand the chemical nature of the thermally crosslinked telechelic PDMS/GO elastomers, a fundamental investigation regarding the reaction between amine terminated telechelic PDMS and GO was pursued first.

3.1. Proposed reaction and evidence of crosslinking

As shown in Scheme 1 (a), only two materials were used to form the highly elastic elastomer, the first being amine terminated telechelic PDMS, which contains a primary amine attached to each end of the polymer chain. The second material, GO, contains numerous oxygen-containing functional groups, including hydroxyls, carboxylic acids, and epoxides, on the surface of the GO sheets [26]. XPS data supports the fact that hydroxyls and epoxides are present in larger proportion than other functionalities (see Figure S2). Once the two materials are mixed together and sufficient energy is applied, in this case heat, an epoxide on GO can react with the nearest primary amine on PDMS by a ring-opening reaction. Such a reaction results in the formation of ethanolamine covalent bonds between the telechelic polymer ends and the epoxy groups on the surface of GO, as shown in Scheme 1 (b). This nucleophilic substitution reaction has been proposed and demonstrated previously by other groups when functionalizing GO surfaces with amines [27,28] as well as for synthesizing chemically crosslinked nyons [29,30], epoxy composites [31–33], and polymer/GO papers [34,35]. However, to the best of our knowledge, no research implementing telechelic polymers with GOs to form a controllable, highly elastic composite material has been reported [20]. It is worth noting that we have also confirmed this reaction proceeds similarly for PDMS materials containing secondary amide end groups [36].

There are three possible scenarios for amine terminated telechelic PDMS to react with GOs: two ends react with two different GO sheets, one end reacts with one GO sheet while the other end forms a dangling strand and remains unreacted, or two ends react with the same GO sheet forming a loop. Considering the high gel content obtained for various samples (which will be discussed in more detail in a later section), it is postulated that the majority of the reaction takes place by forming a crosslink between two different GO sheets, since the latter two possibilities would not contribute to enhancing the gel content and mechanical properties of the composite. Given that reaction is only possible at chain ends, telechelic polymers offer the opportunity to produce elastomers with a predefined and tailorable molecular weight between crosslink points. Considering that this reaction only involves polymer end-groups, the versatility and universality of this crosslinking reaction could be easily demonstrated by applying it to other repeat unit chemistries extending this approach to polymers that are amorphous or semicrystalline, hydrophobic or hydrophilic, synthesized by either addition or step growth polymerizations, etc. Finally, due to the simplicity of the reaction and the fact that no byproducts are formed during crosslinking, it is expected that this facile method could be scalable to mass produce composite materials in a cost-effective way.

As shown in Fig. 1, simply annealing the solution cast telechelic PDMS/GO composite film in vacuum for 24 h resulted in a material that was freely standing, highly flexible and mass producible in different sizes and shapes (note the larger size in Fig. 1 (f)). The transformation of a viscous sol to a self-supporting solid elastomer provided indirect evidence of a reaction forming covalent bonds between the polymer and GO. For comparison, a silanol terminated telechelic PDMS/GO composite remained a soluble viscous liquid even after annealing at high temperature (Fig. S1). This clearly demonstrates the importance of complementary reactive functionalities for performing the reaction that leads to the final elastomers.

While the occurrence of the crosslinking reaction is evidenced by the images in Fig. 1, absorbance spectra obtained using FTIR provide more direct evidence of the chemical changes associated with this reaction (Fig. 2). To understand the crosslinking reaction and formation of the ethanolamine linkage, the telechelic PDMS/GO mixture was investigated before and after annealing. As expected, the majority of the intense peaks in the spectra were related to the PDMS main chain repeat units; detailed peak assignments for Fig. 2 (a) are tabulated in the supporting information in Table S1. For the N1000 telechelic PDMS used in Fig. 2, with amine ends present at approximately 0.6 mol % (i.e., 0.12 wt %), the 1500–1700 cm^{-1} region of the spectra was examined in further detail in Fig. 2 (b). Before annealing, the peak associated with N–H bending was assigned at 1600 cm^{-1} for both N1000 PDMS and N1000 PDMS/1 wt % GO sol samples. However, after annealing the sol to crosslink the sample producing an elastomer, a significant fraction of the N–H bending peak disappeared, indicating that the majority of the primary amine end groups were reacted during this step. In addition, due to a known thermal annealing induced reduction reaction of GO to reduced GO, a stronger C=C ring stretching peak at 1580 cm^{-1} was formed for the N1000 PDMS/1 wt % GO elastomer.
sample. A separate case study on the effect of heat on the transformation of GO to reduced GO is provided in Fig. S2. Because the FTIR spectra after extended annealing were nearly identical to those after 24 h of annealing at 160 °C, it can be concluded that the reaction was essentially completed within 24 h.

Another method to confirm the formation of chemically crosslinked elastomers is to measure the swelling ratio and gel content of the composite material. By controlling the content of GO and the Mn of the PDMS polymer incorporated in this system, the gel crosslink density can be altered effectively (Table 1). For example, increasing the GO content resulted in the introduction of more crosslinking sites and, as a result, dramatically increased the gel content and decreased the solvent uptake of the elastomers. On the other hand, as the Mn of the PDMS was increased from 25 to 30 kg/mol, the mol fraction of the amine end groups was reduced from 0.6% to 0.5%. Table 1 shows that the degree of swelling expectedly increased and gel fraction decreased; the likelihood of amines reacting with the surface epoxy groups of GO would be expected to

Scheme 1. (a) Chemical structures of the two materials used in this study. Each material’s active sites are capable of reacting with complementary functional groups (colored) on the other component. (b) The anticipated reaction mechanism that produces elastomers in this study. No other byproducts are formed during the crosslinking reaction.

Fig. 1. Images of N1000/1 wt % GO elastomers. (a) As prepared material is not only (b) freestanding, but also (c) bendable and (d) flexible. (e) The material is elastic enough to make a knot. (a–e) is a N1000/1 wt % GO elastomer disc 38 mm in diameter, while (f) is 10 cm in diameter.
decrease in this situation reducing crosslinking density. An associated factor is that the mobility of a higher $M_n$ polymer could also be slightly lower due to increased entanglements and viscosity. It is noteworthy that the viscosity of the telechelic PDMS component, which is directly related to the $M_n$ of the polymer, plays an important role in forming a homogeneous crosslinked material following solution casting and thermal annealing. Low viscosity telechelic PDMS, such as 50 cSt and 100 cSt, formed a material showing a gradient in elasticity through the film thickness direction (data not shown). This occurs as GOs (i.e., the highly functional crosslinkers) settled to the bottom of the PTFE dish, resulting higher crosslink density at the bottom while essentially leaving the top as a viscous amine terminated PDMS liquid. A similar phenomena was observed previously by Clarizia and coworkers for PDMS/zeolite composite materials [37]. This can be avoided by using telechelic PDMS with higher molecular weight and viscosity to slow or prevent settling of GO; these observations could be exploited for tailoring these materials further in the future.

### 3.2 Physical properties of telechelic PDMS/GO elastomers

Even though extensive information on the formation of the crosslinked network was obtained from FTIR and gel content measurements, rheological and mechanical properties provide additional understanding of both the elastomer and its precursor. Thus, the dynamic modulus for PDMS N1000/1 wt % GO sol and elastomer samples were investigated in more detail as a function of % strain (Fig. 3 (a)), frequency (Fig. 3 (b)), temperature (Fig. 3 (c)), and during stress relaxation for 10% and 20% strain (Fig. 3 (d)).

As shown in Fig. 3 (a)-(c), the elastomer exhibited a storage modulus ($G'\prime$) that was approximately an order of magnitude higher than the loss modulus ($G''\prime$) over a wide range of strains, frequencies, and temperatures, indicative of stable, solid-like viscoelastic behavior. In contrast, the PDMS/GO uncrosslinked sol exhibited liquid-like behavior ($G''\prime$ being higher than $G'\prime$) over the range of interest and even showed considerable changes across both the frequency and temperature sweep, which is clearly distinguishable from that of the elastomer sample. A stress relaxation test also confirmed the elastomers as viscoelastic solids, which is in agreement with the aforementioned rheological observations shown in Fig. 3 (a)–(c). After relatively short relaxation times, the stress relaxation curve reached an asymptotic equilibrium stress of approximately 20 kPa. On the other hand, the uncrosslinked liquid sol composites exhibited complete stress relaxation within a few seconds.

The mechanical integrity of the elastomer was also investigated using a universal tensile test. As expected, increasing the GO concentration resulted in a higher tensile strength while reducing the elongation at break (see stress–strain curve in Fig. 4 and data tabulated in Table 2). It is noteworthy that the precursor PDMS and PDMS/GO sol are liquids, and it was therefore impossible to perform such experiments on these samples. This result highlights the effect of chemical crosslinks between the polymer and GO, where other studies on physically mixed thermoplastic polyurethane/octadecylamine functionalized GO composites showed no reinforcement at loading levels below 2.5 vol % [38]. Although the gel content for high $M_n$ PDMS (N2000) composites was slightly lower than that of low $M_n$ PDMS (N1000, related data in Table 1), the N2000 samples exhibited moderately higher tensile strength. We hypothesize that a major contributing factor could be related to trapped entanglements between the reacted PDMS chain ends that could act as physical crosslinks within the network increasing the tensile strength.

In general, the physical and mechanical properties of the telechelic PDMS/GO elastomers are very important aspects when considering their use as gas barrier membranes. Depending on the specific application, dense polymer gas barrier membranes may be subjected to high pressures (over 10 atm) with high gas flux penetrating through the thin film membrane [39]. It is critical for
the membrane to withstand both external pressure and gas flow without forming pinholes or rupturing completely.

3.3. Application as an effective gas barrier membrane

There is a strong demand for enhancing the gas barrier properties of polymer nanocomposite materials for prolonging lifetimes of organic electronics or packaged food, among other applications. One of the common approaches to improve the gas barrier properties is to optimize the dispersion of the filler material using suitable processing procedures. Most of the polymer/GO membrane research has involved preparation of samples by solution mixing, in situ polymerization, and melt mixing [20]. While the high barrier performance of these materials has been demonstrated previously by physically mixing two or more components to obtain synergistic properties, a number of recent studies have focused on coating a thin layer of graphene or graphene derivative on top of a flexible substrate, or preparing a free-standing graphene derivative membrane [21]. Such approaches accordingly concentrate the graphene forming a denser layer of gas impermeable filler but they can introduce new challenges such as requiring careful handling to prevent the filler from fracturing or delaminating from the substrate. Exploiting the fact that the telechelic PDMS/GO elastomers exhibit excellent mechanical integrity with the PDMS component effectively encapsulating and protecting the gas impermeable GO from external stress and environmental damage, we envisioned that this material would perform well as a gas barrier or separation membrane.

PDMS (and closely related derivatives) are common materials
used in commercial gas/vapor separation membranes [39]. It is well accepted that by incorporating less permeable fillers (in this case GO) into the composite matrix, the gas barrier properties of the material should increase by introducing tortuous pathways through which the gas permeates [38,40–42]. To quantitatively measure the permeation of various gases, a single gas permeation test was performed, as shown in Fig. 5 and tabulated in Table S2. As a reference case, neat PDMS elastomer was synthesized and tested (Fig. 5(a)), which exhibited identical gas permeability performance to that reported previously by others [39]. For the various gases tested, the gas permeability for both neat PDMS and telechelic PDMS/GO composite elastomer increased in the order of N2, O2, H2, CH4, and CO2. Strongly sorbing penetrants, such as CO2, had the tendency to plasticize the polymer matrix and thereby slightly increase the permeability with increasing pressure. Other low-sorbing gas molecules had less effect on permeability with increases in feed pressure [39]. Reacting 1 wt % GO (0.43 vol %) with telechelic PDMS to form the elastomeric matrix resulted in an average reduction of ~45% in N2, O2, CH4, and CO2 single gas permeabilities. Osman and coworkers achieved a 30% reduction in O2 transmission rate at 3 vol % for physically mixed polyurethane/alumino-silicate nanocomposites [43] while others have shown a 30–50% reduction in single gas permeation for polymer mixed with 1.6–1.8 vol % thermally reduced graphene oxide composites (Table S3, comparing the gas barrier performance of different composite systems) [44]. Finally, as shown in Table 3, the gas selectivity values for the telechelic PDMS/GO composite elastomers were more or less similar to that of the neat PDMS elastomer.

One could speculate that the thermal properties of the neat PDMS and telechelic PDMS/GO elastomer could contribute to differences in their gas permeability data, especially given that the reaction between PDMS and GO might be anticipated to increase the glass transition temperature; Fig. S3 shows that the glass transition temperature (Tg) and melting temperature (Tm) of both materials were essentially identical. This clearly indicates that the significant decrease in gas permeability upon the addition of GO is mostly due to the impermeable GO dispersed phase, which constrains gas molecules forcing them to migrate through irregular amorphous regions of the composite material, and not by the reduction in chain segment mobility in the interstitial amorphous phase. It is evident that incorporation of higher loadings of GO (under the constraint that higher levels of this reactive GO component still allows for formation of mechanically robust elastomers) and enhanced alignment of the dispersed GO platelets would provide better gas barrier performance. Research related to these aspects, accompanied with efforts to model the relevant processes, is currently being investigated and will be reported in the near future.

4. Conclusions

A facile method to prepare elastic polymer/GO composite materials using telechelic polymers was developed. By taking advantage of simple chemistry between amine terminated telechelic polymers and GO, it was possible to transform liquid-like precursor materials into solid, robust elastomers by incorporating less than 1 vol % GO into the system. The GO acted both as a filler material that significantly reinforced the mechanical properties and as a multifunctional crosslinker forming chemical bonds with the polymer establishing the final macromolecular network. The elastomers prepared in this work were highly crosslinked (>75 wt % gel) and could be extended up to 300% when mechanically stretched. The rheological studies revealed that the elastomer is stable to external stress over a wide range of strain, frequency, and temperature. The elastomer showed ~45% reduction in various gas permeabilities by incorporating 0.43 vol% of GO, which highlights the use of this material as an effective gas barrier membrane. Furthermore, this simple, potentially cost-effective materials design approach should enable more versatile usage of polymer/GO composite elastomers in many commercial applications, including as gas barrier or separation membranes.

Author contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

Table 3

<table>
<thead>
<tr>
<th>Material</th>
<th>CO2/N2</th>
<th>CO2/CH4</th>
<th>O2/N2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neat PDMS</td>
<td>10</td>
<td>3</td>
<td>2</td>
</tr>
<tr>
<td>N1000/1 wt % GO</td>
<td>10</td>
<td>4</td>
<td>2</td>
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Fig. 5. Single gas permeability of (a) neat PDMS and (b) telechelic PDMS N1000/1 wt % GO telechelic PDMS/GO elastomers.