REVIEW ARTICLE

Graphene Intercalated Multifunctional Polymer Networks as Acoustic Absorbers for Underwater Applications

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Abstract: Multifunctional polymer networks fortified with the power of graphene and its derivatives as nano-inclusions have excellent sound absorption efficiency in broad frequency range, high loss factor, and matching impedance with that of water along with exceptional thermal, mechanical, and tribological properties are found to be the pre-eminent material for the underwater acoustic applications, particularly for the military tactics. To develop a stealthy underwater acoustic material, various factors need to be carefully considered, including matching acoustic impedance, glass transition temperature, loss factor, tan δ value, compression set and other mechanical properties, thermal stability, adhesion, and other tribological properties, which is briefly summarized in this review. Strategical development of hybrid nano-inclusions, viscoelastic polymer networks, nanocomposites as well as various interpenetrating polymer networks (IPNs), assiduous synthesis and surface modification of graphene are pivotal key approaches that need to be appraised. Simulation studies focusing on various potential models need to be developed for the feasibility studies and designing of the underwater acoustic material.

Keywords: Graphene; Polymer nanocomposite; Underwater acoustics; Sound damping; Interpenetrating polymer networks (IPNs); Viscoelastic polymer networks.

1. INTRODUCTION

Graphene, an extended honeycomb networked two-dimensional sheet which became the pivotal manifestation of nanotechnology, having sp^2 hybridized carbon confined in the hexagonal crystal lattice is the building block of some precious allotropes of carbon like graphite, carbon nanotube, fullerene, etc. In hindsight, the idea of graphene surged the world of science to explore one of the thinnest and strongest materials with long-range π-conjugation, a promising candidate for a wide spectrum of potential applications. Meanwhile, the plethora of material science, nanotechnology, electrical, and bioengineering helms hope in the astonishing properties of graphene on demand (Allen *et al.*, 2010; Zhu *et al.*, 2010). Graphene has captivated the colossal heads of researchers owing to its extraordinary unique properties like large specific surface area (Bonaccorso *et al.*, 2015), high tensile strength, extremely low electrical resistivity, exceptional thermal conductivity (Balandin, 2011; Dhawale *et al.*, n.d.; Pop *et al.*, 2012), high

Young's modulus (Lee *et al.*, 2008), ambipolar field effect (Shin *et al.*, 2010), good optical transparency (Nair *et al.*, 2008; Sheehy & Schmalian, 2009), excellent electronic properties (Castro Neto *et al.*, 2009), high carrier mobility (Bolotin *et al.*, 2008; Morozov *et al.*, 2008), quantum hall effect at room temperature (Novoselov *et al.*, 2007), chemical tunability and excellent acoustic properties (Li *et al.*, 2022), given in Table 1. Furthermore, there is a reliable multifaceted synthetic array of approaches to prepare like graphene-like top-down exfoliation method, bottom-up epitaxial growth, and catalyze-catalyze *in situ* growth on a substrate (Allen *et al.*, 2010; Huang *et al.*, 2012; Meyer *et al.*, 2007; Zhu *et al.*, 2016). Schematics of the skeletal structure of graphite layers and graphene nanosheet are shown in Fig 1a,b respectively, which is also evident from the transmission electron microscopy (TEM) image (Fig. 1c). Also, the electronic band structure of single-layer graphene, in which each unit cell of graphene contains two atoms, leads to the formation of two conical points (K & K'), where the electron energy (E) is linearly dependent on the wave vector, displayed in Fig. 1d. Thus, single-layer graphene behaves differently from other conventional metals which are used as semiconductors and show strong ambipolar electric field effect. Fig. 1e shows the sp2 hybridized carbon atom in a graphene nanosheet.

Property	Value	Ref
Tensile strength	125 GPa	(Lee *et al.,* 2008)
Young's Modulus	1.1 TPa	(Lee *et al.,* 2008)
Ductility	>20%	(Katsiropoulos *et al.,* 2022)
Electron mobility [a]	2×10^5 cm^2 /V s	(Bolotin *et al.,* 2008)
Thermal conductivity [a]	5×10^3 W/m K	(Balandin *et al.,* 2008; Ghosh *et al.,* 2008)
Light transmittance	97.7%	(Nair *et al.,* 2008)
Surface area	2630 m^2 /g	(Schedin *et al.,* 2007; Stoller *et al.,* 2008)
Electrical resistivity	~10^{-6} Ω.cm	(Katsiropoulos *et al.,* 2022)
Optical transparency	~97.7%	(Nair *et al.,* 2008; Sheehy & Schmalian, 2009)
Oxidation temperature	450 °C	(Li Liu *et al.,* 2008)
Permeability	Impermeable to liquid/gases; permeable to protons	(Berry, 2013; W. Yuan *et al.,* 2014)

[a] Measured at 25 °C.

Table 1. Physical and Chemical Properties of Graphene.

The first and foremost isolation of graphene by Andre Geim and Konstantin Novoselov of the University of Manchester triggered the interest in experimentalists and graphene found its way to polymer-based materials invigorating the beauty of nanotechnology, towards polymer nanocomposites. The phrase "nanocomposite" describes a category of composites where one of the elements is evenly distributed within the matrix at the nanoscale, which is on the order of one billionth of a meter (10^{-9} m). Nanocomposites found their way towards high-end applications like underwater acoustics due to their excellent ability to withstand harsh conditions, especially beneath the depths of the ocean, excellent sound-damping properties, and low cost as well as easy fabrication. Thus, nanocomposites took charge as excellent sound-damping material for under vehicles, which can also be used in warfare which is nothing but acoustic stealth technology (David *et al.*, 2022.).

Succinctly, underwater acoustics deals with the propagation of sound in water, which can be exclusively researched and developed to reduce the noise radiated from underwater vehicles or/and reduce the acoustic target strength of the underwater vehicle, which is shown in Fig 2. The former mainly focuses on the effect of sound waves that radiate from underwater vehicles on marine life, which can

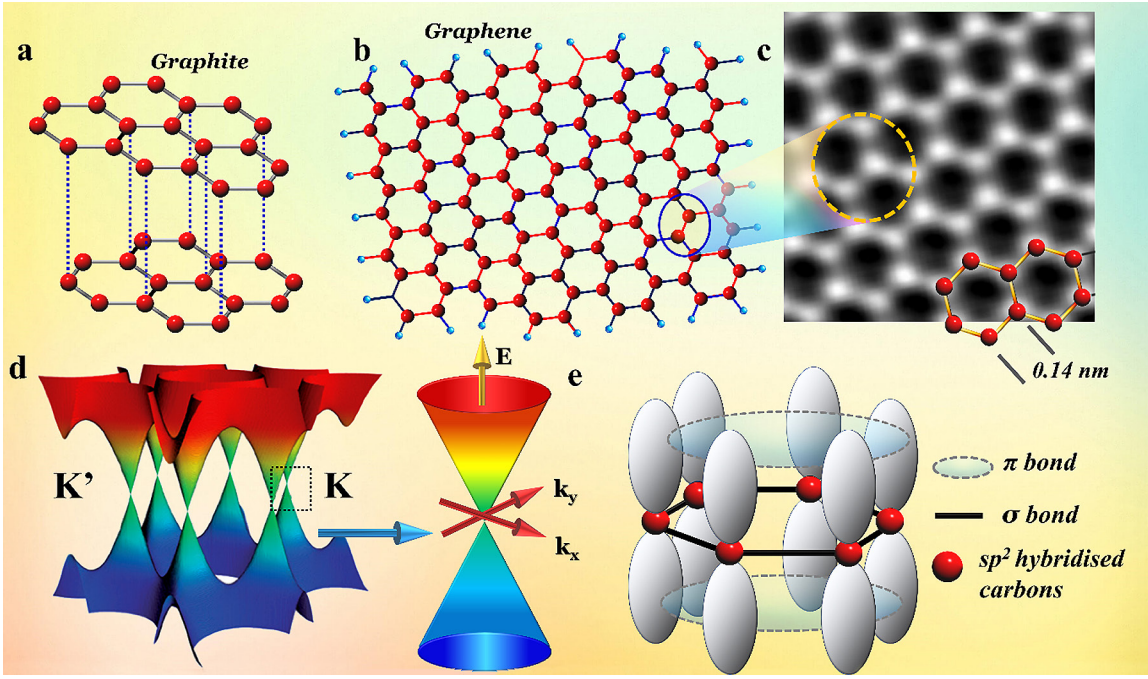

Figure 1. Schematic illustration on the: (a) framework of graphite layers, (b) structure of graphene nanosheet, (c) the high-resolution picture displaying a pristine and flawlessly structured graphene sheet synthesized using the substrate-free gas-phase method. The individual carbon atoms are portrayed as white in this image, (d) electronic energy bands of a solitary layer of graphene nanosheet, and (e) out-plane π orbitals as well as in-plane σ bond perpendicular to the plane of the graphene nanosheet(Dato *et al.,* 2009; Rao *et al.,* 2009).

be detrimental. Later one can be used for SONAR (Sound Navigation Ranging) applications in which the distance and direction of the target object (submarines, etc.) are detected and determined by the time taken for the sound waves to reach back to the SONAR system once it is emitted and hits on the target object. Thus, as a military tactic, the development of polymer-based materials with good sound damping, high loss factor, and matching impedance with water (Acoustic impedance of water = ~1.56 x 10^6 Rayl) is necessary for the underwater vehicles to invade the enemy territory without getting detected by the SONAR system. At the same time, the adhesion as well as water resistance of the surface coating also matter.

Polymer composites are known for underwater acoustics due to their matching acoustic impedance with water, excellent damping property along with good resistance towards ozone, weather, heat, abrasion, water, corrosion, and oxidation (Chen *et al.*, 2023; Huang *et al.*, 2016; Shaid Sujon *et al.*, 2021). Exceptional thermal/electrical insulation properties as well as economical and easy fabrication of the polymer composites, make it a multifunctional material that adds to various high-end applications. A critical and consolidated review on the development of graphene-based polymer composites pivoting around the tuning of filler-polymer systems' alignment and orientation, and the size and composition of filler that determines the sound damping property is important to brace up the researchers in the facile development of various polymer-based systems for the sonar waves absorption without compromising its mechanical, dynamic mechanical and thermal properties.

2. UNDERWATER ACOUSTICS VS. POLYMERIC MATERIALS

The story of underwater acoustic damping materials pivots around decades when the SONAR system was once introduced that makes use of active and passive array methods. In the active array method, the target object (submarines, ships, etc.) will respond by resonating and re-radiating the ultrasound that comes from the SONAR system and thus, the distance as well as the direction of the target object is detected. Foresee, as an effective strategy, the

active method aims to befog the location as well as the direction of the target by various brilliant techniques. Likewise, passive signals can also detect target objects but compared to the active method, the primary goal of the passive approach is to enhance the submarine's stealthy nature and thus reduce its detectability using methods such as quiet propellers and engines, streamlined hull designs, and surface treatments that minimize ultrasonic reflections. Moreover, passive materials have garnered more interest and broader applications due to their excellent performance-to-cost ratio, ease of manufacturing, and dependable service. The frequencies of underwater soundwaves can be counted between 10 Hz and 1 MHz and the SONAR system employed in military applications needs to detect targets effectively over a certain distance which limits its operational frequency range, typically falling in the range of 1-100 kHz, corresponding to wavelengths between 1.5 m-1.5 cm (Andrews, 2003). Since it is difficult for the underwater vehicles to surpass the enemy territory without penetrating deep into the seabed by making a noise below 1kHz. Above 100 kHz are also not focused because they are absorbed very quickly. Submarines were introduced to invade the enemy territory which can be driven deep underwater focusing on its stealthy behaviour. Starting from natural resources, underwater sound-damping materials found their way to advanced underwater acoustic materials, piezoelectric, and metamaterials, which can be used for active as well as passive SONAR systems. This was taken on grand by military forces even in World War II as submarines as well as anti-submarine vessels.

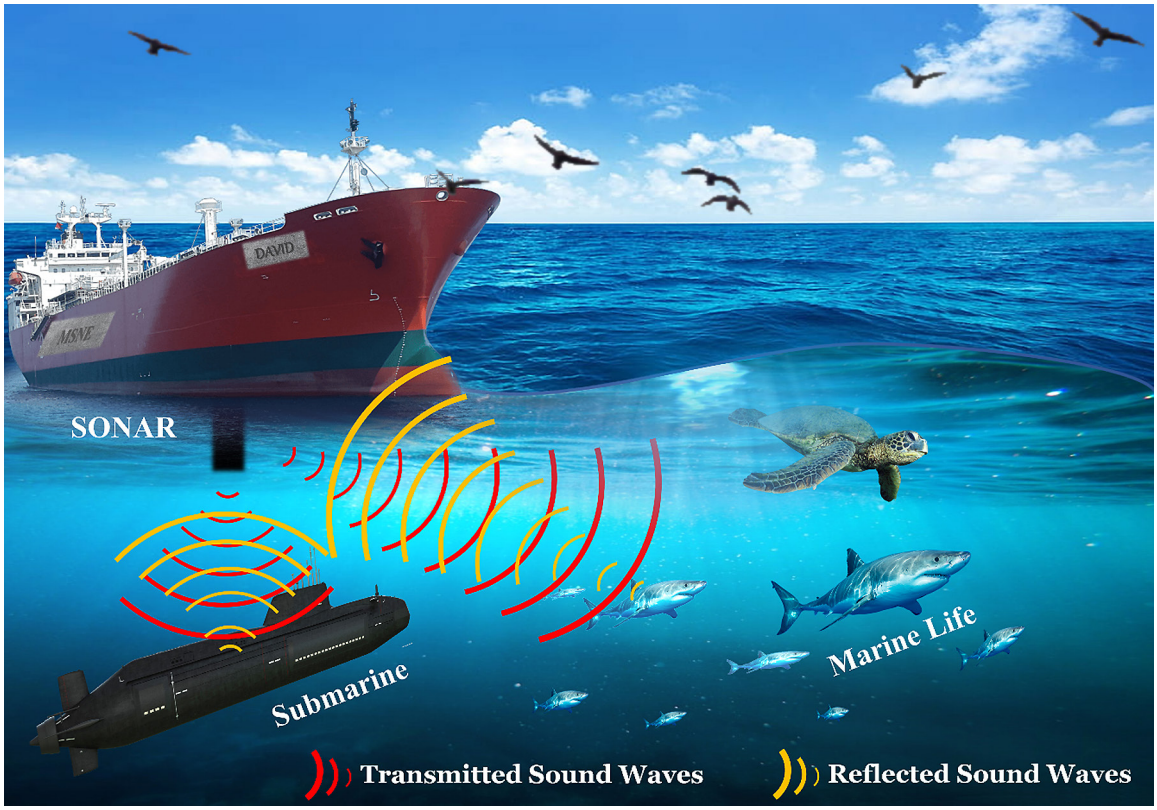

Figure 2. Illustrates the detection of the target object by SONAR system using sound waves.

Visco-elastic relaxation absorption, viscous absorption, and heat conduction absorption are the three main sound damping mechanisms associated with acoustic materials, shown in Figs. 3a,b. The plethora of polymer-based acoustic materials correlates with the visco-elastic relaxation absorption that results in the attenuation of sound waves. When the sound waves propagate through the material, molecules within the material will start vibrating and thus the increased movement of the molecular chain results in the development of internal friction. The energy dissipated will be converted to heat. In sound-damping materials, the energy is efficiently dissipated and converted to heat. Thus, the

characteristic properties required for the material to be acoustically stealthy include:

(i) matching impedance of the acoustic material with water, so that soundwaves, that travel all way from the sonar underwater towards the target, can enter into the acoustic material with sound damping property without being reflected;
(ii) the ability of the acoustic material to attenuate and disperse the sound waves that can be transmitted into the material, so that the sound waves that entered into the acoustic material can be effectively damped by effectively dissipating the energy by converting to heat and thus reduce the chances of the soundwaves to reach back to the SONAR system;
(iii) adhesion of the surface coating material demands an efficient metal-material bonding, that can reduce the damage of the propellent coating material due to cavitation. The acoustic impedance can be calculated using Equation 1.

$$\text{Acoustic impedance, } Z = \rho \cdot c \qquad \text{Eq. (1)}$$

Where 'ρ' is the density of the material and 'c' is the speed of the sound waves traveling in the medium.

For an ideal medium possessing infinite thickness and one-dimensional sound waves, the characteristic specific acoustic impedance (represented as Z_o) equals the product of the volumetric mass density (denoted as ρ) of the medium and the speed (designated as c) at which sound waves propagate within that medium (Fu *et al.*, 2021). The impedance of water is 1.56×10^6 Rayl. The noise reduction coefficient is another important parameter, which is a single numerical value ranging from 0.0 to 1.0. This value characterizes the average sound absorption capability of a material which describes the average sound absorption performance of a material in which the value 0.0 indicates the material doesn't attenuate but rather reflects the sound waves.

Sound waves can exclusively travel through elastic substances. When sound waves irritate the material, the polymer chain experiences repetitive compression and stretching on a micro/nanoscale. During this continuous and dynamic process, there is a delay in the strain compared to the stress, and this delay is characterized by a phase angle δ ranging from 0 to π/2 radians. The ratio of the imaginary part to the real part in this context is referred to as the tangent of the phase angle δ, commonly known as the loss factor, given in Equations 2 and 3. A higher loss factor is crucial for materials designed to absorb underwater sound.

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$$\tan \delta = E''/E' \qquad \text{Eq. (2)}$$

$$E^* = E' + E'' j \qquad \text{Eq. (3)}$$

Where E' is the measure of the energy stored elastically and E" is the equivalent energy loss in the form of heat during collision (shown in Fig. 3b), or else, the ratio of storage modulus with loss modulus. Thus, without any shadow of a doubt we can say that, the dynamic mechanical property E^* can be used to evaluate the sound-damping property and absorption capacity of the polymer for underwater acoustics, as shown in Fig. 3c. Higher the materials' loss factor (tan δ value), better would be the sound damping property, thus polymer-based materials are likely to be having higher sound damping property. The sound absorption coefficient (α) is the ability of the material to dampen the sound waves and it is quantified as the ratio of absorbed sound energy (E_a) to the incident sound energy (E_i), expressed in Equation 4.

$$\alpha = E_a / E_i \qquad \text{Eq. (4)}$$

Polymeric materials have captivated colossal heeds with its acoustic damping performance due to their close acoustic impedance with water, showing excellent sound damping properties by the visco-elastic relaxation absorption due to their chain flexibility that correlates with its elasticity. Also, polymeric materials are having high tan δ compared to metals, glass, etc. due to their long chain molecules. Be that as it may, the glass transition temperature (T_g), adhesion (metal-polymer coating), acoustic impedance, and compression strength also matter. Also, the excellent resistance to weather, abrasion, corrosion, water, heat, ozone, and oxidation fortifies the polymer-based materials for underwater applications since these materials need to be able to endure more severe conditions, particularly in the depths of the ocean (Garu & Chaki, 2012; Jung *et al.*, 2002). polymeric materials also attract experimentalists due to their low cost and easy fabrication as well as good thermal/electrical insulation

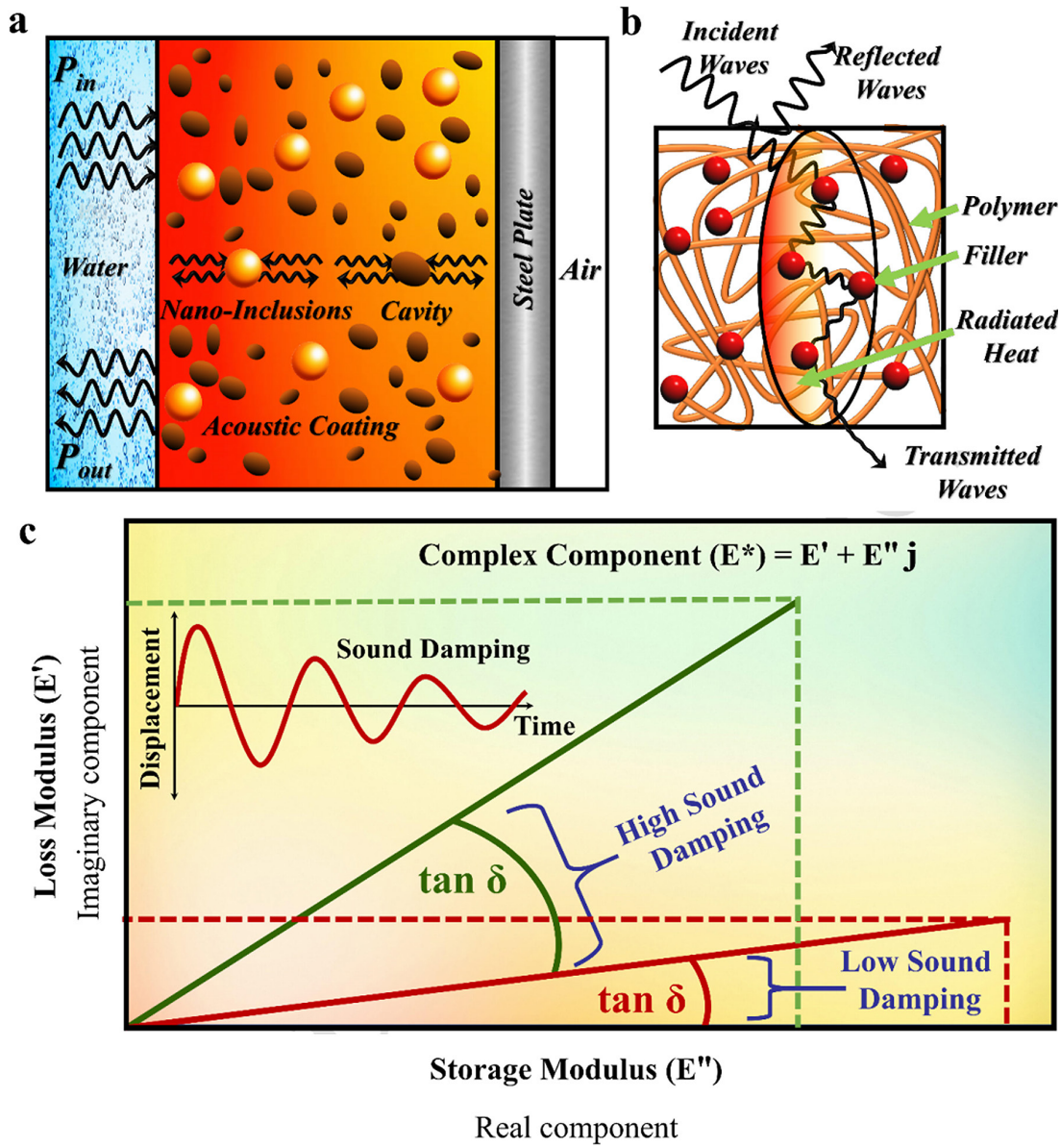

Figure 3. Schematic illustration on the: (a) sound absorption mechanism of sound absorption coating, (b) damping of sound waves by the incorporation of fillers in visco-elastic polymer matrix, (c) correlation of tan δ value and sound damping property (Goken *et al.*, 2018; Zhang *et al.*, 2023).

properties. To reduce the acoustic target strength of underwater vehicles, various polymer-based materials are widely used in which, interpenetrating polymer networks (IPN), and polymer foams are some of the most commonly used polymers. Various kinds of inclusions are incorporated into the polymer matrix to enhance the underwater sound absorption capabilities to a greater extent. Nanofillers like graphene, CNT, etc. have captivated the high demand as non-inclusions in polymer matrix that can effectively damp the soundwaves.

The plethora of polymers still demands novel polymers or interpenetrating polymer networks for underwater acoustic application. The majority of homogeneous polymers exhibit limited effectiveness in absorbing underwater sound due to their impedance matching and low loss factor. For instance, tan δ of polydimethylsiloxane (PDMS), epoxy, and polyurethane (PU) are more or less than 0.225, 0.38, and 0.5, respectively. To enhance sound absorption properties even further, a novel synthetic macromolecule is developed by

linking more than two polymers through a cross-linked network, namely IPNs (shown in Fig. 4a), which need to be introduced. For instance, tan δ value can be increased if an IPN of PU and PDMS is developed, without compromising mechanical properties, which is shown in Figs. 4(b-d). Finite element method (FEM) modeling can be used to simulate and correlate the acoustic data like absorption coefficient (AC), reflection coefficient (RC), and transmission coefficient (TC) calculated using Equation 5, 6, and 7, respectively. Overall, the IPN have a high tan δ value compared to PU-based acoustic absorbers (Jayakumari *et al.*, 2011).

$$RC = 10 \log (I_r/I_i) = 20 \log (P_r/P_i) \quad \text{Eq. (5)}$$

$$TC = 10 \log (I_t/I_i) = 20 \log (P_t/P_i) \quad \text{Eq. (6)}$$

$$TC + RC + AC = 1 \quad \text{Eq. (7)}$$

where, I_r, I_i, and I_t are the intensities of the reflected, incident, and transmitted waves, and P_r, P_i, and P_t are the corresponding pressure amplitudes, respectively. The total intensities of these signals equals one on a linear scale.

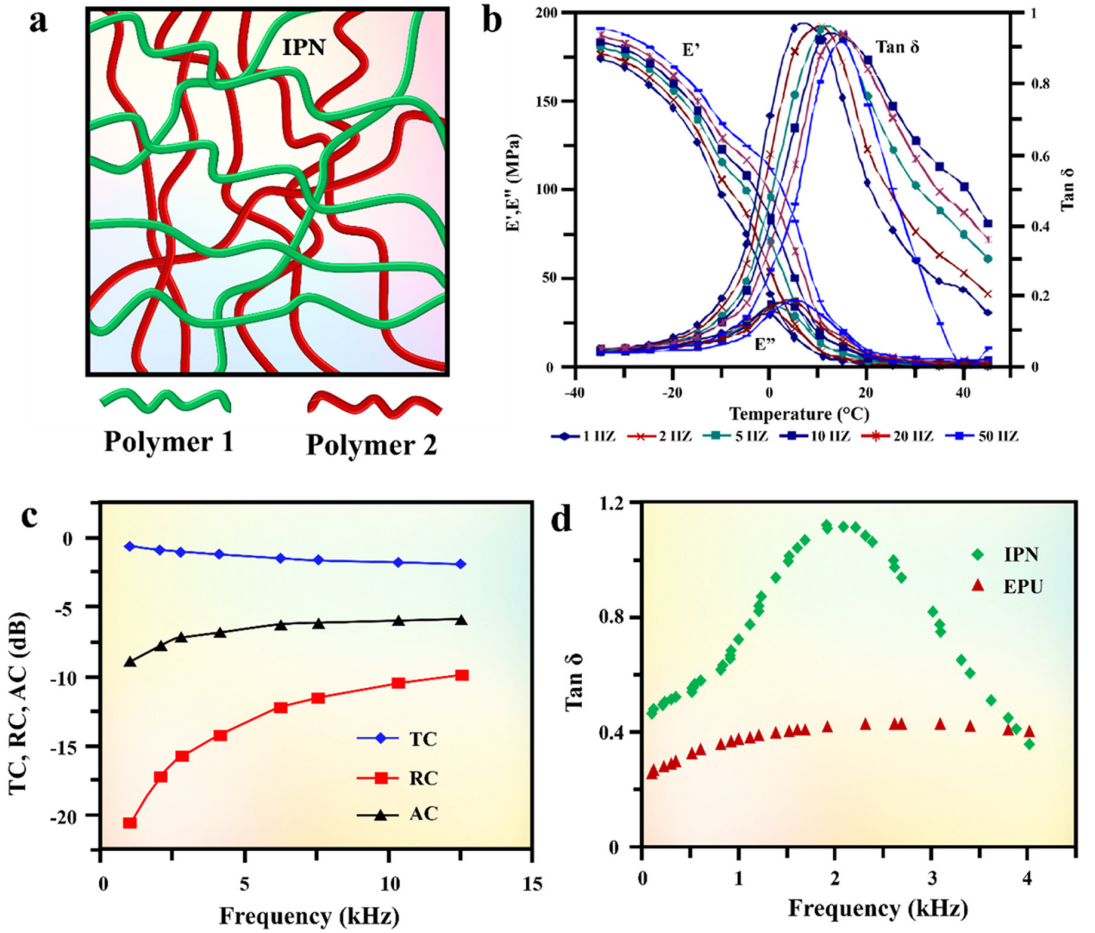

Figure 4. (a) Schematics of an IPN, (b) plot of dynamic mechanical analysis of IPN based acoustic absorbers composed of PU with PDMS, (c) acoustic data of IPN from FEM modeling (water-tile-water configuration), and (d) comparison of the tan δ of IPN and PU based acoustic absorbers(Jayakumari *et al.*, 2011).

3. GRAPHENE SYNTHESIS: BOTTOM-UP AND TOP-DOWN APPROACHES

Recently, various potential methods have been used to synthesize graphene which is briefly summarized in Fig. 5. Starting from the bottom-up approach that includes the mostly adapted chemical vapor deposition (CVD) method in which direct synthesis of graphene from carbon sources either by growth on a transition metal catalytic substrate mainly Ni, Cu,

Ru, etc., or substrate-free, that results in the large surface area single to few-layer graphene films with high quality (Muñoz & Gómez-Aleixandre, 2013). Despite the trajectories of CVD research, plasma-enhanced chemical vapor deposition (PECVD) were also widely accepted for its low catalytic reaction temperature (~650 ˚C) and reaction time, which results in large surface area single layer graphene with high yield compared to CVD method (Terasawa & Saiki, 2012). Physical vapor deposition (PVD) stands as an alternative to CVD method which relays hope on high-quality graphene with high yield along with low thickness, sharp edges, and controllable growth, set up at lower temperatures on a variety of substrate materials using vacuum evaporation, filtered cathodic vacuum arc deposition (FCVA) and ion beam-assisted deposition (IBAD) or sputter deposition and ion plating, which can be used based on the type of energy source supplied for the deposition process on which it is done in vacuum or on generating plasma thus bombarding it with solid carbon source so produce ions or free radicals (Oldfield *et al.*, 2015). The epitaxial growth of graphene via the thermal decomposition of hexagonal single-crystal silicon carbide (4H-SiC and 6H-SiC) lead the excess carbon to aggregate and thus form a hexagonal network structure of carbon atoms which is graphene (Huang *et al.*, 2021). Another versatile and reliable approach towards graphene synthesis via thermal fusion of the polycyclic aromatic hydrocarbons (PAHs), which already contain fused hydrocarbon rings which are the building units of a one-atom-thick graphene film, at high temperature (1100 ˚C), through a simple organic reaction route (Wan *et al.*, 2012). Other bottom-up graphene synthesis methods like substrate-free gas phase (SFGP) synthesis of graphene also grabbed the attention of experimentalists (Liu & Zhang, 2010).

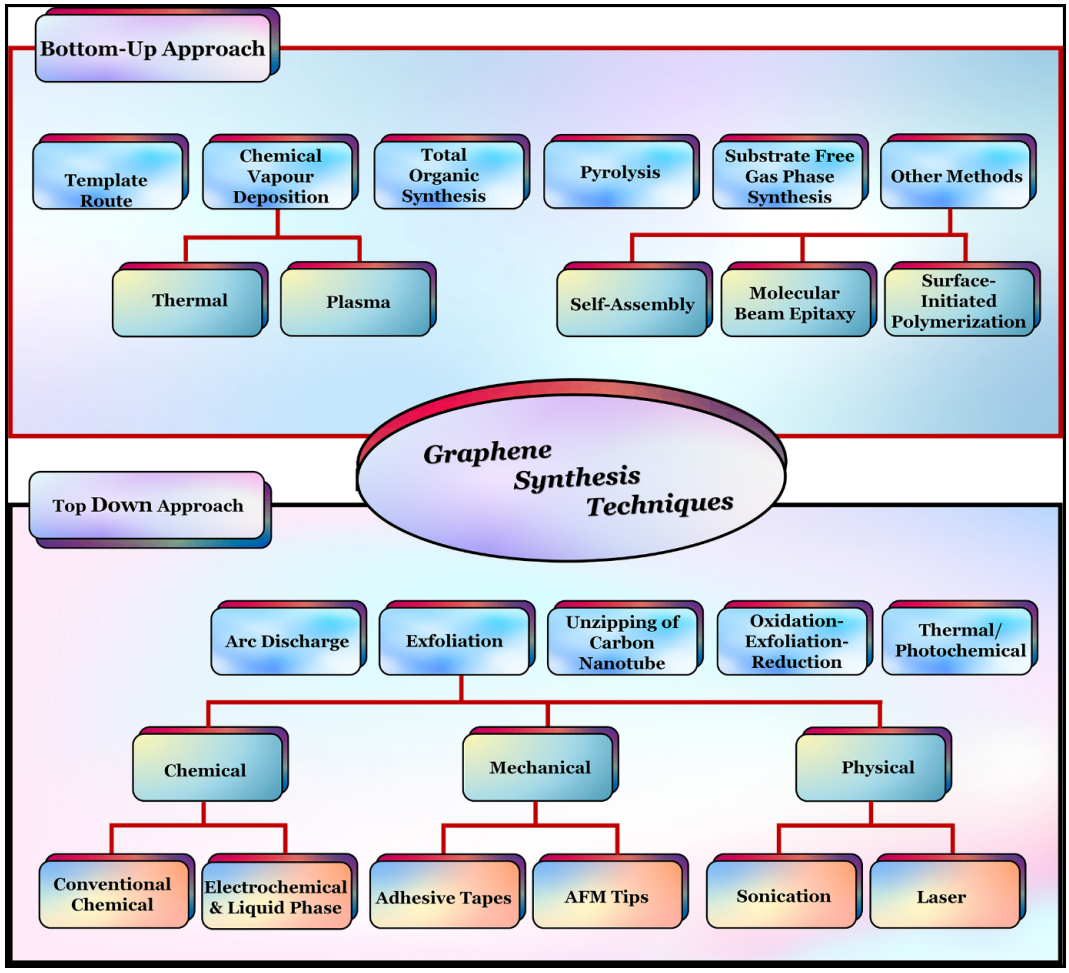

Figure 5. Graphene synthesis by various methods based on top-down approach and bottom-up approach.

The top-down approach towards graphene synthesis includes the exfoliation of graphite or graphite intercalation compounds (GICs) by mechanical exfoliation, electrochemical exfoliation, and liquid phase exfoliation (LPE), appending with the other methods including unzipping of carbon nanotubes (CNTs), arc discharge and oxidation-exfoliation-reduction of graphite oxide. To separate a graphene sheet from the graphite, it needs to overcome the van der Waals force of attraction between them. The trajectories of the exfoliation method were paved from the mechanical "peeling off" of graphene from the graphite molecule since the graphite is composed of stacked graphene layers held by the week van der Waals force. Either exfoliating the graphene layer from the ordered pyrolytic graphite by a sharp single crystal diamond wedge, which is purely mechanical thus saves labor cost and time. Or by using a pressure sensitive tape, an adhesive tape like polyvinyl chloride (PVC), by a three-roll mill but the difficulty in recovery of the graphene along with the adhesive and production cost was in concern (Yi & Shen, 2015). LPE includes the exfoliation of graphite flakes by the intercalation of a potential intercalates like naphthalene with the help of a solvent via sonication in which more than 40 types of aqueous or non-aqueous solvents like N-Methyl-2-Pyrrolidone (NMP), with a high boiling point of >200 ˚C and/or surface tension of 40-50 mJm^{-2} is best suitable, can be used in graphene synthesis and results in single-layer graphene (Xu *et al.,* 2014; Xu *et al.,* 2018). The electrochemical method in ionic liquids makes use of a carbon source like graphite as an electrode and H_2SO_4-KOH solution as electrolytes, which results in a few nanometre thick graphene (Yu *et al.,* 2015). Intercalation of substances like alkali metals (like lithium salt) into graphite *in situ* to expand the graphite, which is known to be GICs followed by sonication results in the exfoliation of graphene layers from the GICs, thus the formation of graphene (Yoon *et al.,* 2015).

CNTs are made of graphene layers which are folded in to tube-like cylindrical structure that can be unzipped by breaking or cutting off the C-C bond in the axial or longitudinal direction to make it back to flat layers resulting in graphene nanoribbon (GNR) which are quasi one-dimensional materials, with different number of layers and properties depending on the structure of CNTs i.e., single-walled CNTs (SWCNTs) or multi-walled CNTs (MWCNTs) were used for the process (Shinde *et al.,* 2011). The electrical arc discharge method make use of two electrodes, in which the cathode consists of a graphite rod and the anode consists of a carbon-rich material as carbon precursor like petroleum asphalt, in a non-conductive medium like air, gas, water, or in any inert gas conditions. Plasma is generated by the dissociation of the gas in the medium on the passage of electric current

Method	Thickness	Lateral	Advantage	Disadvantage
Micromechanical exfoliation	Few layers	μm to cm	Unmodified and large-size graphene sheets	Very small-scale production
Electrochemical exfoliation	Single to few layers	500-700 nm	High electrical conductivity of the functionalized graphene	High cost of ionic liquids
Direct sonication of graphene	Both single and multiple layers	μm	Inexpensive and unmodified graphene	Low yield
Reduction of carbon monoxide (CO)	Multiple layers	Sub-μm	Un-oxidized sheets	Contamination with α-Al_2S and α-Al_2O_3
Epitaxial growth on SiC	Few layers	Up to cm size	A very large area of pure graphene	Very small scale
Unzipping of carbon nanotubes	Multiple layers	few μm long nanoribbons	Size depends on the starting nanotubes	Expensive and oxidized graphene
CVD	Few layers	Very large e (cm)	Large size; high quality	Small production scale

Table 2. An overview of the growth mechanisms of graphene through top-down and bottom-up approaches (Mbayachi *et al.,* 2021).

and the plasma generated can decompose the carbon precursor in anode resulting in high quality large surface area graphene (Li *et al*., 2010). Oxidation-exfoliation-reduction of graphite oxide method includes the oxidation of graphite oxide, by various potential methods like Brodie method, Staudenmaier method, Hofmann method, and Hummer's method which are widely accepted in which Hummer's method grabbed great attention, results in GO and on further reduction, desired graphene sheets are synthesized, with high yield and low cost of production (Chen *et al.,* 2013; Feicht *et al.,* 2019; Poh *et al.,* 2012). An outline of the peculiarities of the graphene synthesized by various methods is summarized in Table 2.

Structural characteristics of graphene can be revealed using various techniques. Raman spectra of graphene sheets show peaks at G-band, D-band and 2D-band at ~1580 cm^{-1}, 1350 cm^{-1}, and 2700-2900 cm^{-1}, respectively, due to in-plane vibration of carbon atoms, second-order two-phonon process, and defects in graphene lattice, shown in Fig. 6a. In a monolayer graphene, the 2D-band is a single sharp peak, while in multilayer graphene, it becomes broader and more complex due to the presence of additional van Hove singularities. X-ray diffraction pattern of graphene also shows no peak in the spectra due to its two-dimensional nature, shown in Fig. 6b. But, in case of multi-layer graphene, graphene oxide and graphite, XRD peaks are obtained which is due to its three-dimensional structure, shown in Fig. 6b. Also, Field emission scanning electron microscopy (FESEM) and TEM images of graphene are also shown in Figs. 6c,d, having a sheet-like structure, as well as the atomic force microscopy (AFM) image of the single-layer and bi-layer graphene is also shown in Figs. 7a,b along with its height profile.

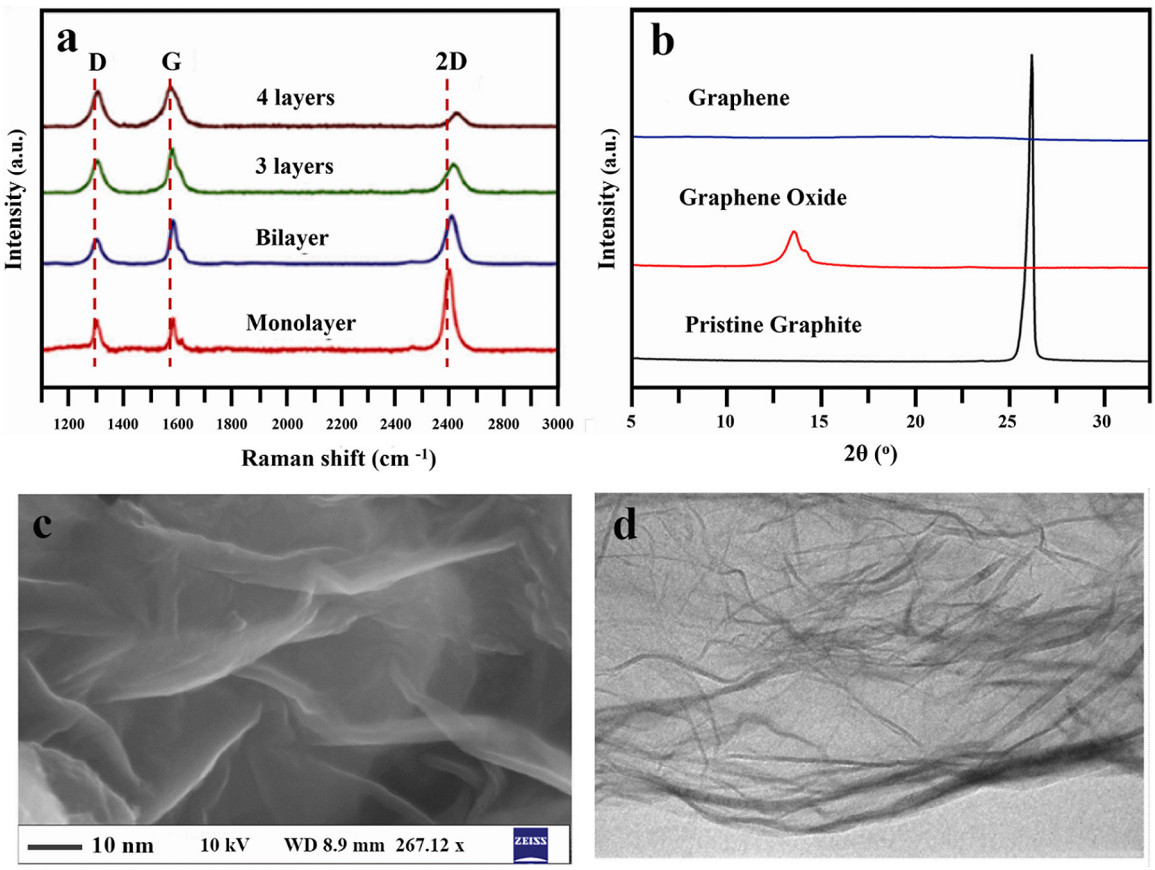

Figure 6. (a) Raman spectra of the monolayer to few-layer graphene sheets, (b) XRD pattern of graphene, graphene oxide, and pristine graphite, (c) FESEM image, and (d) TEM image of graphene sheets (Mbayachi *et al.,* 2021; Subramanya & Bhat, 2015; Bin Zhang *et al.,* 2010).

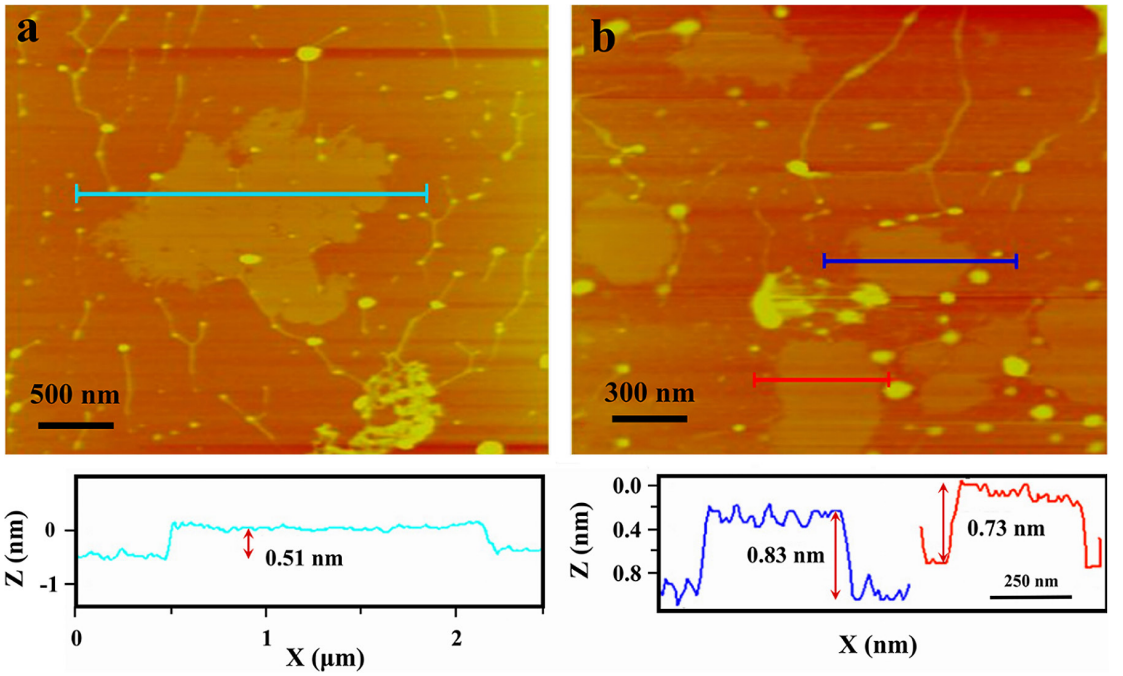

Figure 7. AFM images of representative (a) monolayer grapheneand (b) bilayer graphene (The histogram also shows the height profiles) (Shen *et al.*, 2011).

4. SYNTHESIS OF GRAPHENE-BASED POLYMER NANOCOMPOSITES

In general, polymer composites are defined as a multi-component system in which a single-layered or multi-layered graphene is incorporated into the polymer matrix. Foresee, the challenge is to tune the system for sound wave absorption without compromising its mechanical and thermal attributes. Graphene polymer composites can be prepared by various methods including in-situ polymerization, melt mixing, solution/latex stage blending, electrospinning, and two-roll milling/internal mixing. In-situ polymerization is a widely used chemical route for the preparation of graphene polymer composites that involves the formation of a covalent bond between the elastomeric matrix and the functionalized graphene on applying heat or radiation (Zheng *et al.*, 2004). Covalent bond formation gives a strong interaction between filler and polymer. Polymers like epoxy (Guo *et al.*, 2011), Nylon-6 (Xu & Gao, 2010), PMMA (Potts *et al.*, 2011; Wang *et al.*, 2012), polyimide (Wang *et al.*, 2011), polypropylene(Polschikov *et al.*, 2013; Shevchenko *et al.*, 2012), PVDF, poly (butylene terephthalate)(Fabbri *et al.*, 2012), PU (Wang *et al.*, 2011), etc. are reported. In some cases, the solvent is also used in the in-situ polymerization process. Melt mixing, is a cost-effective, eco-friendly, and fast method for the low or even massive production of graphene polymer nanocomposites that involves the direct dispersion of graphene in a molten polymer matrix at high temperature and shear force without the use of any harmful solvents. Melt mixing can be done by extrusion, injection molding or even by internal mixing (Verma & Goh, 2018), and can be adapted for polar as well as non-polar polymers. At high filler stacking, the viscosity increases results in poor dispersion of graphene within the polymer matrix and thus, overall properties of the nanocomposite including the sound absorption properties are reduced (Singh *et al.*, 2011).

Suspension of graphene is prepared and incorporated into polymer matrix which is in latex stage or solution stage, simply by magnetic stirring and/or ultra-sonication leads to enveloping of graphene by polymer latex/solution (Kuilla *et al.*, 2010). Latex stage mixing method is particularly used for the preparation of elastomeric composites (Liu *et al.*, 2019; Mao *et al.*, 2013; Xu *et al.*, 2016; Zhang & Cho, 2018) and coagulation of latex helps in reducing agglomeration of graphene. Further evaporation of solvent results in the graphene reinforced polymer nanocomposite with enhanced dispersion of graphene in polymer matrix. Be that as it may, the sonication time, solvent used as well as the

mixing time and composition of graphene in polymer matrix may affect the overall properties of the prepared nanocomposites dramatically. Another unique and powerful approach towards the prepare of graphene polymer nanocomposites is the electrospinning method which was firstly experimented with the colloidal suspension like natural rubber latex and graphene nanoplatelet (Cacciotti *et al.*, 2015). Graphene suspension is prepared with appropriate solvents, for example chloroform, by magnetic stirring or/and ultra-sonication process. Electrospinning techniques depends on the viscosity, concentration, conductivity, molecular weight, solvent volatility and surface tension of the polymeric solution as well as the processing conditions like temperature, feed rate, distance between source and collector plate, diameter of needle of syringe, effect of voltage and collector plate(Ray *et al.*, 2016; Wasim *et al.*, 2018). High voltage of around 14-16 kV is applied to the graphene-polymer suspension in a suitable solvent (polymeric solution) filled in the syringe needle connected to a spinneret result in ejection of charged liquid jet. The electrostatic potential generated within the polymeric solution helps in overcome the surface tension and the ejects out the polymeric solution that is collected by the metal collector (Babul Reddy *et al.*, 2015; Cacciotti *et al.*, 2015; Zárate *et al.*, 2020). Graphene polymer nanocomposites with fibrous structure and good dispersion as well as orientation will be obtained with this technique.

In two-roll milling process, mechanical shearing force is applied through two opposing horizontal rolls rotating in opposite direction in different speed. The thickness of the nanocomposite sheet is tuned by changing the nip gap, which is the gap between the two rolls(Potts *et al.*, 2013). In internal mixing process, the compounding ingredients including the graphene and polymer matrix is add into the chamber with specific volume. The rotors within the chamber will be rotating giving a shear force and the temperature can be applied if needed (Freakley & Wan Idris, 1979; Rodgers & Waddell, 2013). Melt mixing can also be done using the internal mixer like Brabender. Mixing time, nip gap, fill factor, chamber volume, rotating speed of rotors/rollers, and temperature can affect the overall properties of the prepared graphene polymer nanocomposite(Potts *et al.*, 2012). Two-roll milling method can be used particularly for elastomeric matrix like natural rubber, acrylonitrile butadiene rubber (NBR), styrene butadiene rubber (SBR), etc. (Amutha Jeevakumari *et al.*, 2020; Potts *et al.*, 2012). Other than these methods, reversible addition fragmentation chain transfer (RAFT) polymerization is a multi-step chemical method for the synthesis of graphene polymer nanocomposites mediated by a RAFT agent like dithiocarbamates, xanthates (dithiocarbonates), pyrene based, etc. (Cui *et al.*, 2012; Liu *et al.*, 2010). Polymers experimented were polystyrene (Ding *et al.*, 2015; Gu *et al.*, 2014), PMMA(Tumnantong *et al.*, 2020), etc. Xiao *et al.*, (Xiao *et al.*, 2010) developed a freestanding chemical vapor deposited graphene (CVDG) using a microwave plasma-enhanced CVD system. Subsequently, they prepared graphene/epoxy composites by dispersing the CVDG directly into a curing agent consisting of diamine epoxy and then subjecting it to thermal curing with diepoxides. Even though the CVDG lacks reactive or polar groups, the epoxy matrix with the functionalities/bonds like, C-H bonds, aromatic rings, and hydroxyl functionalities, can engage in diverse π interactions (such as CH-π, π - π, and OH-π) with the abundant π-electrons present on the CVDG. This, in turn, led to substantial interfacial interactions between the graphene and epoxy matrix, efficient load transfer, and a noteworthy enhancement in properties. These graphene-polymer systems can be used for the sound damping application without compromising the mechanical properties like tensile strength, etc., tuning the uniform dispersion of graphene content in the polymer matrix, shown in Fig. 8a. The TEM images of graphene/natural rubber nanocomposite is shown is Fig. 8b. Figs. 8c, d gives the Ashby plot of Young's modulus vs. tensile strength and Young's modulus vs. density that compares the properties of graphene as well as graphene based polymer nanocomposites with conventional materials, which is supported by absorption coefficient vs. frequency plot of graphene/Styrene Butadiene Rubber nanocomposite, shown in Fig. 8e.

5. GRAPHENE-POLYMER SYSTEM VS. UNDERWATER SOUND DAMPING

Graphene, being a two-dimensional material, having high surface area to volume ratio, can be harnessed to achieve effective sound damping property of the acoustic material due to its nano size, complex interactions at graphene-polymer interphase (even in low filler loading) and exceptional contribution towards mechanical as well as tribological properties of the polymer nanocomposite

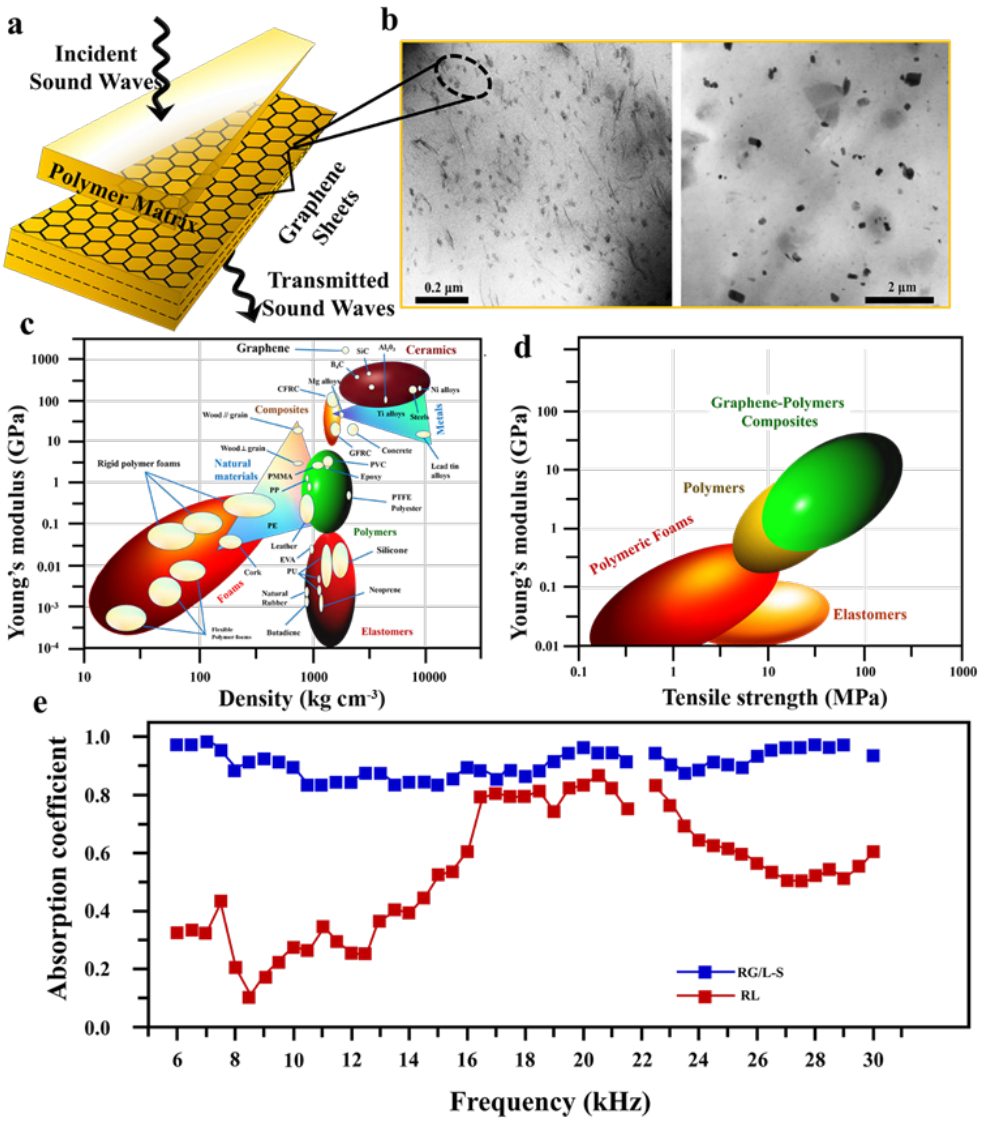

Figure 8. (a) Schematic illustration of graphene in polymer matrix for sound damping, (b) TEM images of graphene in natural rubber matrix (at low and high resolution), (c) Ashby plot of Young's modulus as a function of density comparing the properties of graphene to those of conventional materials (Please be aware that the axes on the graph are presented using a logarithmic scale, and the density of graphene used in this comparison is 2200 kg. m^{-3}), (d) Graph illustrating Young's modulus vs. tensile strength, comparing the properties of graphene-polymer nanocomposites with other materials , and (e) Underwater absorption coefficient of graphene/Styrene Butadiene Rubber nanocomposite, (Kinloch *et al.,* 2018; Li *et al.,* 2017; Verdejo *et al.,* 2011; Zhan *et al.,* 2011).

(Wu *et al.*, 2018). Acoustic waves are longitudinal waves, usually generated when a vibrating surface comes in contact with the surrounding medium and displace the particles of the medium to create alternate zones of high pressure (compression) and low pressure (rare fraction). In this case, the wave length, pressure amplitude, time period, frequency, and phase speed of the sound waves matters. As the polymer adheres to the filler, there's a possibility of a reduction in its free volume, leading to an increase in the glass transition temperature (T_g) of the polymer region which is directly in contact with the filler, typically within an interfacial thickness of about 50-100 Å. Consequently, when a filler is introduced, the T_g peak tends to broaden and also shifts to a higher temperature. Various vital points

need to be considered while incorporating fillers into polymer systems. This includes the morphology, orientation and composition of the filler. The filler-polymer interface also plays a vital role in the development of polymer composites based acoustic absorbers, which is schematically shown in Fig. 9a. Despite from the trajectories of graphene history, researchers are interested in experimenting the performance of graphene into sound damping applications due to its versatile contribution towards polymer matrix.

High surface to volume ratio of the graphene allowing it to interact effectively with sound waves, along with its high mechanical strength that enables it to withstand and dissipate vibrational energy efficiently. Low-cost large production of graphene with high purity is another advantage that makes graphene a potential nano-inclusion for underwater acoustics. Ease of intercalation and low mass density of graphene ameliorates the use of graphene-polymer system towards high-end coatings, etc., as well as its biocompatible also plays a vital role. The high thermal conductivity of graphene (2000-5000 W m^{-1} K^{-1}), contributes high towards withstanding the heat which is formed by the dissipation of vibrational energy, can be a major merit of graphene-polymer systems. The orientation of the graphene in the polymer matrix also effects the acoustic property of the underwater acoustic material. Directionally antagonistic graphene-polymer systems show great sound damping property which is schematically shown in Fig. 9b, and the polymer matrix chosen was polyurethane (Oh *et al.*, 2018). Along with all its benefits, graphene can enhance the visco-elastic property of the graphene-polymer system, which is crucial for the sound damping property. The hydrophobic nature of graphene contributes higher to the water-resistant property of the sound damping materials since the material is used for underwater application, which also prevents biofouling of the acoustic coating. Thus, graphene-polymer system is suitable for developing robust underwater sound damping materials. Graphene which is evenly distributed in the polymer matrix can form a continuous network which enables multiple scattering of sound waves and wave-mode conversion to enhance sound damping property of the material. Graphene based polymer nanocomposites shows sound absorption co-efficient greater than 0.8 at 6-30 KHz and performs well under high hydrostatic pressure up to 3 MPa. Unlike other traditional fillers, the adaptability of graphene into its derivatives, like graphene oxide (GO), reduced-GO (rGO), makes it appropriate for acoustic applications.

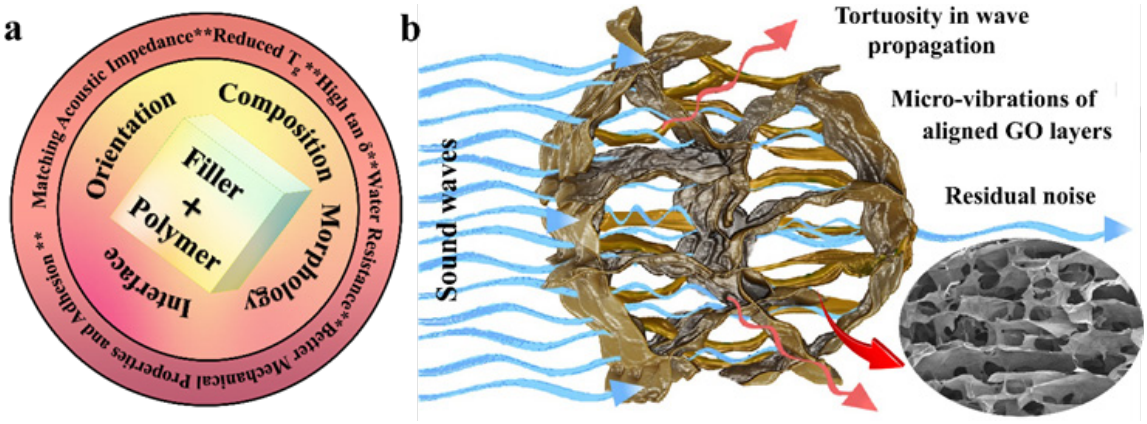

Figure 9. Schematic illustration of the (a) factors to be considered while designing filler-polymer networks based acoustic absorbers and the expected vital properties (b) mechanism of sound absorption in a directionally antagonistic (parallel) graphene sound absorber (Oh *et al.*, 2018).

Thus, viscoelastic polymer networks being the versatile materials with excellent sound damping properties fortified with the exceptional physico-mechanical properties of graphene and its derivatives as nano-inclusions can be used for stealthy acoustics applications, in a wide range of temperature, frequency, and strain domains (Li *et al.*, 2017; Mohamad *et al.*, 2017). Studies on the multifunctional graphene intercalated styrene-butadiene rubber (SBR) nanocomposite show enhanced sound damping property of sound absorption coefficient more than 0.8 (6-30 kHz) having direct correlation with the water pressure. The nanocomposite also shows enhanced thermal and

mechanical properties which makes them multifunctional materials. Baihua Yuan *et al.,* (2018) developed the graphene nanoplatelet (GNP)-modified acrylonitrile-butadiene rubber-based materials for underwater acoustics. Elastomers-based underwater acoustic material shows excellent sound damping properties due to its elastic nature. GNPs contribute higher towards sound absorption (from 0.35 to 0.73) which can be attributed to the enhanced interfacial contact area between filler-matrix system, along with a significant enhancement in mechanical properties, which makes the material capable for underwater acoustic application. Katsiropoulos *et al.,* (2022) investigated the graphene nanoplatelets (GNPs) doped epoxy and carbon fibre/epoxy polymer composite (CFRPs). The results show that GNPs concentration (in between 0.15-1 wt. %) effects significantly as well as very likely, which supports the interfacial shear friction as the pivotal damping mechanism (also in Rafiee *et al.,* 2019). Li *et al.,* (2019) developed poly (vinyl alcohol)/ graphene nanocomposite membrane-type acoustic materials to actively adjust the anti-resonant frequency of the membrane, showing excellent sound attenuation properties in the frequency range of 369.2 to 420 Hz.

Effective stress transfer mechanism also enables the enhancement in storage modulus, which can also be functioned by the surface modification of graphene (Pious *et al.,* 2020). Bin Li *et al.,* (2011) evaluated the surface modification of graphene nanoplatelets (GNP) by silanization and found that, 3 wt. % surface modified GNP in polyetherimide matrix shows enhanced storage modulus and thus enhances sound damping property with an enhanced damping factor 3 times higher than PEI. Dashtkar *et al.,* (2021) studied the carboxyl functionalized graphene in a PU matrix and found an increase in the sound absorption by 37% (200-300 Hz), 34% (500-600 Hz), and by 32% (700-1000 Hz), which can be widely used in various acoustic applications. Nautiyal, et.al. (2017) studied the damping property of graphene foam-based hierarchical polyimide composites which supports multiscale mechanisms including rippling, spring-like interlayer van der Waals interactions and flexing of graphene foam branches, also enhancing the interfacial adhesion, maintain the damping property in the extreme temperature range. The inclusion of just 1.5% by weight of graphene foam in the polyimide matrix results in a substantial enhancement of approximately 300% in the loss tangent.

Forsee, as an effective alternative, hybrid non-inclusions were also introduced to enhance the acoustic properties of the material (Fu, 2022; Navidfar & Trabzon, 2022; Simón-Herrero *et al.,* 2019). Graphene being one of the best nano-inclusions for sound damping, graphene-based hybrid nano-inclusions with CNT, wollastonite, fiberglass, etc., found their way towards sound damping applications (Elkasaby *et al.,* 2020). Lei Liu *et al.,* (2019) have developed eco-friendly low-cost melamine-based composite foam with a semi-open cellular structure incorporated with tiny self-assembled GO sheets disrupted by functionalized CNT, resulting in a significant increase of 100% and 20% in a frequency band ranging from 250 to 1600 Hz, that can be sued for wide range of acoustic application, that can also be adapted in case of underwater acoustics due to its optimal air-flow resistance, tortuosity against sound wave propagation, smaller viscous characteristic lengths, internal reflection and interfacial damping. On the other hand, Wu *et al.,* (2017) reported for the first time, a three-dimensional graphene-based composites of poly(dimethyl siloxane) (PDMS) incorporated with graphene foam and CNT, specifically for the low-frequency sound damping of 100-1000 Hz with an absorption coefficient higher than 0.3. The same nanocomposite can absorb 70% of the incident sound waves at a low frequency between 100 and 200 Hz, which makes it viable for commercial application. Rafiee *et al.,* (2019) have developed multiscale laminated fiberglass/epoxy composites modified with various carbon nanofillers, including MWCNT, GO, RGO, and GNP, which showed an enhanced the damped natural frequency of the nanocomposites but in higher nanoparticle loading, an inverse effect was found which can be due to the agglomeration of the nanoparticle in higher concentration. If not carefully tuned, the acoustic absorption property of the material can decrease due to the increased crystallinity and unsuitable structure, which could be a challenge (Verdejo *et al.,* 2008). Structure, composition as well as orientation of graphene within the matrix also affects the acoustic damping property of the underwater acoustic material (Lee *et al.,* 2023; J. Lee & Jung, 2019; Lu *et al.,* 2020; Yang *et al.,* 2020; Zong *et al.,* 2021). Gong *et al.,* (2022) studied the effect of interfacial sliding of two-dimensional multilayer graphene oxide in carbon fiber reinforced polymer composite in which, a damping factor of 0.0345 is observed at a strain of 0.235 and frequency 1 Hz, whereas the energy

dissipation ability of the material also matters. Kiddell *et al.*, (2023) used flash graphene with different flake diameters, flake thicknesses and oxygen content which was produced from different feedstocks in flexible polyurethane foam (PUF). Studies show that, 0.025 wt. % of FG in PUF itself showed enhanced sound absorption properties than raw PUF without compromising its mechanical properties. Structural and feasibility studies can be done using simulation studies while developing material for underwater acoustics using various simulation models. Lee *et al.*, (2023) have done an algorithmic approach to validate the experimental results on the sound damping property of multi-layer GO/PU foam, shown in Figs.10a,b (for double-layer structure in which each layer is 4 mm thick) along with the sound absorption curve. Studies show that the average sound absorption coefficient was significantly increased by up to 153% in comparison to the original PU. Also, it was found that the sound absorption property can be enhanced even up to 49% just by optimizing the layer arrangement with the same GO content in the polymer matrix. Xu *et al.*, (2020) studied the sound radiation as well as transmission loss parameters of functionally graded porous graphene-reinforced nanocomposite plates using a simple quasi-3D high-order shear deformation theory (HSDT) integrated with other modeling theories based on Hamilton's principle, which showed that, the sound damping property of a material is significantly influenced by various parameters include temperature, thickness, and porosity of the material, angle of incidence of sound waves, etc. The effect of the weight fraction of functionally graded porous graphene on the sound radiation of nanocomposite is shown in Fig. 10c.

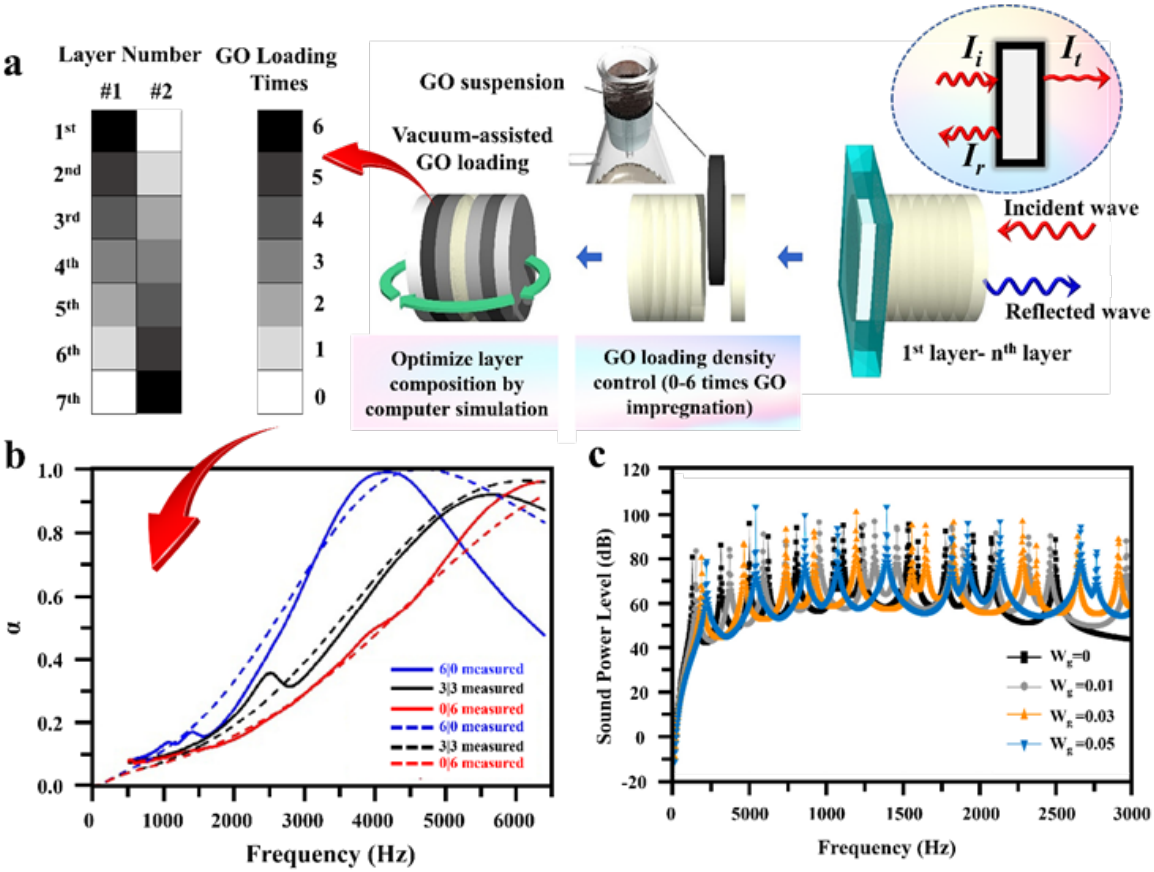

Figure 10. Simulation studies on the (a) optimal arrangements of a double-layered structure (each layer 4-mm-thick) of the graphene oxide-based polymer networks, (b) measured and calculated sound absorption curves of various double-layered structures, and (c) quantitative analysis on the effect of concentration of functionally graded porous graphene in the polymer system by simulation studies (Lee *et al.*, 2023; Zhichao Xu *et al.*, 2020).

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The mechanism of sound damping by nano-inclusions can be debated based on the terms 'resonance', 'mode conversion', and 'friction'(Fu *et al.*, 2021). Figs. 11a,b illustrate the sound damping mechanism of the graphene-polymer networks by various approaches and Figs. 11c,d show the effect of the concentration of graphene as nano-inclusions on the T_g value and sound-damping properties (Fu *et al.*, 2021; Pang *et al.*, 2022; Yuan *et al.*, 2018). Firstly, graphene incorporated into the polymer matrix has its natural frequency and that will start to resonate with the frequency of the incident sound wave. Thus, the peak value of the sound absorption coefficient appears at the resonance frequency, in other words, sound absorption can only occur around the resonance frequency. Foresee, in this case, the working frequency range of the sound-damping material becomes narrow. Thus, to broaden the

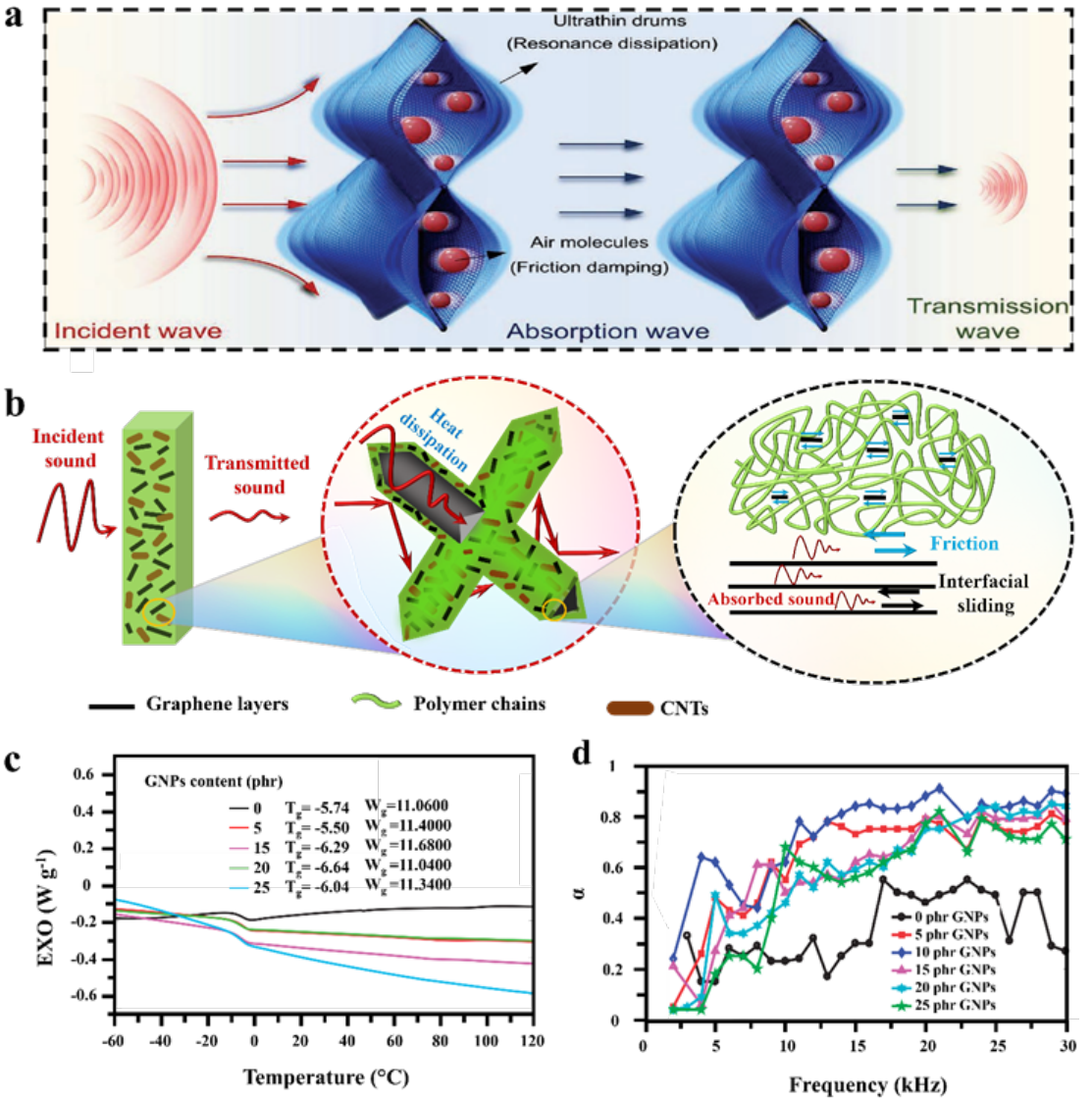

Figure 11. (a) Illustrates the sound damping mechanism of the graphene-polymer-based absorber by the resonance of ultrathin graphene and air friction damping in the pores, (b) Schematics representation of the attenuation of sound waves due to interfacial sliding and friction occurring between carbon nanotubes (CNTs), layers of graphene, and its interfaces, (c) Differential scanning calorimetry graph of the GNPs/NBR nanocomposites and (d) Spectra depicting the underwater sound absorption coefficients (α) of nanocomposites with varying levels of graphene nanoplatelet (GNP) content (Fu *et al.*, 2021; Pang *et al.*, 2022; B. Yuan *et al.*, 2018).

band of sound absorption coefficient, nano-inclusion should be of mixed size and multi-layered, so theoretically, multi-layer graphene would be a better choice. Secondly, in solid material, sound waves can exist in longitudinal as well as shear waves, but they can only propagate as longitudinal waves in water. This is attributed to mode conversion. In this case, the intercalation of graphene (being a rigid nano-inclusion) can convert longitudinal waves to shear waves at the graphene-polymer interphase. Wen *et al.*, (2011) have modeled the same using the finite elemental method. The advantage of mode conversion is that only the longitudinal waves can propagate back to the water, not the shear waves, which also contribute to the sound-damping property of the material. Thirdly, when surfaces in contact move relatively to each other, the friction between two surfaces (graphene and polymer matrix surface, at the interphase) converts kinetic energy to thermal energy, and thus heat will be dissipated since graphene and polymer matrix have different displacements(Sharma *et al.*, 2017).

Challenges of this system would be, the narrow band which needs to be carefully studied and tackled by using multi-sized and multi-layered graphene systems. Also, due to the deformation of both matrix and nano-inclusion, there is a possibility for a change in the mechanical properties of the polymer nanocomposites at high pressure. The consequence would be the sound absorption may shift to a higher frequency. This can be tackled by tuning and optimizing the graphene loading in the polymer matrix to enhance the mechanical properties like compression set, without compromising the impedance matching and sound damping property. This also helps in optimizing the thickness of the material focusing on low thickness and high sound damping properties, since it is attributed to the materials' cost-effectiveness and sailing resistance in water. Lastly, the multifunctionality of the graphene-polymer system also helps in reducing the biofouling as well as reducing the surface roughness which hurts the drag.

Foresee, as a counterpart, experimentalists in the field of underwear acoustics are also interested in developing underwater acoustic sensors like micro-electro-mechanical systems-based sensors working on the principles of piezoelectrics, particularly focusing on the detection of low frequency stealthy underwater vehicles and the system also uses graphene-based systems (Prabhu *et al.*, 2021; Smitha Pai *et al.*, 2023; Wu & Chou, 2016). Graphene-based materials are also used in other underwater acoustic applications like transistors as underwater microphones also focus broadband wireless communication underwater, etc. (Li *et al.*, 2018; Wang *et al.*, 2020).

6. SUMMARY AND PERSPECTIVES

Wide studies have been done on graphene and its derivatives for the development of polymer networks-based acoustic absorbers, without compromising but enhancing its multifunctional properties, even using simulation studies on various potential models. Researchers were interested in experimenting with various viscoelastic polymer networks, particularly focusing on their characteristic acoustic impedance, T_g value, thermal stability, mechanical and tribological properties, water resistance, sound absorption capacity, and loss factor. Graphene-based polymer networks show excellent sound-damping properties and thus can be used for underwater acoustic applications. Surface modification of graphene as well as the development of hybrid nano-inclusions also significantly enhanced the sound damping property of the material. Methodical as well as heedful development of IPNs is found to be revolutionary in the evolution of stealthy materials for underwater acoustics.

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Conflicts of Interest

The authors have no conflicts of interest. ♦

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