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RESEARCH PAPER

Exploring graphene origami-enabled metamaterials: A review

Hosein Ezzati [a], Farzad Ebrahimi [b], Erfan Salari [c*]

[a] *School of Mechanical Engineering, College of Engineering, University of Tehran, Tehran, Iran*

[b] *Department of Mechanical engineering, Faculty of Engineering, Imam Khomeini International University, Qazvin, Iran*

[c] *Department of Civil Engineering, Lakehead University, Thunder Bay, Ontario, Canada*

Abstract

Auxetic metamaterials are a class of material with tunable Poisson's ratio. Nonetheless, most auxetics have weak mechanical properties, thus, implementing graphene origami (GOri) as a strong reinforcement can overcome this deficiency. This review explores the burgeoning field of auxetic metamaterials enhanced through the integration of graphene origami. This study also presents an in-depth investigation of the background of negative Poisson's ratio auxetic metamaterials, most importantly, graphene origami. Furthermore, it focuses on the notable mechanical characteristics such as increased flexibility, strength, and dynamic response of GOri-reinforced composites. Leveraging Hamilton's principle and other theoretical frameworks, this paper collates and examines the equation of motion derivations and the impact of various parameters on GOri behaviour.

Keywords: Graphene origami; Auxetics; Metamaterial; Negative Poisson’s ratio.

1. Introduction

The realm of material science has been invigorated by the advent of metamaterials; a novel category of synthesized substances characterized by extraordinary properties seldom encountered in naturally occurring materials. Spanning recent decades, a plethora of these artificial metamaterials has been engineered, showcasing an array of atypical physical qualities across various domains of scientific inquiry [1]. These innovative materials have opened up a spectrum of possibilities, challenging conventional understanding and offering groundbreaking applications in technology and scientific research [2]. In recent discourse within the field of material science, the paradigm of metamaterials has undergone notable expansion. This new category comprises materials distinguished not only by the properties emanating from their macroscopic composition but also significantly influenced by their intrinsic architecture [3]. Such materials are recognized for their extraordinary attributes, including but not limited to enhanced static modulus and specific density considerations [4], as well as their notable efficacies in terms of energy absorption capabilities [5-7]. The acoustic and phononic characteristics of these materials are exceptionally advanced, presenting groundbreaking potential in sonic manipulation and vibration control [8-10]. Moreover, their thermal conductivity aspects offer innovative approaches to heat transfer management [11, 12]. Additionally, these materials fall under the purview of intelligent materials due to their responsive nature. A particularly striking feature is the manifestation of negative Poisson's ratio (NPR) [13-15], which further underscores the unconventional and superior behavioural patterns evident in this emergent class of metamaterials. Utilizing nanocomposites in an auxetic arrangement enables the enhancement of structural elements' stiffness and brings out features including NPR while simultaneously reducing their weight [16-18].

Recent scientific inquiries have been particularly focused on two-dimensional materials such as graphene,

*Corresponding author.
E-mail address: esalari@lakeheadu.ca

compelled by their extraordinary integration of mechanical resilience, electrical conductivity, optical transparency, and thermal endurance. This burgeoning interest is motivated by the exceptional attribute of these materials to maintain remarkable structural thinness while exhibiting unparalleled strength. The conductive capabilities of graphene and similar materials enable rapid and efficient electrical signal transmission, thus presenting vast potential for advancements in nanoelectronics. Moreover, their optical properties offer translucency and flexibility that could revolutionize display technologies. Lastly, their ability to withstand high temperatures and effectively dissipate heat makes them ideal for enhancing thermal management in a myriad of applications, ranging from advanced computing systems to renewable energy technologies [19-24]. The manipulation of these atomically slender, flat materials into intricate three-dimensional configurations through processes such as bending, creasing, or folding could profoundly modify their mechanical robustness, electrical performance, and optical traits. This paves the way for expansive innovation in the crafting of functional apparatuses and frameworks [25]. The mechanical behaviour of graphene is intriguingly similar to that of paper, which allows for the conceptual transfer from paper crafts to graphene structures [26]. Drawing inspiration from the art of origami, graphene can be engineered into three-dimensional forms by applying loads within its plane or perpendicular to it, akin to folding a paper model. The creation of such origami-inspired configurations showcases the incredible durability and remarkable flexibility of graphene when strategically folded. Through molecular dynamics simulations, scientists illustrated how the geometrical design of graphene origami structures allows for effective adjustments in both flexibility and Poisson's ratio. This approach offers the potential to achieve super-flexibility alongside a highly adjustable negative Poisson's ratio, setting it apart from any current graphene-based configurations [27]. Researchers demonstrated that graphene origami serves as an effective method for transforming graphene into complex nanostructures with atomic precision. Utilizing low-temperature scanning tunnelling microscope manipulation, they systematically folded and unfolded graphene nanoislands (GNIs) in a direction of their choosing [28]. Regarding graphene origami (GOri)-enabled metallic metamaterials, researchers discovered that incorporating a larger amount of GOri into the copper matrix not only results in a greater NPR but also enhances the elastic modulus of the nanocomposite [29]. These class of materials have highly tunable physical properties including thermal conductivity, Poisson's ratio, and Young's modulus [30, 31].

Graphene origami-enabled auxetic metamaterials represent a novel frontier in auxetic materials, offering unprecedented mechanical characteristics that are pivotal in the evolution of engineering applications. With graphene's exceptional strength and flexibility, combined with the tailored geometrical properties of origami, this paper introduces the transformative potential of such composites. Although there are other review papers regarding graphene origami and its potential, their main focus is on the chemical side of this material. Hence, we will have more of a mechanical view, meaning the mechanical characteristics of graphene origami-reinforced composites including their buckling and vibration behaviour in this article. It outlines the objective to holistically review and distil the amalgamation of current research about GOri mechanics, manufacturing processes, and theoretical models.

2. Theoretical Background

Auxetic metamaterials are distinguished by their counterintuitive property: a negative Poisson's ratio, expanding laterally when stretched, unlike conventional materials. Graphene, a single layer of carbon atoms arranged in a hexagonal lattice, brings unique electronic and mechanical benefits. When applied with origami principles, it metamorphoses into 3D structures with tailorable characteristics - an interplay of art and science resulting in materials with programmable properties.

2.1. Auxetic Metamaterials

Usual mechanical metamaterials are known for their negative values like negative Poisson's ratio (NPR), negative compressibility (NC), or negative normal stress [4]. Among the various kinds of mechanical metamaterials, auxetic materials stand out due to their non-standard way of changing shape. For example, regular materials tend to get thicker (or thinner) in the sides when you compress (or stretch) them. But, auxetic materials do the opposite - they get thinner (or thicker) when compressed (or stretched), just like in Figure 1. This unique trait has caught the attention of more and more scientists, leading to a rise in research and publications in the area of auxetic materials and structures.

When subjected to tension and compression, auxetic materials and configurations demonstrate non-standard deformation behaviours. Drawing from established principles of elasticity theory, various characteristics of materials are influenced by Poisson's ratio (ν). A straightforward and commonly cited instance of this relationship is seen in the calculation of the shear modulus, denoted as G :

$$G = \frac{E}{2(1+\nu)} \tag{1}$$

Where E stands for Young's modulus.

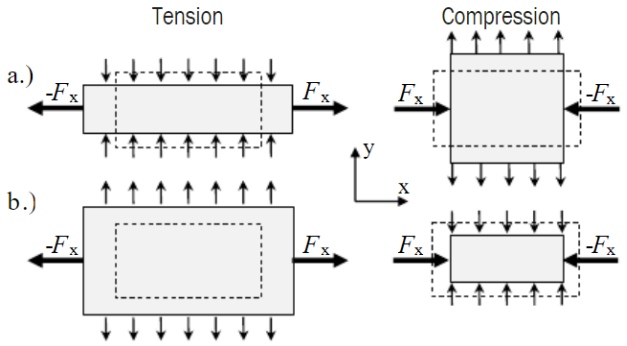

Figure 1: Responses to tensile and compressive forces: (a) for non-auxetic material; (b) for auxetic material [32].

It is apparent that when the Poisson's ratio ν trends toward −1, a boundary for isotropic substances, the corresponding shear modulus markedly escalates toward infinity. Other such properties that exhibit this inclination towards boundless increase as ν nears −1 encompass resistance to indentation, endurance against thermal shock, and robustness against fractures. These enhancements have been assessed and noted to improve significantly. The following sections will delve into these aspects more thoroughly, underlining the improved characteristics of auxetic materials owing to their negative Poisson ratio. These atypical properties confer a range of sought-after features, including enhanced resistance to shearing forces [33], improved capacity to withstand indentations [34], greater fracture toughness [35], synclastic curvature under stress [36], adjustable filtration capabilities [37], and more efficient energy dissipation and absorption [38-42]. Numerous studies have taken the remarkable properties of auxetics into account and analysed the effect of their incorporation into other materials [43-46].

2.2. Graphene Origami (GOri)

Graphene origami refers to the theoretical and experimental study of folding graphene, a single layer of carbon atoms arranged in a two-dimensional honeycomb lattice, into various structures. This concept borrows from the traditional Japanese art of paper folding, applied at the nanoscale to create three-dimensional configurations from two-dimensional materials.

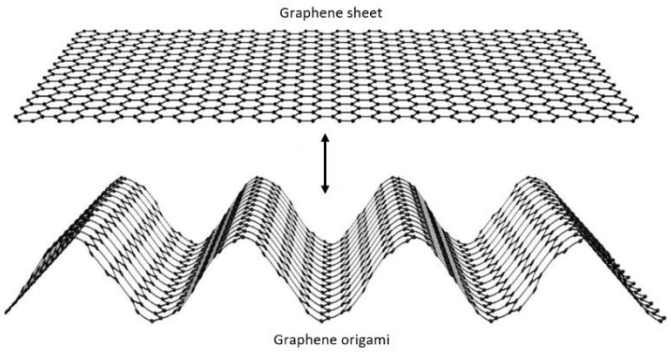

Figure 2: 2D graphene sheet into 3D origami structure.

The theoretical background of graphene origami involves understanding the material's mechanical properties, its elastic and plastic deformation behaviours, and the effects of such deformations at the atomic scale. The geometric configuration of graphene origami plays a pivotal role in determining the mechanical behaviour and performance of GOri-based auxetic metamaterials. The specific folding patterns, such as the type of crease or fold angles, and the scale of the origami structures directly influence the material's flexibility, strength, and auxetic properties (i.e., negative Poisson's ratio). For instance, variations in the folding angle or the number of folds in a given pattern can

significantly impact the material's response to tensile and compressive stresses. Additionally, Graphene is noted for its exceptional strength and flexibility, which makes it an ideal candidate for precise folding and manipulation. Graphene can exhibit reversible elastic deformation, which is essential in origami to ensure that structures can be folded and unfolded without permanent damage. Figure 2 demonstrates graphene origami fabricated from a regular 2D graphene sheet.

3. Synthesis and Fabrication Techniques

Methods of GOri synthesis have evolved since the creation of its simulations. Here we undertake various fabrication methods for achieving the 3D structure from graphene sheets.

3.1. thermos-responsive self-folding

Drawing inspiration from origami artistry, Figure 3 illustrates a thermally activated technique for the reversible construction and deconstruction of monolayer graphene into complex three-dimensional shapes [47]. In this process, the graphene's surface is coated with nanoscopic films of polydopamine and thermally sensitive poly (N-isopropyl acrylamide) (PNIPAM) layers. Precision micropatterning of this composite leads to autonomous origami-like folding into coordinated three-dimensional configurations, driven by the temperature-responsive fluctuations of the PNIPAM layers as they alternate between contracted and expanded phases. Upon elevating the temperature to approximately 45°C within an aqueous environment, a designed graphene structure mimicking a flower spontaneously closes its petals inward, transitioning from an open to a closed conformation. Conversely, cooling the structure down to 25°C induces the folded graphene flower to revert to its original unfolded stance. This self-assembly method not only sculpturally transforms graphene but also allows for the modulation of its electrical attributes. In its flat form, the material exhibits a direct current (I)–voltage (V) relationship with a consistent sheet resistance. However, the act of folding morphs the I–V profile into a non-linear characteristic, reflecting the altered electronic properties at the fold interfaces [25].

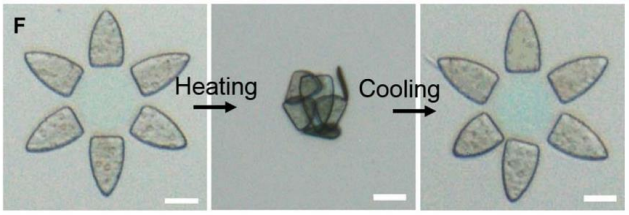

Figure 3: Reversible self-folding of graphene [47].

3.2. folding by growth and patterned transfer

In their innovative study, Kim et al. [48], outlined approaches for engineering multi-fold geometries within graphene sheets, which they aptly named 'grafolds,' as depicted in Figure 3. The intricate details of these configurations were made visible through transmission electron microscopy (TEM), conducted on graphene in both suspended forms and supported by substrates. Impressive control over the fold patterns was achieved by skilfully varying the surface curvatures throughout the processes of graphene synthesis or its subsequent transfer stage. A particularly effective method for achieving precise fold patterns was initiated by synthesizing graphene on electron-beam lithographed copper substrates (as seen in Figure 3(a)); these copper surfaces catalyse the chemical vapor deposition (CVD) of graphene. Following the graphene formation and the subsequent removal of the copper foundation, the graphene was moved to homogeneous substrates. This transference led to consistent folding along the pre-etched copper designs, with the TEM images in Figure 3(b–c) showcasing these directional folds. Moreover, the research not only reveals the practical applications of specifically oriented grafolds but also paves the way for experimental verification of previously theoretical models. Such models have posited that the way multiple graphene layers are arranged and the interactions between these layers could considerably affect the material's electrical and optical attributes. The authors speculate that intricate multi-layer folding techniques—akin to creating pleats—could serve as a practical test bed for these theoretical projections, emphasizing the significant impact of stacking sequences and interlayer couplings in determining graphene's functional properties.

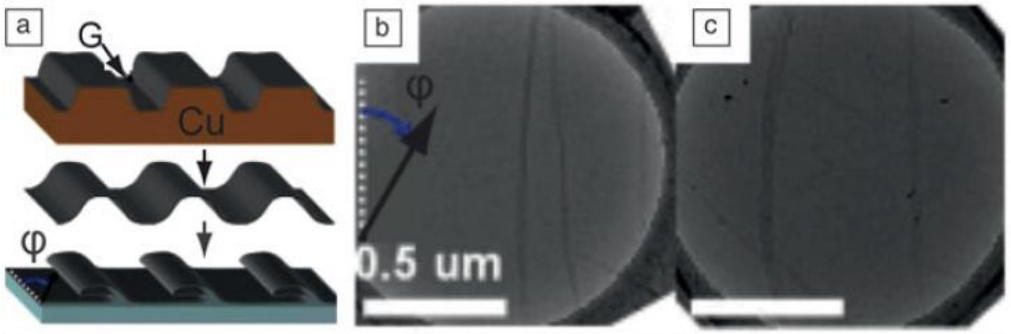

Figure 4: Graphene origami. (a) Directional control of folding formations in graphene during synthesis. Graphene synthesized using an etch-patterned copper substrate and transferred to Quantifoil holey carbon transmission electron microscopy (TEM) grids show (b–c) graphene folds along the direction of the copper patterns (vertical dark lines) [48].

3.3. Folding induced by capillary forces

Leveraging the power of molecular dynamics simulations, Patra et al. [49] (as shown in Figure 4(a)) have presented compelling evidence that the dynamics of water nanodroplets can initiate and influence the folding process in graphene nanostructures. Their research suggests that various modes of nanodroplet movement, including bending, sliding, rolling, or a zipper-like action, could induce the formation of both stable and temporary configurations in graphene such as layered stacks, encapsulated forms, intricate knots, and circular rings. The creation of these advanced 3D graphene constructs opens up possibilities for their integration into nanodevices with bespoke mechanical, electrical, or optical characteristics. However, a significant technical challenge lies in the precise positioning of nanodroplets to facilitate this process.

Further advancements in this realm were reported by Guo et al. [50], who explored the reversible hydration-induced folding and unfolding of uniform graphene oxide (GO) liquid crystal phases exhibiting extensive structural order (illustrated in Figure 4(b-c)). It was found that GO sheets with known lateral dimensions naturally arrange into nematic liquid crystal phases, which can be strategically organized into high-order supramolecular structures employing techniques such as surface anchoring, the manipulation of fluid dynamics, and deliberate micro confinement. The resulting orientations are predominantly influenced by the excluded volume effect and the adsorption energy due to the partially water-repellent properties of GO's basal planes, fostering a perpendicular orientation at the GO-water interface. Upon the process of dehydration, certain aligned GO liquid crystal phases solidify into fixed GO forms. Intriguingly, re-exposure to water causes the solid GO to swell disproportionately, regaining its pre-drying dimensions and form. This foldability and un-foldability are characteristics that set GO apart from other molecular frameworks.

The authors underscore the necessity for further study into alternative solvents, ionic interactions, and the impact of various drying rates to master the control of these transformational patterns. Utilizing these principles, a new generation of GO-based adaptive, stimulus-reactive materials can be designed. Specifically, the hydration-sensitive folding/unfolding traits of GO could prove instrumental in precision release mechanisms within nano/microelectromechanical systems, or as dynamic materials for volumetric expansion or sealing purposes, showcasing the material's potential in a wide array of technological applications [26].

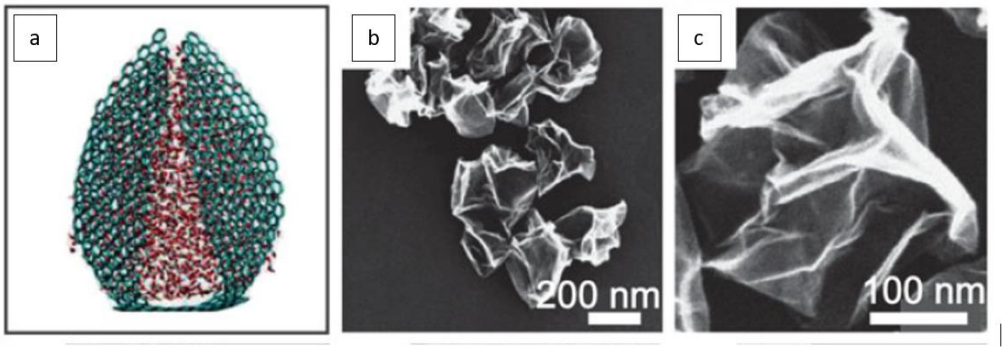

Figure 4: (a) Nanodroplet-assisted folding of a star-shaped graphene flake [49]. (b-c) Crumpled graphene nanoparticles fabricated by continuous microdroplet drying of colloidal graphene oxide suspensions. Scanning electron microscopy images show folded sheet structures and extended creases, whose sharp edges suggest plastic deformation [51].

3.4. folding by mechanical force

In a novel approach, Zhang et al. [52] successfully manipulated graphene sheets into folded structures by immersing them in a solvent and applying a 200-watt ultrasound treatment. This process exerts random but forceful mechanical energies on the material, as detailed in Figure 5.

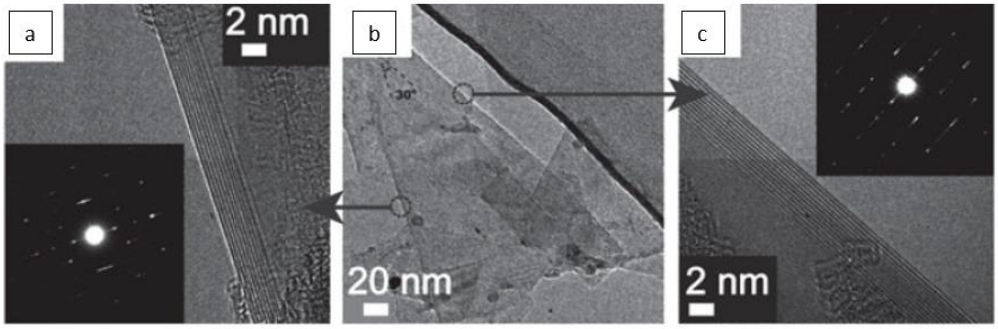

Figure 5: TEM image (center) of a multilayer graphene sheet with HRTEM images (left and right) of the two preferential folding directions formed by intense mechanical stimulation; the preferred folding directions are 30° apart. The diffraction patterns indicate that the left image (a) is an armchair folded edge, and the right image (c) is a zigzag folded edge [52].

The authors noted that while graphene displays considerable stiffness within its plane, it reacts with considerable flexibility to out-of-plane forces, which encourages folding along certain preferential axes when subjected to mechanical stress. A meticulous statistical analysis of 100 consistently folded edges illustrated that when faced with intense mechanical forces, free-floating graphene sheets displayed a proclivity for folding predominantly along the armchair (0°, 29 edges) and zigzag (30°, 27 edges) orientations. To provide theoretical backing for this folding pattern preference, Zhang et al. conducted comprehensive atomistic simulations to compute the energy states associated with various potential folded configurations. These simulations revealed distinct global and local minima in energy levels corresponding precisely to the 0°- and 30°-fold directions, thus giving a quantitative explanation as to why these specific orientations are favoured. These insights not only deepen the understanding of graphene's behaviour under force but also have significant ramifications for the controlled mechanical folding of graphene in future nanotechnology applications.

Regarding manufacturing cost and complexity, adding GOri to form auxetic metamaterials could potentially increase the complexity of manufacturing, as the unique folding and structural configurations of graphene origami require precise control over fabrication techniques. Nonetheless, combining graphene origami to form auxetic structures could lead to more efficient designs by optimizing material usage and enhancing mechanical properties (e.g., strength, flexibility, and energy absorption), potentially offsetting the initial increase in complexity. While the addition of GOri may introduce some challenges to scaling the production, its unique properties could offer significant advantages for industrial applications where performance, such as flexibility, lightness, and strength, is critical. Future developments in hybrid manufacturing processes, including automated folding and alignment techniques, may help bridge the gap between small-scale lab-based experiments and large-scale industrial production. The scalability could be further enhanced by combining GOri with other materials or manufacturing platforms that support high-volume production, enabling the incorporation of auxetic properties into practical applications.

4. Mechanical Characteristics

The flexibility and strength of GOri metamaterials surpass those of traditional composites, allowing for otherwise unachievable designs. Utilizing Timoshenko beam theory and computational models, the dynamic response of these materials reveals an ability to handle a range of stressors while maintaining integrity. Studies often employ Hamilton's principle to delineate motion equations, which provide analytical insight into the material's response under various conditions. Theoretical frameworks like Timoshenko beam theory and Hamilton's principle are instrumental in understanding the mechanical behaviour of graphene-origami metamaterials. Timoshenko beam theory accounts for both bending and shear deformations, providing a more accurate description of stress and strain in thin structures such as graphene-origami, which is essential for analysing stiffness and flexibility. In addition, Hamilton's principle facilitates the derivation of equations of motion, enabling the analysis of dynamic response and vibrational modes. Together, these frameworks support modelling and simulation, offering insights into nonlinear behaviours, characterizing material properties, and optimizing designs.

4.1. Flexibility and Strength

Utilizing molecular dynamics simulations, researchers have discovered that a nano-twinned copper (nt-Cu) matrix with closely spaced twins can induce distinctive wave-shaped wrinkles and sawtooth-like buckling in graphene structures when subjected to uniaxial compression. This nt-Cu matrix is characterized by a symmetrical lattice structure that favours a lattice rotation coinciding with the annihilation of dislocations, facilitating the transition of encapsulated graphene from two-dimensional to three-dimensional forms with high uniformity. The introduction of new twin boundaries (TBs) within the nt-Cu matrix enhances the ability of graphene to resist out-of-plane deformations, allowing the material to maintain stable wrinkling or buckling morphologies over a vast range of strains. These three-dimensional textured structures of graphene demonstrate significant flexibility, and their microstructural parameters can be precisely modulated by the application of varying levels of compressive strain. This precise control over the stable three-dimensional configurations of graphene through strain modulation underscores the potential for creating graphene-based materials with tailor-made textural attributes, paving the path for their exploitation in cutting-edge applications that require finely tuned mechanical characteristics [53].

In another article, Zhao et al. [15] analyse the buckling and post-buckling characteristics of functionally graded (FG) graphene origami (GOri)-enabled auxetic metallic metamaterial (GOEAM) beams. Using the Timoshenko beam theory, von Kármán type nonlinearity, and Differential Quadrature Method (DQM), the behaviours of FG-GOEAM beams have been studied. The study uses gradient-plasticity (GP)-aided micromechanical models to predict the material characteristics of layers within the FG beams. The research on FG graphene origami-enhanced auxetic metallic metamaterial beams reveal that incorporating graphene origami increases the buckling and post-buckling strength significantly compared to pure copper beams (Figure 6). The symmetrical distribution of graphene origami within the beams, especially when concentrated in outer layers, confers the highest resistance to buckling and improves post-buckling resilience. Although increasing the degree of folding in graphene origami decreases overall structural rigidity, these enhanced beams still outperform pure metal beams. Moreover, these beams show reduced sensitivity to temperature changes, highlighting their advanced thermo-mechanical stability. The study indicates that the mechanical properties of functionally graded beams can be finely tuned using graphene origami, offering an innovative approach to designing robust, high-performance engineering structures.

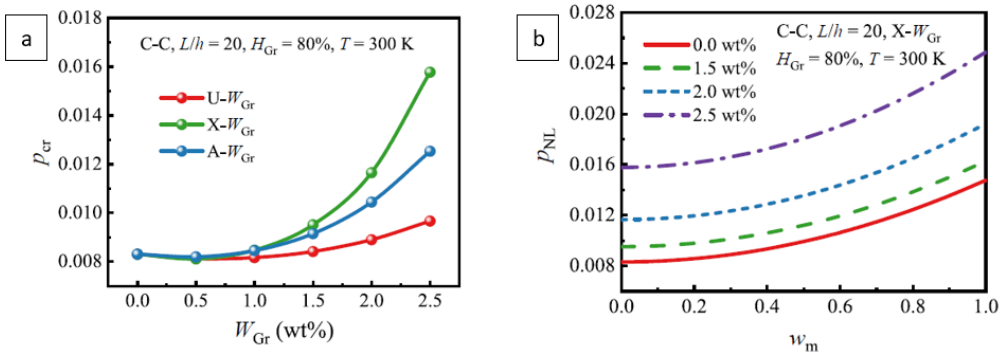

Figure 6: (a) Effect of GOri content on the dimensionless critical buckling loads of FG-GOEAM beams with various GOri distribution patterns. (b) Effect of GOri content on post-buckling paths of FG GOEAM beams [15].

Forecasting the material properties of complex composites presents a significant hurdle when relying on conventional micromechanical models. To surmount this challenge, Zhao et al. [54] introduced an advanced micromechanical modelling technique that merges molecular dynamics (MD) simulations with a genetic programming (GP) algorithm. This innovative GP-enhanced Halpin-Tsai model is meticulously calibrated using data derived from MD simulations, allowing for the precise prediction of Young's modulus in graphene origami/Copper (GOri/Cu) metamaterials. The model skilfully accounts for variations in graphene origami folding degrees, graphene concentrations, and operating temperatures. It achieves an impressive coefficient of determination (R^2) close to 0.95, exemplifying the high accuracy and efficiency of the proposed modelling approach in predicting the elastic properties of GOri metamaterials. The characteristic features of the material including Young's modulus (E), Poisson's ratio (ν), the Coefficient of Thermal Expansion (α), and the density (ρ) of the GOEAMs are characterized by the GP-assisted micromechanical models delineated in the following section as [54]:

$$E = \frac{1+\xi\eta V_{GR}}{1-\eta V_{GR}} E_{Cu} \times f_E\left(H_{GR}, V_{GR}, T\right) \tag{2}$$

$$\nu = \left(\nu_{GR}V_{GR} + \nu_{Cu}V_{Cu}\right)\times f_{\nu}\left(H_{GR}, V_{GR}, T\right) \tag{3}$$

$$\alpha = \left(\alpha_{GR}V_{GR} + \alpha_{Cu}V_{Cu}\right)\times f_{\alpha}\left(V_{GR}, T\right) \tag{4}$$

$$\rho = \left(\rho_{GR}V_{GR} + \rho_{Cu}V_{Cu}\right)\times f_{\rho}\left(V_{GR}, T\right) \tag{5}$$

Where the factors of material (η), and geometry (ξ) can be calculated in the following form [55]:

$$\eta = \frac{\left(\frac{E_{GR}}{E_{Cu}}\right) - 1}{\left(\frac{E_{GR}}{E_{Cu}}\right) + \xi} \tag{6}$$

$$\xi = \frac{2l_{GR}}{t_{GR}} \tag{7}$$

In which l_{GR} , and t_{GR} relate to the length and thickness of the GOri, respectively. The transformation functions denoted by $f_{E,\nu,a,\rho,}\left(H_{GR}, V_{GR}, and\, T\right)$ were deduced by applying the GP algorithm instituted by Zhao and colleagues. These functions encapsulate the interrelations of the physical properties within the material system as influenced by variables such as *H* for the amount of covered H atoms (0%-100%) which is an indicator to measure the GOri folding degree, *V* for volume, and *T* for temperature. These functions are formulated as [54]:

$$f_{E}\left(H_{GR}, V_{GR}, T\right) = 1.11 - 1.22V_{GR} - 0.134\left(\frac{T}{T_0}\right) + 0.559V_{GR}\left(\frac{T}{T_0}\right) - 5.5H_{GR}V_{GR} + 38H_{GR}V_{GR}^{2} - 20.6H_{GR}^{2}V_{GR}^{2} \tag{8}$$

$$f_{\nu}\left(H_{GR}, V_{GR}, T\right) = 1.01 - 1.43V_{GR} + 0.165\left(\frac{T}{T_0}\right) - 16.8H_{GR}V_{GR} - 1.1H_{GR}V_{GR}\left(\frac{T}{T_0}\right) + 16H_{GR}^{2}V_{GR}^{2} \tag{9}$$

$$f_{\alpha}\left(V_{GR}, T\right) = 0.794 - 16.8V_{GR}^{2} - 0.0279\left(\frac{T}{T_0}\right)^{2} + 0.182\left(\frac{T}{T_0}\right)\left(1 + V_{GR}\right) \tag{10}$$

$$f_{\rho}\left(V_{GR}, T\right) = 1.01 - 2.01V_{GR}^{2} - 0.0131\left(\frac{T}{T_0}\right) \tag{11}$$

In the field of engineering, the ability to customize materials with negative thermal expansion (NTE) was recognized as crucial for various applications. However, the options for significant tunability in the coefficient of thermal expansion (CTE) within metal matrix composites (MMCs) were notably limited. Zhao and their co-workers [56] addressed this limitation by formulating novel folded graphene (FGr) structures aimed at tailoring the CTE of MMCs, which were then embedded into a copper (Cu) matrix via molecular dynamics (MD) methods. In addition to developing these NTE materials, this research analysed their thermal buckling resistance capabilities, applying the classical Euler-Bernoulli beam theory combined with the Ritz method. Atomistic investigations, conducted through MD simulations, demonstrated that the CTE of FGr/Cu nanocomposites could be effectively adjusted by applying mechanical prestress and varying the temperature, especially in the temperature span from 200 K to 400 K. With the specialized origami-like patterning of FGr utilized by the researchers, the maximum observed negative and positive CTE values were approximately −95.42 × 10−6 K−1 and 56.32 × 10−6 K−1, respectively, under a prestress of 1000 MPa at 300 K and 200 K. More significantly, a quantifiable relationship for the temperature- and prestress-dependent material properties of the FGr/Cu composites was established. Building on this material model, the FGr/Cu composite beams exhibited a considerable enhancement in thermal buckling performance when compared to pristine

graphene/Cu beams, signalling an advance in the development of composite materials with improved thermal stability profiles.

Moreover, the bending and buckling behaviour of graphene origami (GOri) metamaterial irregular plates were investigated under various boundary conditions [57]. These analyses were conducted based on the higher-order shear deformation theory. With special attention devoted to the impact of nonlocal parameter ratio (NPR), another study examined the buckling behaviour of functionally graded graphene oxide-enhanced aerogel (FG-GOEAM) beams with variable thickness, which were submerged in a fluid environment [58]. They reported that H coverage on a beam result in enhanced auxetic properties, which in turn cause a reduction in both the critical buckling load as a consequence of the decreased stiffness of the beam.

Another aspect of structure stability is the GOri-reinforced structures'' fatigue response. While graphene origami offers significant enhancements in flexibility and strength, the unique folding and unfolding mechanisms that provide these benefits may also introduce fatigue-related concerns. To address these challenges, ongoing research is exploring ways to improve the fatigue resistance and long-term stability of GOri materials. This includes the development of composite GOri structures, where graphene is combined with other materials that offer better fatigue resistance or enhanced durability [59]. Additionally, the optimization of folding patterns, such as using more gradual or less sharp folds, could help to reduce stress concentration and enhance the material's performance under cyclic loading. Coating or surface treatment techniques may also be employed to improve the material's resistance to wear and environmental degradation. Additionally, perforating crease lines has been widely adopted as a practical method for designing thick engineering origami structures. Perforations inevitably reduce crease stiffness and thus affects the mechanical characteristics and fatigue performance of origami structures [60]. Utilizing this concept for thick origami structures and inculpating the concept for thin graphene sheets may enhance the fatigue response of GOri structures.

4.2. Dynamic Response

The FG-GOEAM beam presents a distinct benefit in obtaining extensive adjustability in the vibrational attributes of beam frameworks via the manipulation of Gori parameters (Figure 7). The theoretical analysis indicates that combining the *X*-W_{Gr} and *X*-H_{Gr} patterns significantly boosts the stiffness and natural frequency of FG beams. Increasing the GOri content to create a metamaterial substantially enhances the natural frequency while reducing dynamic deflection. Despite a rise in forced vibration amplitude with more H coverage, it is still considerably less than that of a pure Cu beam, highlighting the dynamic robustness of FG-GOEAM beams. Moreover, these beams show a diminished sensitivity to temperature changes compared to pure Cu beams, indicating their strong structural integrity [61].

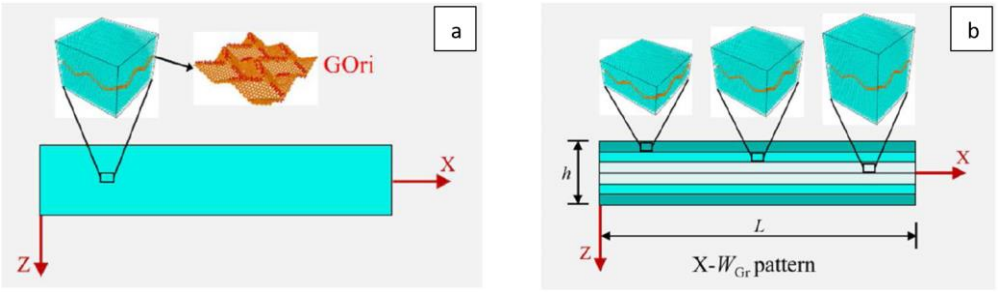

Figure 7: Isotropic homogeneous GOEAM beams and FG-GOEAM beams with gradients in (a) graphene folding degree and (b) graphene content and X distribution pattern [61].

Moreover, the integration of GOri allows for the adjustment of the vibrational behaviour of the GOEM plate's architecture, providing valuable insights for the development of high-efficiency engineering constructs. Increasing the weight fraction of GOri content transforms GOri/Cu composites into auxetic metamaterials with a negative Poisson's ratio, remarkably enhancing the natural frequency of FG-GOEM plates. Among all four distribution patterns, the *A* pattern exhibits the highest natural frequency capability and increases the natural frequency with the growth of GOri content. In the case of GOri folding degree patterns, *A*-H_{Gr} has the highest natural frequency values followed by *X*, *U*, and *O*. As the folding degree of GOri increases, there is a decline in the dimensionless natural frequency trajectory of FG-GOEM plates (Figure 8) [62].

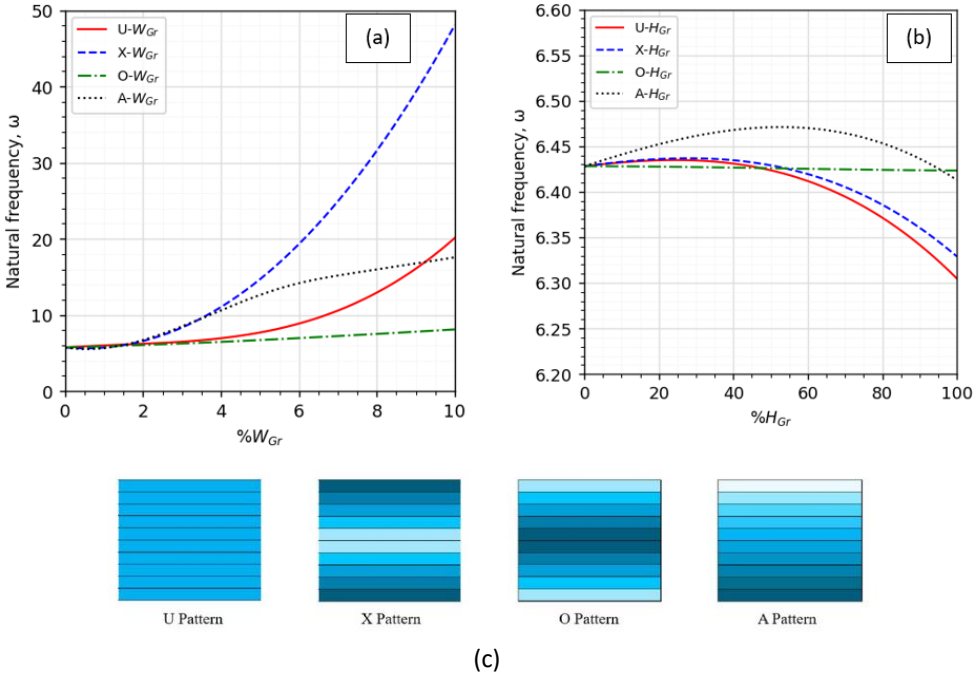

Figure 8: Variation of dimensionless natural frequency with respect to (a) GOri content and different distribution patterns, (b) different graphene folding degree patterns, and (c) GOri nanoparticles' distribution patterns in the copper matrix [62].

The evaluation of how metamaterials with negative Poisson's ratio (NPR) influence the nonlinear properties of functionally graded (FG) materials was carried out by Zhao et al. [63]. Their findings indicated that increasing the content of GOri leads to the creation of NPR mechanical metamaterials, resulting in FG-GOEAM beams that have greater linear and nonlinear vibration frequencies compared to pure copper beams. Beams with GOri closer to their surfaces are stiffer and exhibit higher nonlinear frequencies. Even with more GOri folding, which reduces the nonlinear frequency, FG-GOEAM beams still surpass pure Cu beams in nonlinear vibration characteristics. Layer-wise distribution of metamaterials across the beam lessens the impact of temperature on nonlinear vibration sensitivity. The adaptable nonlinear vibration features of FG beams are fine-tuned through GOri material parameters, aiding in the design of high-performance metamaterial structures. Alongside the previous studies, the free vibration analysis of a composite cylindrical panel was probed by Vali and Arefi [64]. The outcome of their work showed that boosting the graphene weight fraction in a cylindrical panel raises its natural frequencies, as the increase in stiffness caused by additional graphene outweighs the corresponding increase in mass. Similarly, a higher porosity coefficient results in elevated natural frequencies, since the induced stiffness increment surpasses the gains in density or mass. The study into the effect of graphene origami's folding degree reveals that more folding mildly reduces the natural frequencies, attributable to diminished structural stiffness. Thickness augmentation in porous layers of the panel leads to higher natural frequencies as structural stiffness is enhanced. Conversely, enlarging the span angle of the cylindrical panel results in lower natural frequencies due to reduced geometric stiffness. An examination into the impact of adding more graphene origami layers establishes that an increase in layer count leads to a rise in natural frequencies.

Furthermore, complex analyses of the nonlinear free vibration behaviour of fluid-immersed FG-GOEAM tapered beams have been thoroughly conducted [65]. The integration of GOri into the manufacturing process transforms GOri/Cu into a GOEAM metamaterial, which substantially improves the FG-GOEAM beams' performance under nonlinear vibration beyond that of traditional metal beams. Specifically, when GOri is strategically placed in the outermost layers of the beam, there's a notable increase in stiffness, leading to enhanced nonlinear behaviour. This metamaterial also allows for the adjustability of these responses in beams immersed in fluids. Furthermore, immersion in a fluid medium adds a dampening effect to vibrations while simultaneously increasing the beam's capacity to withstand post-buckling loads. Figure 9 demonstrates the dimensionless nonlinear free vibration of FG-GOEAM beams with clamped-clamped boundary conditions.

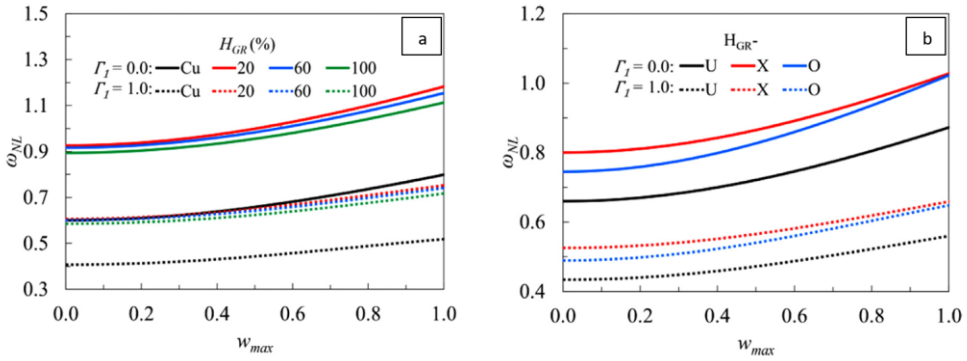

Figure 9: Dimensionless nonlinear free vibration of FG-GOEAM beams: (a) Effect of GOri content (*X-W_{GR}*); (b) Effect of GOri distribution pattern (C–C, *H_{GR}=100%, L/h=10, U-H_{GR}, W_{GR}=2.5%*) [65].

Their findings in another investigation revealed that incorporating GOri into a copper matrix substantially increases stiffness, improving the vibration resistance of FG-GOEAM composite beams, especially when GOri content exceeds 1.0wt%. A higher folding degree of GOri magnifies its auxetic behaviour but reduces stiffness, and although this lowers the fundamental frequency, it is still higher than in pure copper beams. Locating more GOri near the surface optimizes stiffness and fundamental frequency due to the high NPR. Density increases have a marginal added mass effect in shallow fluids or with a clamped-free beam configuration. Temperature variations have a minimal effect on the FG-GOEAM beam's vibration in fluid, which is not the case for pure copper or homogeneous metamaterial beams. Lastly, although beam thickness variation slightly impacts the fundamental frequency trend, a thicker beam is better at resisting deformation [66].

Last but not least, in the case of wave propagation in a functionally graded graphene origami-enabled auxetic metamaterial beam, Ebrahimi and Parsi [67] indicated that the dynamics of FG-GOEAM beams are significantly influenced by their material composition, with auxetic designs leading to higher wave frequencies but lower phase velocities. Increasing the content of graphene origami in the copper matrix amplifies stiffness and thus dynamic resistance, raising both wave frequency and phase velocity. However, a high folding degree in GOri enhances flexibility and auxetic behaviour, yet decreases stiffness, and consequently lowers both wave frequency and phase velocity. Distributing more GOri towards the outer layers enhances beam rigidity and wave frequency, benefiting from a higher negative Poisson ratio near the surface. Figure 10 delineates the effect of Gori content, distribution pattern, and Gori folding degree for the under-study beam. Despite temperature variations, GOri-infused metamaterials exhibit robust mechanical properties with stable wave frequencies and phase velocities. Furthermore, tweaking Winkler and Pasternak parameters can lead to incremental gains in wave frequency and phase velocity. Lastly, adjusting GOri material parameters offers a pathway to optimize wave propagation characteristics in advanced metamaterials, with potential applications in aerospace and defence.

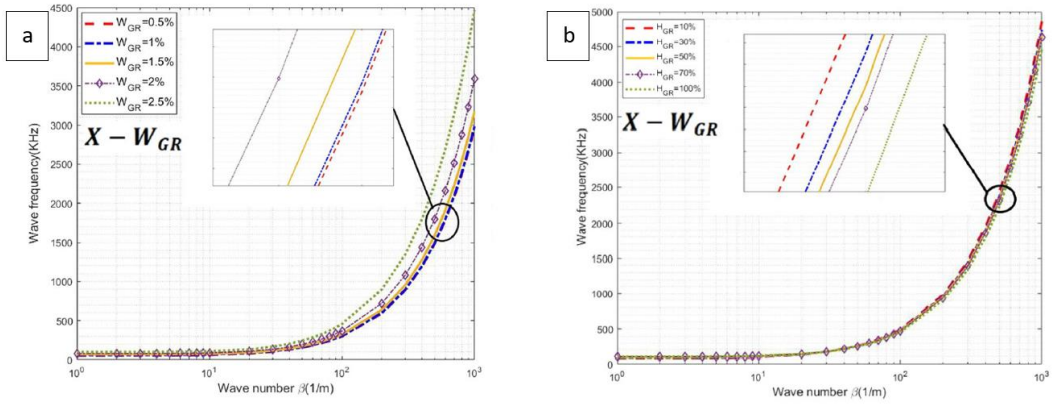

Figure 10: (a) Wave frequency variation vs number of waves for various GOri content (*X−W_{GR}, H_{GR}=100%*); (b) Wave frequency variation vs number of waves for various graphene origami folding degrees (*X−W_{GR}, W_{GR}=2.5%*).

5. Future challenges

Research on graphene origami faces several significant limitations and challenges that hinder its progress and application. One major hurdle is the difficulty in synthesizing high-quality graphene at scale, as contaminants and defects can adversely affect the material's mechanical properties. Precise fabrication of complex nanoscale structures is essential but challenging, with current techniques being costly and time-consuming. Furthermore, accurately predicting the mechanical behaviour of folded graphene proves complex due to variations in geometric configurations and the inherent nonlinear nature of the material. The need for high-resolution characterization tools, which are expensive and require specialized expertise, adds another layer of complexity to the research process. Additionally, understanding the long-term durability and stability of graphene origami structures under varying environmental conditions remains a challenge. Future efforts should focus on refining modelling techniques to better capture nonlinear elasticity and large deformations in GOri-based auxetic metamaterials. Emerging methods, such as nonlinear finite element analysis, machine learning models, and multi-scale simulations, show promise in overcoming current limitations. These approaches are expected to improve the accuracy of predictions for large deformations and nonlinear behaviours, thus making models more applicable to industrial-scale manufacturing and practical applications. Continued research into hybrid modelling approaches combining theoretical physics and materials science could lead to more robust models that address these challenges. Integrating graphene with other materials for practical applications can lead to compatibility issues, complicating the development of composite structures. Existing theoretical frameworks often fall short in capturing the unique behaviours of graphene origami, and the high costs associated with graphene production limit accessibility for researchers. Overall, overcoming these challenges necessitates interdisciplinary collaboration across materials science, engineering, and nanotechnology to unlock the full potential of graphene origami in various innovative applications.

6. Conclusion

This review underlines the transformative potential of graphene origami-enabled auxetic metamaterials. We undertake the background of such metamaterial along with the research advances in the fabrication of graphene origami along with the analytical investigations on the static and dynamic responses of composites reinforced with Gori. As research progresses, these materials promise not only to enhance existing products but also to catalyse new technologies altogether. The mechanics of these composites present a rich domain for exploration, and their increased adoption in practical applications awaits the resolution of current production and theoretical challenges. The intersection of materials science, mechanical engineering, and theoretical physics provides fertile ground for future research opportunities, particularly in optimizing material properties and expanding the range of practical applications.

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