

Graphene Oxide Aerogels with Ultra-Low Thermal Conductivity

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Abstract

Graphene oxide (GO)-based hydrogels and reduced graphene oxide (rGO)-based aerogels were synthesized and their thermal transport properties were characterized using steady-state and infrared thermography methodologies. GO was produced via a modified Hummers' method and characterized by X-ray diffraction, Raman spectroscopy and X-ray photoelectron spectroscopy analysis. Self-assembled GO hydrogels exhibited thermal conductivity equal to 1.50 W/m·K, attributed to the formation of continuous thermally conductive networks at relatively high GO concentrations. In contrast, ultralight rGO aerogels with porosity approaching 99% demonstrated thermal conductivity of 0.021 W/m·K, which is 26% less than currently reported values for similar systems. The observed effect was attributed to extensive phonon scattering and suppressed solid and gaseous heat transport. The results highlight the strong dependence of thermal behavior on the microstructure and porosity of graphene-derived gels and demonstrate their potential for thermal management and insulation applications.

1. Introduction

The ongoing global energy crisis, related to rising energy demands, exhaustion of fossil fuel resources, and necessity to minimize greenhouse gases emissions, has spurred the development of new energy-saving materials [1], [2]. In a most highlighted case, buildings alone, currently account for approximately 36% of global energy consumption, with heating and cooling representing the dominant share of this demand and heat, in specific, accounting for a massive 74% of the total energy consumption in the buildings sector [3]. The major share of these enormous energy demands are thermal losses stemming from poor thermal insulation in buildings [4]. The main problem concerns the high thermal conductivity of cement-based building materials that are widely used in modern construction [5], [6]. Hence, the development of lightweight materials with low thermal conductivity has become a critical research priority for reducing energy consumption and enhancing sustainability [7]. Among the emerging solutions, aerogels have attracted considerable attention because of their highly porous nanostructure, ultra-low density, and outstanding thermal insulation performance, making them promising candidates as prospective energy-saving materials for next-generation energy-efficient construction and insulation applications [8], [9].

Among the different fillers proposed in literature for cement, nanocarbons such as graphene oxide (GO) and its reduced variant, rGO, have attracted extensive scientific attention due to their exceptional mechanical, optical, electrical, and thermophysical properties [10], [11], [12]. GO contains abundant oxygen functional groups, which enhance its dispersibility in aqueous media and enable tunable chemical reactivity [13]. Upon reduction, the resultant rGO exhibits improved electrical and thermal transport behavior [14][15]. A typical extended range of thermal conductivity regarding GO-based structures is 0.2–70.0 W/m·K [16][17], while correspondingly for rGO-based is 1.8–172.5 W/m·K [18][19]. These characteristics have driven the integration of graphene-based nanostructures into numerous applications, including energy generation, water treatment, polymer and cement reinforcement [20], [21][22], [23][24].

One of the most interesting categories of graphene-related materials is the GO-based gels which can be classified mainly into hydrogels and aerogels [22]. Three-dimensional GO hydrogels formed through self-assembly mechanisms and their derived rGO aerogels represent a particularly promising class of materials mainly due to the hierarchical porosity, ultralow density, and high surface area of the latter ones [22]. The self-assembly method is driven by non-covalent interactions such as hydrogen bonding, van der Waals forces, and π - π stacking, leading to monolithic and compact structures without the need for polymeric crosslinkers or additives [24]. Although GO exhibits significantly lower intrinsic thermal conductivity compared to pristine graphene [8][11], due to the presence of oxidative groups and structural defects, its self-assembled hydrogels can exhibit significantly higher macroscopic thermal conductivity values than its pristine counterparts, of the order of 5 or even to 10 W/m·K [27]. This is mainly attributed to the continuous and dense microstructure that results from the solidification of the hydrogel, which allows efficient heat transfer along and between GO sheets, while such hydrogels can contain more than 90% water [28].

The thermal reduction of GO hydrogels produces lightweight rGO aerogels with porosity exceeding 99% [29]. The morphology of the aerogels, dominated by air-filled interconnected pores and a thin carbon framework, endows extremely low thermal conductivity ($\sim$ 0.004–0.05 W/m·K), making them attractive candidates for thermal insulation applications [30]. These values are comparable to, and in some cases lower than those of benchmark thermal insulation materials currently used in the construction and aerospace sectors [31]. For example, conventional expanded polystyrene (EPS) and extruded polystyrene (XPS) typically exhibit thermal conductivities of approximately 0.030–0.040 W/m·K, mineral wool and glass wool generally range between 0.032 and 0.045 W/m·K [32], while silica aerogels, regarded as state-of-the-art super-insulating materials, commonly display values of about 0.013–0.020 W/m·K [8]. Additionally, ceramic materials like SiC aerogels have been reported with conductivity equal to 0.039 W/m·K [33]. The exceptionally low thermal conductivity reported for graphene-derived aerogels therefore places them among the most promising lightweight super-insulating materials, while simultaneously offering additional advantages such as low density, mechanical resilience and electrical functionality.

Recent studies have investigated thermally conductive hydrogels [34], mechanically reinforced hydrogels [35], and nitrogen-doped aerogels with enhanced elasticity and multifunctionality [36]. On the other hand, the relationship between GO concentration, microstructure of the resulting aerogel, and measured thermal conductivity, remains insufficiently explored. Several studies have examined the thermal diffusivity of GO nanofluids [37] and the effect of GO flake size on thermal conductivity [38], yet only limited information is available on the quantification of the thermal behavior of rGO aerogels produced directly from GO hydrogels. Additionally, most reported characterization methods require specialized, high-cost instrumentation [39], [40][30], and accessible methodologies for assessing ultralight graphene aerogels remain scarce.

This work aims to contribute to the understanding of structure–property relationships of GO-derived gels and to provide a straightforward methodology for evaluating their thermophysical performance. To this end, GO was synthesized and used as starting material in the gels. It was subsequently characterized by X-Ray diffraction, Raman spectroscopy and X-Ray Photoelectron Spectroscopy. GO films were prepared after the drying of GO solutions at 50°C. GO-based hydrogels were synthesized by processing of stable colloidal GO suspensions at 90°C without any other reactive agents or polymers. rGO-based aerogels with controlled porosity were prepared by thermal treatment of self-assembled, chemical crosslinker-free, GO-containing hydrogels and using a green approach for their drying, the ambient-pressure drying method, while they were characterized by Scanning Electron Microscopy. Next, the temperature profiles during cooling of GO films with different concentrations were recorded, and the heat capacity of both GO-based hydrogels and aerogels was also estimated. For the needs of thermal conductivity calculation regarding the three-dimensional hydrogels and aerogels, an assembly of a custom-built experimental setup is presented. Thermal conductivity of the aerogels, and density-based calculations were employed to estimate the rGO-to-air volume ratio, which was found to be approximately 0.1% rGO and 99% air—consistent with literature reports [41]. In this way, the as-prepared graphene-related materials and the adopted methodology are suggested as a novel and efficient solution for the thermal assessment of the nanofillers before being added in cementitious matrixes.

The work demonstrates that GO-based gels can exhibit broad thermal conductivity range, extending from relatively highly conductive hydrogel structures to ultralow-conductivity aerogel architectures, depending primarily on their microstructure, density, and porosity. The study also proposes a practical and experimentally accessible methodology for thermophysical characterization using infrared thermography, modified DSC analysis, and a custom steady-state thermal conductivity measurement setup, avoiding the need for highly specialized instrumentation. Most importantly, the results obtained are not limited by the nature of the target application, a fact which enables exploitation of the developed materials in a wide range of thermal management tasks.

2. Experimental Section

2.1 Materials and samples

2.1.1. Preparation of Graphene Oxide dispersion

In the present work, graphene oxide was synthesized via a modified Hummers' method which included a graphite pre-oxidation step [42]. To ensure complete oxidation, graphite powder was initially pre-oxidized in a mixture of sulfuric acid, phosphorus pentoxide, and potassium persulfate at 80°C for 1 h before being treated with potassium permanganate in concentrated sulfuric acid under controlled temperature (35–40°C). Following oxidation, the suspension was diluted with deionized water and treated with hydrogen peroxide for termination of the reaction. Residual metal ions and acids were removed by separation through cellulose membranes. The obtained GO was dispersed in deionized water for the formation of stable colloidal suspensions of controlled concentrations [43].

2.1.2. Synthesis of Graphene Oxide hydrogels

Starting from the GO which was prepared as described in the above paragraph, three-dimensional (3D) graphene oxide hydrogels were synthesized using the self-assembly method, which relies on the spontaneous organization of GO sheets into three-dimensional interconnected networks [44]. Therein, the aqueous GO suspensions, comprising more than 90% water by weight, were poured in Teflon molds with volumes of 40 ml and diameters of 6 cm, for uniformly shaped structures, as well as in small glass vials for control of sample dimensions. The filled molds and vials were heated in an oven at 90°C for three days, which enabled gradual water evaporation as well as self-assembly of GO sheets. For each formulation, a total volume of 25 mL was prepared which was subsequently divided into three equally sized samples using a precision cutter to ensure accuracy and repeatability. Initially, the physical dimensions and mass of the samples were measured using a Vernier caliper and a precision analytical weight meter. The resulting hydrogels maintained the cylindrical shape of the vials and the disc-like shape of the Teflon mold. Through the process, GO sheets were rendered hydrophobic and interacted through π – π stacking, hence resulting in the formation of robust 3D hydrogels [21].

2.1.3 Synthesis of reduced graphene oxide aerogels

rGO aerogels were synthesized from graphene oxide (GO dispersion, as already described) and ascorbic acid as primary reagents. Initially, the components were dissolved and thoroughly stirred to obtain homogeneous mixtures. The solution was then heated at 95°C for one hour, for induction of partial GO reduction and hydrogel formation [24]. At this point, it is very important to separate these hydrogels which are partially reduced GO thanks to the use of ascorbic acid, according to Pham et al. [45], from the neat GO hydrogels which did not contain any reducing agent (previous paragraph). This intermediate partially reduced GO hydrogel was subsequently cooled to -27°C and maintained at this temperature for 24 hours to stabilize the 3D structure of the material. Following thawing at ambient temperature, the hydrogel was subjected to an additional heat treatment at 95°C for five hours, for further GO reduction and enhancement of gel structure [45]. The product was washed extensively for removal of impurities and adjustment of the pH. The final graphene aerogels were obtained after complete removal of water, which was achieved by drying at 60°C for 24 hours. A mean apparent density equal to 7 mg/cm³ has been obtained for these macrostructures.

The exceptionally high porosity obtained in the present work can be mechanistically attributed to the synergistic combination of self-assembly, mild chemical reduction, and controlled ambient-pressure drying [24]. During hydrogel formation, graphene oxide sheets spontaneously organize into a three-dimensional interconnected network through π - π interactions, hydrogen bonding, and van der Waals forces, thereby generating a mechanically stable porous scaffold before drying [46]. The subsequent reduction with ascorbic acid partially restores the graphitic structure while avoiding the rapid gas evolution and framework shrinkage that can occur under harsher reduction conditions. Most importantly, the ambient-pressure drying step removes water gradually, which reduces capillary stresses acting on the pore walls and helps preserve the pre-formed cellular architecture [47]. Previous studies on graphene xerogels and aerogels prepared by ambient-pressure drying have shown that the maintenance of an interconnected hydrogel skeleton during solvent evaporation is critical for minimizing pore collapse and retaining a large fraction of the original pore volume [45][22]. As a result, the present synthesis route yields an ultralight rGO aerogel composed predominantly of interconnected air-filled pores separated by a thin graphene framework, leading to porosity approaching 99% and correspondingly suppressed heat transport through both the solid and gaseous phases.

2.2 Characterization of GO

2.2.1. Spectroscopic and Morphologic characterization

X-ray diffraction (XRD) analysis was performed using a Bruker D8 Advance diffractometer to investigate the crystalline structure and phase composition of the prepared samples. The instrument provides high-resolution diffraction patterns suitable for identifying crystalline phases and evaluating structural changes induced by the processing conditions. The measurements were performed in ambient conditions.

Raman spectra of the GO dispersion were collected with an InVia Reflex (Renishaw PLC, Gloucestershire, UK) MicroRaman equipment using a 514 nm laser excitation source. Dispersions were initially drop-casted and then spin-coated on SiO₂/Si wafers. In all experiments, representative spectra were collected by probing several random locations on each specimen using a grating with 1200 grooves per mm and an objective lens with $\times 100$ magnification power. The power of the laser beam was kept below 1 mW to avoid heating of the specimen at the probed location. Raman spectra analysis involved baseline correction and fitting of peaks with Lorentzian functions. When GO peaks were superimposed onto the peaks of the substrates, the necessary deconvolution process was applied. In this analysis, the Lorentzian components assigned to the substrates were held fixed, having had their parameters (position, full width at half-maximum) evaluated from the spectra of the bare substrates.

X-ray photoelectron spectroscopy (XPS), measurements were performed in a Bruker D8 Advance X-ray diffractometer (Bruker Corp., Billerica, Massachusetts, USA). Surface analysis measurements were performed in a UHV chamber operating at ca. 5×10^{-10} mbar, equipped with a SPECS Phoibos 100-1D-DLD hemispherical electron analyzer (SPECS Surface Nano Analysis GmbH, Berlin, Germany) and a non-monochromatized dual-anode Mg/Al X-ray source. XPS spectra were recorded using an MgK α source of 1253.6 eV photon energy and an analyzer pass energy of 10 eV, giving a full width at half maximum (FWHM) value of 0.85 eV for the Ag3d_{5/2} line. The analyzed area appertained to a round spot of 3 mm diameter. Atomic ratios were calculated from the intensity (peak area) of the XPS peaks weighted with the corresponding relative sensitivity factors (RSF) which were derived from the Scofield cross-section, considering the electron transport properties of the matrix (the inelastic mean free path (IMFP)). Spectra collection and analysis, including fitting, was performed using the software SpecsLab Prodigy, v. 4.12.0r49869 (SPECS Surface Nano Analysis GmbH, Berlin, Germany) was used. XPS peaks were deconvoluted with a sum of Gaussian-Lorentzian peaks after a Shirley-type background subtraction. Scanning electron

microscope (SEM) images were obtained using a LEO SUPRA 35VP with a maximum resolution of 1.5 nm at high vacuum and 20 KV, and 2 nm in low vacuum and 30 KV.

2.2.2. Thermographic characterization

The heat flow rate in GO was assessed by recording the cooling rates, to room temperature, of heated thin film samples of the material, via infrared thermography [48]. For this purpose, films were prepared by evenly dispensing 2mL of graphene oxide (GO) solutions, of varying concentrations, onto glass microscope slides. Solutions were subsequently dried at room temperature and then heated on a hot plate at 100°C, until a uniform temperature was achieved across their entire surface. Once thermal equilibrium was attained, film samples were immediately transferred onto a thick flat metallic baseplate, where temperature decrease was monitored from the top, with a high-performance HIKMICRO M30 thermal imaging camera (Hangzhou Hikmicro Sensing Technology Co., Ltd, Hangzhou, China), which embodies an uncooled microbolometric detector and operates in the long-wave infrared (LWIR, 7.5–14μm) spectrum with excellent sensitivity to temperature differences (noise equivalent temperature difference, NETD of less than 35 mK at 25°C) [49]. The thermal camera was placed at a height of approximately 0.5 m from the films and temperature variation was recorded by capturing thermographs of the samples at video mode (25 frames per second), during the whole time period until samples returned to room temperature. The collected cooling profiles allowed an indirect observation and determination of the thermal conductivity of samples and the evaluation of the effect of GO concentration on its thermal properties.

2.2.3. Measurement of heat capacity for the hydro- and aero-gels

Differential scanning calorimetry (DSC) is the relevant thermal analysis technique that provides the temperatures and heat flows associated with endothermic or exothermic chemical transitions in materials as a function of time and temperature. The specific heat capacity of the solid phase was measured by modulated DSC (MDSC). MDSC thermograms were obtained using a TA Instruments DSC Q100 Series (TA Instruments, Delaware, USA). The heating rate was 5°C/min in a temperature region from 60 to 115°C.

Since the aerogel densifies under the pressure of the sealed pan, the signal obtained corresponds to the rGO skeleton since the porous structure has collapsed. For the hydrogel, a GO film was dried thoroughly before sealed in the pan, so the measurement refers to the solid GO phase. The reversing heat capacity was fitted as a polynomial in temperature over the range of interest, and the mean value was obtained using:

$$\langle C_p \rangle = \frac{1}{\Delta T} \int_{T_c}^{T_h} C_p(T) \times dT$$

1

Because the calorimetric measurements refer to the solid phase only, the fluid filling the pore space was calculated as such:

$$\rho \times C_p = \frac{m_{solid} \times \langle C_{p,solid} \rangle + m_{fluid} \times \langle C_{p,fluid} \rangle}{V}$$

2

Where m_{solid} corresponds to the mass of the GO in the hydrogel or the rGO in the aerogel and m_{fluid} is the mass of the air in the aerogel and water in hydrogel. The specimens were cylindrical monoliths and were used throughout. Dimensions were measured using calipers and the mass on an analytical balance.

2.3. Thermal diffusivity and effective thermal conductivity

The effective thermal conductivity of the specimens was estimated from thermal diffusivity and volumetric heat capacity, according to:

$$k = \alpha \times \rho \times C_p$$

3

where k corresponds to thermal conductivity coefficient, α to thermal diffusivity, ρ is the density of the examined sample and C_p is the specific heat capacity. A transient approach was utilized to obtain the α while C_p was determined through MDSC, as already mentioned.

For the transient measurements, the specimens were mounted on top of a hot surface and covered with foam, with the lateral surface insulated so that heat flow could be treated as one-dimensional along the cylinder axis. Thermocouples in contact with the two faces

recorded the hot and cold face temperature. The specimens were mounted with the thermocouples, insulated and placed on top of the heating surface, while the temperature was constantly recorded.

Diffusivity

For one-dimensional conduction in a slab with a prescribed temperature at one face and a finite thermal resistance at the other, separation of variables gives a temperature field expressed as a series whose terms decay as $\exp(-\frac{\alpha \lambda_n^2 t}{L^2})$. The eigenvalues λ_n are the roots of:

$$\lambda \times \cot \lambda = -Bi$$

4

At sufficiently long times, the temperature response is dominated by the first eigenmode and the approach to steady state becomes a single exponential of time constant $\tau = L^2 / \alpha \lambda_1^2$. Thus, the diffusivity can be calculated from:

$$\alpha = \frac{L^2}{\tau \lambda_1^2}$$

5

Time constant

The cold-face temperature was fitted to:

$$T_c(t) = T_{c,\infty} - A \exp(-t/\tau)$$

6

where $T_{c,\infty}$ is the steady-state cold-face temperature, A is the exponential amplitude and τ is the characteristic time constant. The time constant τ is the time at which the exponential factor equals $e^{-1} = 0.368$, that is, at which the temperature has covered 63.2% of its total rise. By recording the rise of T at the cold side, a fit was employed to determine the whole temperature decay.

Biot number

Heat loss at the cold face is governed by the combined resistance of the insulation and the environment represented as an effective heat transfer coefficient h . Thus:

$$k \frac{T_h - T_c}{L} = h(T_c - T_{amb})$$

7

By rearranging the parameters, the Biot number can be calculated by:

$$Bi = \frac{h \times L}{k} = \frac{T_h - T_c}{T_c - T_{amb}}$$

8

3. Results and Discussion

3.1 Characterization of GO

3.1.1. Spectroscopic and Morphologic Characterization

X-ray diffraction (XRD) analysis (Fig. 1A) of the produced GO revealed two main diffraction peaks at angles of $2\theta \approx 10^\circ$ and $\approx 25^\circ$. The peak at ca. 10° corresponds to the (001) plane and indicates the presence of strongly oxidized layers with increased interlayer spacing (d -spacing), resulting from the incorporation of oxygen-containing functional groups (such as -OH and -COOH) and the adsorption of

water molecules [50]. In contrast, the peak near 25° is associated with the (002) plane of incompletely oxidized graphite or partially restored graphene layers [51].

The Raman spectrum of GO (Fig. 1B) exhibits two characteristic peaks: the D peak and the G peak. The D peak is attributed to defects and sp^3 hybridized carbon atoms introduced during the oxidation process, which disrupts the ideal sp^2 carbon lattice. The G peak corresponds to the intramolecular vibrations of sp^2 -bonded C-C atoms. Furthermore, the absence or weak presence of the 2D peak at ca. 2700 cm^{-1} is indicative of a multilayer structure and a significant degree of oxidation. In the present study, Raman analysis of the synthesized GO showed D and G peaks at 1348 cm^{-1} and 1593 cm^{-1} , respectively, with an intensity ratio $I_D/I_G \approx 0.87$. These values are consistent with literature reports attaining successful graphite oxidation and controlled defect introduction [52].

X-ray photoelectron spectroscopy (XPS) (Fig. 1C, D) revealed an elemental surface composition of the produced GO of 70.9% carbon (C) and 28.6% oxygen (O), corresponding to a O/C atomic ratio of approximately 0.40. This ratio pertains to a moderate degree of oxidation and satisfactory incorporation of oxygen-containing functional groups, while maintaining the structural integrity of the sp^2 carbon framework [53][54].

Scanning electron microscopy (SEM) observations (Fig. 2) reveal that the synthesized reduced graphene oxide (rGO) aerogel exhibits a highly porous three-dimensional cellular architecture consisting of interconnected graphene sheets that form an open and continuous network [22], [23]. At low magnification, the material is characterized by numerous irregular macropores with dimensions ranging from several tens to hundreds of micrometers, indicating successful preservation of the hydrogel structure during its drying process [55]. Higher-magnification images further demonstrate that the pore walls are composed of thin, highly wrinkled and folded graphene nanosheets interconnected through multiple junctions, generating a lightweight framework with a very small solid volume fraction [56].

3.1.2. Thermographic Characterization

Temporal variations of the average surface temperature of GO film samples (Fig. 3A-C) of two different GO concentrations are compared in Fig. 3D. Therein, in the initial cooling phase (100°C to 65°C), all curves overlap, suggesting that heat transfer is governed by bulk conductive phenomena rather than GO microstructural details. In the intermediate range (65°C – 40°C), clear differentiation emerges. The sample with the lowest concentration (1.43 mg/mL) reaches 40°C first, indicating faster thermal diffusion; conversely, the highest concentration sample (3.38 mg/mL) exhibits a pronounced delay, indicative of reduced thermal conductivity. This disparity is even more distinct in the final cooling stage, of temperatures between 40°C and room temperature (25°C – 27°C), where the 3.38 mg/mL sample shows the slowest cooling—a manifestation of poor thermal transport. Such behavior aligns with scarce recent reports in literature such as Khabibullin's et al. [57] who found that higher GO concentrations can inhibit heat flow due to enhanced phonon scattering, often caused by aggregate or network formation at elevated nanoparticle density. Jiménez-Pérez et al. [37] further highlight a nonlinear trend wherein initial increases in GO concentration raise thermal diffusivity until a critical threshold is reached, beyond which a plateau or even drops are noted due to aggregation or saturation effects. The present results align with such maximization at an optimal concentration, with higher loadings of the nanomaterial leading to reductions due to additional scattering and decreased phonon mean free path [25][37].

3.1.3. Measurement of GO's and rGO aerogel's heat capacity

The typical variation of GO's heat capacity as a function of temperature that was established via MDSC and used for calculating the thermal conductivity of GO hydrogels (Fig. 5A), is presented in Fig. 5B, wherein the black line represents the experimental data, and the red line represents a third order polynomial fit. The theoretical expression derived for the $C_{P,GO}(T)$ regression, subsequently used as input to the integral of Eq. (1), was:

$$C_{P,GO}(T) = 0.51137 + 0.03875 T - 4.68426 \cdot 10^{-4} T^2 + 1.60933 \cdot 10^{-6} T^3$$

9

As for the aerogels, samples were fully dry (Fig. 5D) and this did not inhibit the direct measurement, via MDSC, of their heat capacity variation with temperature, as was the case in hydrogels. Hence the $C_{P,GO}(T)$ function was established independently for these materials and is presented in Fig. 5E, wherein the black line represents the experimental data, and the red line represents a fifth order polynomial best-fit. The theoretical expression of the $C_{P,GO}(T)$ regression, subsequently used as input to the integral of Eq. (1), was calculated as:

$$Cp_{GO} (T) = -30.96489 + 2.04495 \cdot T - 0.05208 \cdot T^2 + 6.5408110 \cdot 10^{-4} \cdot T^3 - 4.0396 \cdot 10^{-6} \cdot T^4 + 9.80858 \cdot 10^{-9} \cdot T^5$$

10

The opposite temperature dependence of the heat capacity observed for GO (Fig. 5B) and the rGO aerogel (Fig. 5E) can be attributed to the profound structural and chemical transformations occurring during the reduction and aerogel formation processes. Pure GO (Graphene Oxide dispersion) contains abundant oxygen-containing functional groups (hydroxyl, epoxy, carbonyl and carboxyl groups) together with physically adsorbed water, both of which introduce additional vibrational degrees of freedom and progressively decrease the heat capacity with increasing temperature [26][27][15]. Following thermal reduction, a significant fraction of these oxygen functionalities is removed, and the material is converted into a highly porous three-dimensional rGO framework [22]. The resulting aerogel consists predominantly of air-filled pores separated by a sparse carbon skeleton, where heat storage is governed mainly by the reduced graphene framework and interfacial phonon interactions rather than by oxygen functional groups and confined water. Moreover, the highly disordered porous architecture and extensive phonon scattering modify the phonon density of states, leading to a different temperature dependence of the specific heat compared with pure GO [46]. Similar effects associated with the reduction of graphene oxide and the evolution of porous graphene architectures have been reported in previous studies investigating the thermophysical properties of graphene-derived materials [14][58], [59].

3.1.4. Transient response

In Figs. 5C and 5F, the transient response of the hydrogel and the aerogel are presented, respectively. The equations that fit the response of the materials are the following:

$$T_{c,aer}(t) = 70.707 - 33.381 \times \exp\left(-\frac{t}{32.988}\right)$$

11

$$T_{c,hyd}(t) = 81.081 - 49.406 \times \exp\left(-\frac{t}{72.787}\right)$$

12

3.2 Thermal properties of GO-enhanced hydrogels and aerogels

In the following Tables 1, 2 and 3, the properties of the examined specimens, the corresponding values of heat capacity and the resultant thermal conductivities are shown. In aerogel, the solid skeleton carries most of the heat capacity despite occupying only 1% of its volume, whereas in the hydrogel, water dominates.

Table 1
Specimen properties

Thickness, L (mm)	Aerogel	Hydrogel
	11	8.5
Diameter (mm)	13	15
Volume (cm ³)	1.46	1.502
Mass (mg)	37.5	1610
Density (mg/cm ³)	25.7	1072

Table 2
Volumetric heat capacity

$\langle C_{p,solid} \rangle (J/g \cdot K)$	Aerogel	Hydrogel
	0.916 (Eq. fitted C_p)	1.432 (Eq. fitted C_p)
$\langle C_{p,fluid} \rangle (J/g \cdot K)$	1.01	4.196
Solid mass (mg)	37.5	206
Fluid mass (mg)	1.43	1404
$\rho \times C_p (J/m^3 \cdot K)$	2.45×10^4	4.12×10^6

Table 3
Calculated thermal conductivity

$T_h (^\circ C)$	Aerogel	Hydrogel
	115	88
$T_C (^\circ C)$	70.71	81.08
$T_{amb} (^\circ C)$	28.5	28.5
Mean Temperature ($^\circ C$)	92.9	84.5
Bi (Eq. 8)	1.049	0.132
λ_1 (Eq. 4)	2.051	1.65
τ (s)	32.99	72.79
α (m^2/s)	8.72×10^{-7}	3.65×10^{-7}
k ($W/m \cdot K$)	0.021	1.5

Hydrogels in general contain approximately 90–93% water [60]. In this work, the samples were prepared with a GO concentration of 2.72 mg/mL, which is considered relatively high for GO-based gels [61]. This increased content contributed significantly to the formation of a tightly connected, continuous thermal conduction network, which limits the presence of empty regions within the material and minimizes the thermal resistance between the atomic sheets. The calculated value of the thermal conductivity coefficient obtained from the experimental data is equal to 1.50 W/m·K. This value lies between the usual range reported in literature for graphene oxide hydrogels (which is typically between 0.5 and 3 W/m·K) [29][62]. The relatively high thermal conductivity value measured is considered quite reasonable, taking into account that the preparation method promotes the homogeneous distribution and optimized interconnection of graphene oxide sheets, the high concentration of GO, which enhances the creation of a continuous thermally conductive network, and the microstructure of the hydrogel, which ensures limited thermal losses and enhances heat transfer through directed paths.

For the case of rGO aerogel, experimental measurements gave $T_c = 70$ °C and $T_h = 115$ °C. Under steady-state conditions, the thermal conductivity coefficient was calculated as $k = 0.021$ W/(m·K), a value very close to the average of aerogels with densities between 11.1 and 35.0 mg/cm³ [29], $k = 0.023$ – 0.026 W/(m·K). The measured thermal conductivity of 0.021 W/m·K is among the lowest values reported for graphene-based aerogels prepared by relatively simple chemical reduction and ambient-pressure drying [47], [63]. Although even lower values have been reported for ultralight graphene aerogels possessing substantially lower densities and highly optimized

pore architectures, the present material combines very low thermal conductivity with a facile synthesis route and an experimentally accessible methodology for quantitative thermophysical characterization.

Graphene aerogels exhibit exceptionally low thermal conductivity even though graphene itself possesses intrinsically high thermal conductivity. This phenomenon may be primarily attributed to the highly porous and disordered three-dimensional architecture of the aerogel network. Scientific studies have suggested that the ultralow density and porosity of graphene aerogels, which often exceeds 95–99%, dramatically suppress heat transport by limiting continuous phonon propagation pathways through the solid framework. In these materials, the graphene sheets form a tenuous interconnected skeleton separated by large volumes of air-filled nanopores, thereby reducing the effective solid conduction contribution. Moreover, the tortuous morphology and weak inter-sheet contacts introduce substantial interfacial thermal resistance, strongly scattering phonons and hindering efficient heat transfer across the network. Qiu et al. reported thermal conductivity values as low as approximately 4.7×10^{-3} – 5.9×10^{-3} W/m·K for ultralight graphene aerogels with densities near 4 mg/cm³, values even lower than the thermal conductivity of air under ambient conditions, emphasizing the dominant role of interface resistance and structural disorder in thermal transport suppression [46]. In general, increasing porosity reduces the number of solid heat-conduction pathways while simultaneously increasing pore tortuosity and phonon-boundary scattering, leading to progressively lower thermal conductivity. Table 4 concentrates several published previous works, including synthesis technique and conditions, density, porosity (air content) and thermal conductivity of the produced graphene/rGO aerogels to facilitate a direct comparison with the aerogels reported in the current work. Reviews on nanoporous aerogels further explain that highly porous aerogel structures suppress both solid-state conduction and gaseous conduction because nanoscale pores reduce the mean free path of gas molecules and weaken gas-phase heat transfer through the Knudsen effect [64], [65]. Graphene-based aerogels can therefore offer promising thermal insulation potential due to their combination of ultralow density, extremely high porosity, and correspondingly low thermal conductivity [66].

Table 4
Comparative table with physical properties and thermal conductivity for the present work and other published.

Reference	GO/rGO	Synthesis method	Drying method	Density (mg/cm ³)	Porosity (%)	Thermal conductivity (W/m·K)
Xie et al., Carbon (2016) [46]	rGO aerogel	Hydrothermal self-assembly	Freeze drying	~ 4	> 99.8	0.0047–0.0059
Huang et al., Nanomaterials (2019) [29]	rGO aerogel	Thermal-flow self-assembly	Freeze drying	11–35	98–99.5	0.023–0.026
Oh et al., Adv. Funct. Mater. (2021) [30]	Graphene foam	Compartmentalized assembly	Freeze drying	2.4	> 99	0.00575
Present work	rGO aerogel	Ascorbic acid reduction + hydrogel formation	Ambient-pressure drying	25.7	99	0.021

Conclusions

This work demonstrated the strong dependence of the microstructure and porosity of graphene-derived gels on their thermal transport properties. Self-assembled GO hydrogels exhibited relatively high thermal conductivity (1.50 W/m·K), attributed to the formation of continuous heat-conduction pathways between well-dispersed graphene oxide sheets. In contrast, ultralight rGO aerogels with porosity approaching 99% achieved an exceptionally low thermal conductivity of 0.021 W/m·K, highlighting the dominant role of pore architecture, interfacial thermal resistance, and phonon scattering, in suppressing heat transfer.

Beyond material performance, the study proposes a practical methodology for evaluating the thermophysical properties of graphene-derived porous materials by combining infrared thermography, differential scanning calorimetry, and a custom steady-state thermal conductivity setup. The proposed approach provides an accessible alternative for the characterization of lightweight thermal insulation materials without relying on highly specialized instrumentation.

The combination of tunable thermal conductivity, low density, and controllable microstructure demonstrates the potential of GO-derived hydrogels and rGO aerogels for advanced thermal management and energy-efficient insulation applications. Future focus pathways include tailoring the pore architecture and degree of chemical reduction to further optimize thermal transport, evaluating the long-term

mechanical and environmental stability, and integrating the materials into multifunctional composites and cement-based systems for sustainable building technologies.

Declarations

Conflicts of Interest:

The authors declare no conflict of interest.

Author Contribution

G.G., G.T., S.M., G.P., A.K., N.K., and L.S. performed experiments for the synthesis and characterization of the graphene oxide-based materials. G.G. and S.M. drafted the manuscript, C.G. participated in the design and coordination of the manuscript, while K.D. designed, supervised all search procedures and revised the manuscript. All authors have read and agreed to the published version of the manuscript.

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Data Availability

All data supporting the findings of this study are available within the paper .

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Figures

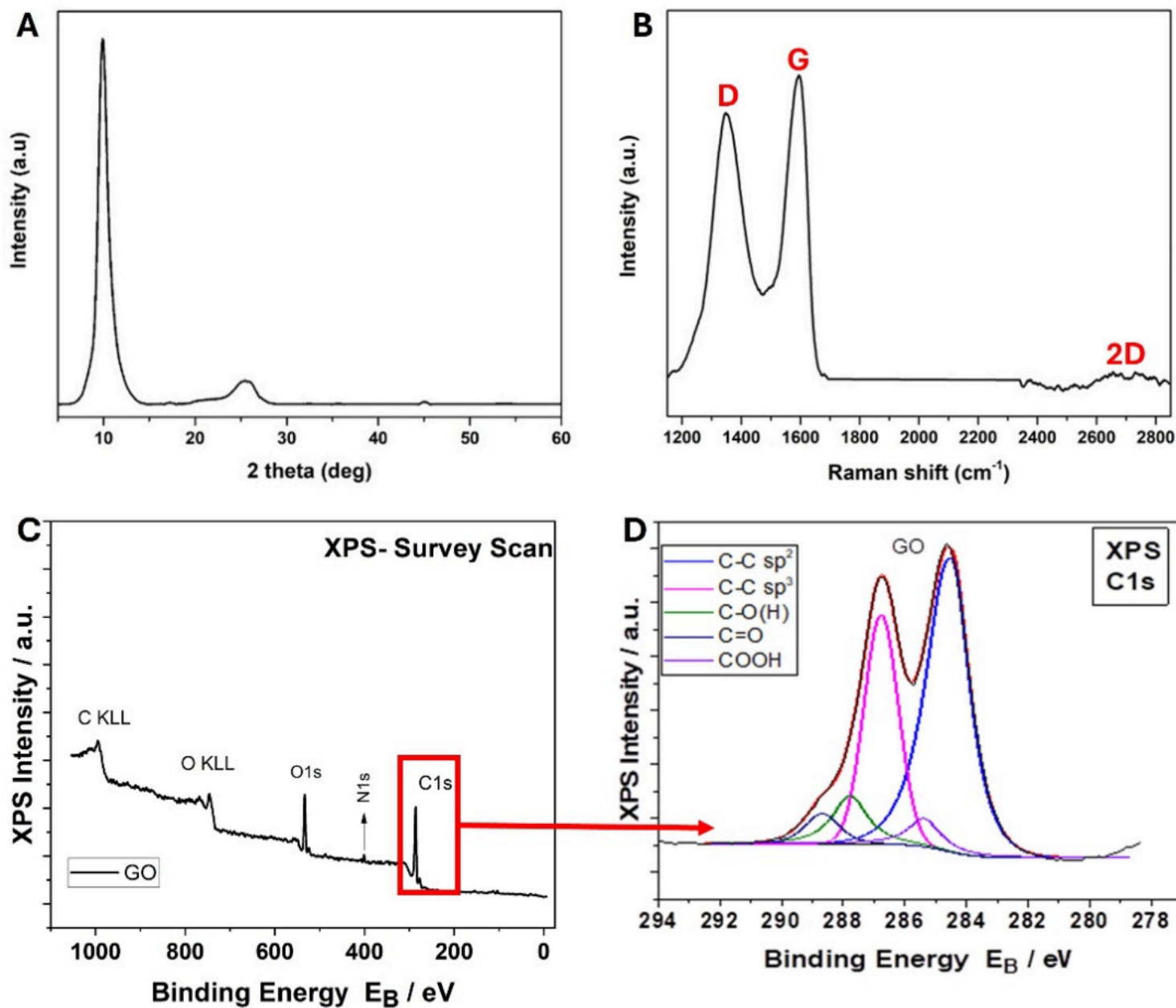


Figure 1

Spectroscopic characterization of graphene oxide (GO): (a) X-ray diffraction (XRD) pattern, (b) Raman spectrum with characteristic D and G peaks, (c) X-ray photoelectron spectroscopy (XPS) survey scan, (d) decomposition of the C1s region into individual bonds C-C(sp^2), C-C(sp^3), C-O(H), C=O, and COOH.

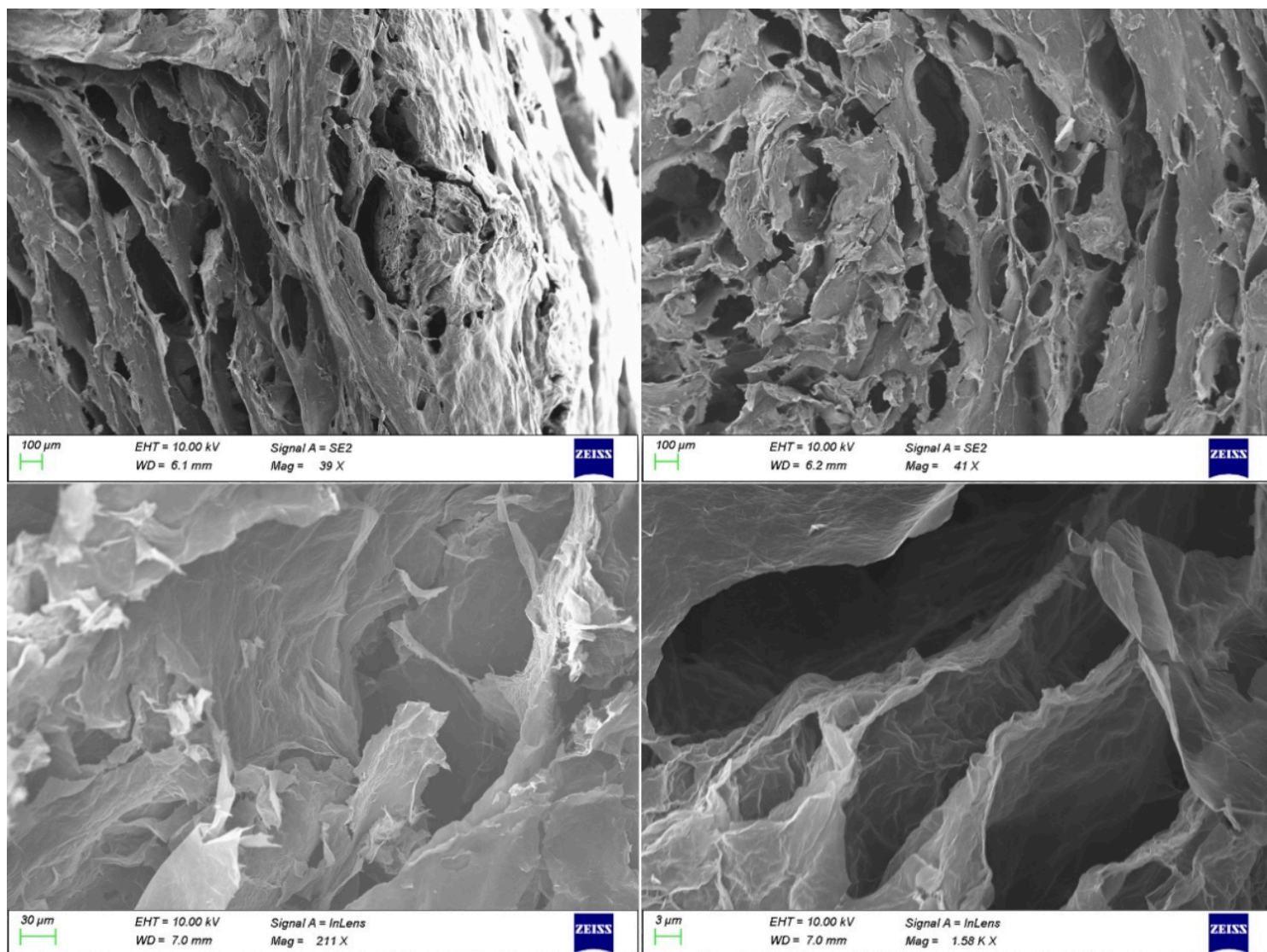


Figure 2

Scanning Electron Microscopy photos of the as-synthesized graphene aerogels, with different scale-bars.

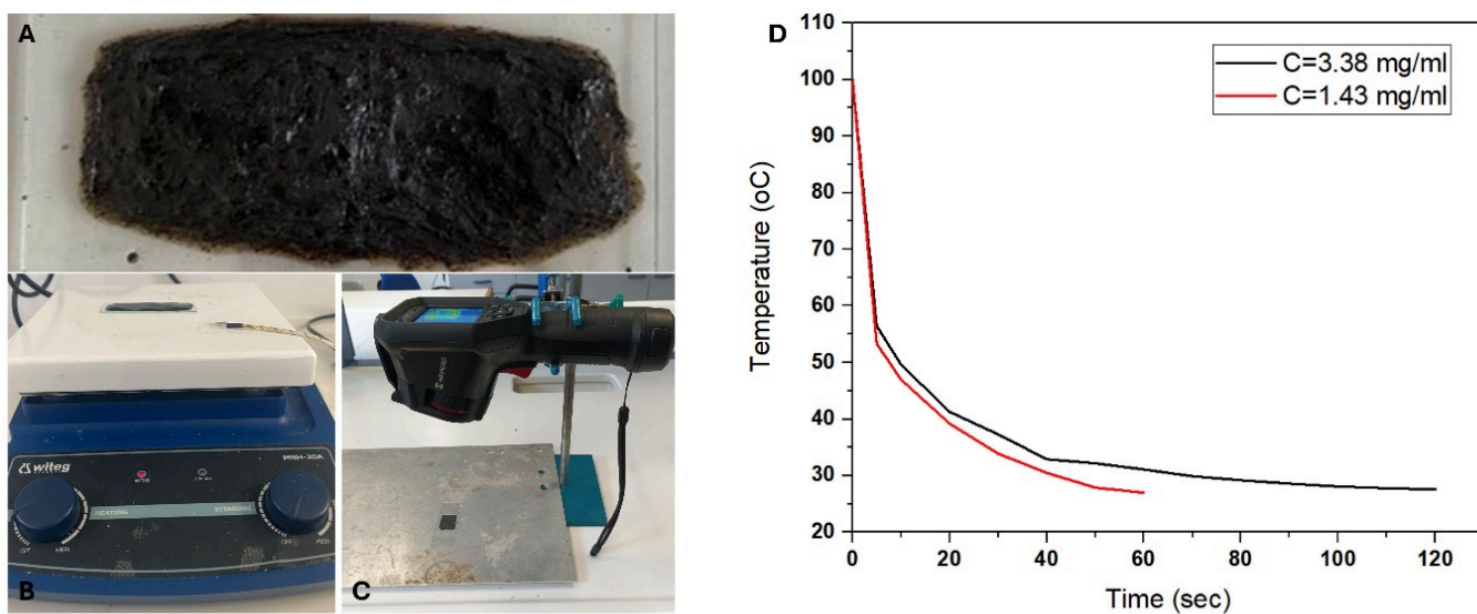


Figure 3

A) The resultant GO film after the drying of the starting GO dispersion, **B)** the GO dispersion was drop-cast onto glass microscope slide and dried mildly with the assistance of a heater, and **C)** the setup adopted for the collection of temperature profiles of GO films depicting the thermal camera which was standing directly above the sample. **D)** Cooling profiles of GO samples of varying GO concentration. The material with higher GO content (3.83 mg/mL, black line) cools down slower than its counterpart with lower GO content, indicating lower thermal dissipation.

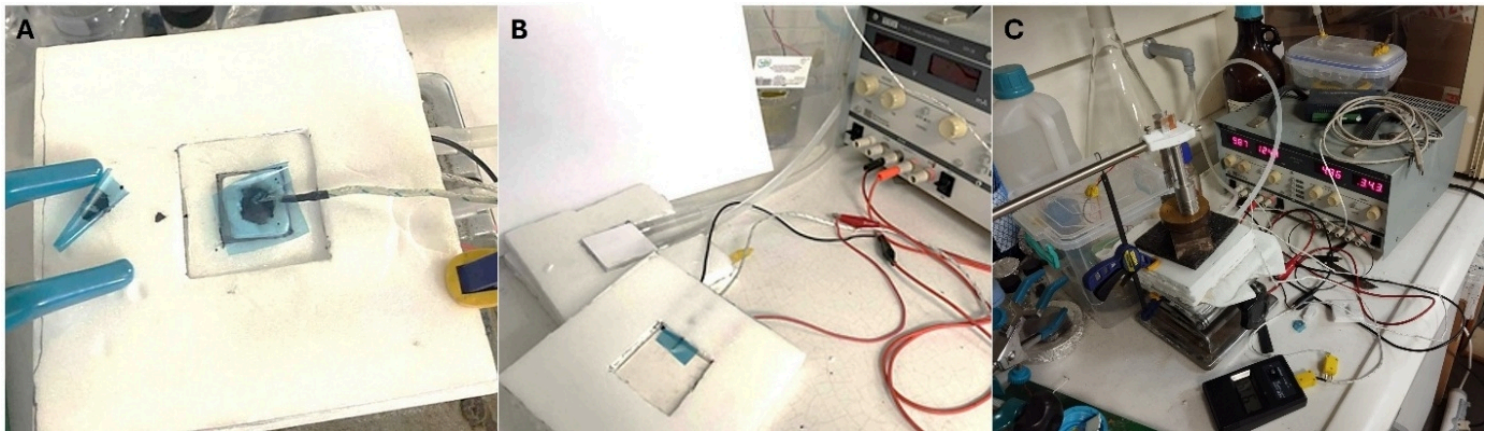


Figure 4

Experimental setup for measuring steady-state thermal conductivity. **A)** The heating setup includes a heating element attached to the upper surface of the sample, with thermal insulation around the perimeter to prevent lateral losses. **B)** Complete experimental setup, with DC power supply connection for heating via the Joule effect and cooling circuit at the base of the sample. **C)** For achieving a better heat isolation for our setup, some bronze objects were used to apply more pressure on the top. In this way, a thermally insulated and stable experimental setup was developed, which ensures accurate and reliable measurements.

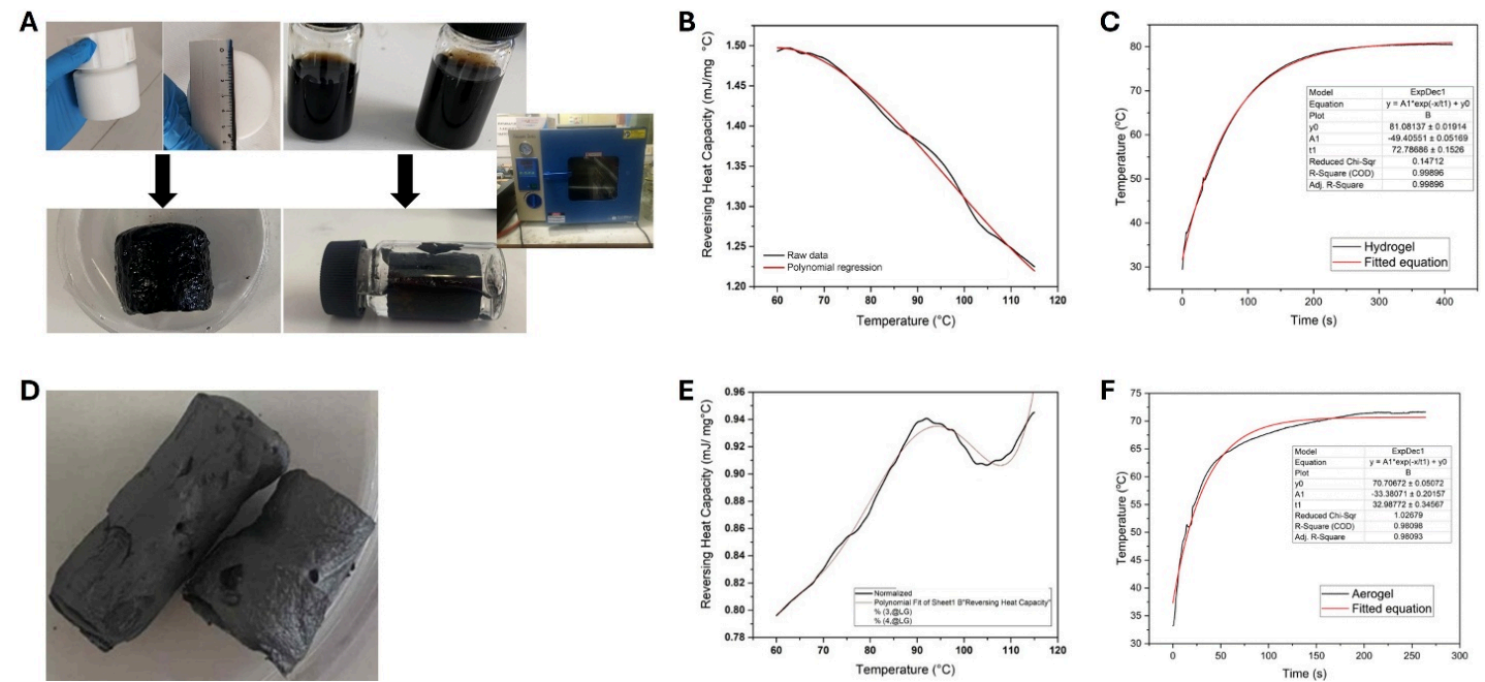


Figure 5

A) Formation of GO hydrogels. On the left, the used Teflon mold and the hydrogel of that shape are shown, while correspondingly on the middle, the glass vials and the cylindrical hydrogel are shown. For both cases, temperature of 90°C was applied and all samples were mounted in the furnace shown on the right side. **B)** Heat capacity curve for pure GO, as a function of temperature, as obtained from

MDSC thermal analysis. C) Transient response of the GO hydrogel, as obtained from MDSC thermal analysis. D) rGO aerogels and E) their heat capacity variation with temperature, obtained by MDSC thermal analysis. F) Transient response of the rGO aerogel, obtained by MDSC thermal analysis.