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Review

Nanoreinforcement effects in multifunctional polyurethane foams—Scientific status hitherto and future

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Abstract: Polyurethane is a multipurpose polymer with valuable mechanical, thermal, and chemical stability, and countless other physical features. Polyurethanes can be processed as foam, elastomer, or fibers. This innovative overview is designed to uncover the present state and opportunities in the field of polyurethanes and their nanocomposite sponges. Special emphasis has been given to fundamentals of polyurethanes and foam materials, related nanocomposite categories, and associated properties and applications. According to literature so far, adding carbon nanoparticles such as graphene and carbon nanotube influenced cell structure, overall microstructure, electrical/thermal conductivity, mechanical/heat stability, of the resulting polyurethane nanocomposite foams. Such progressions enabled high tech applications in the fields such as electromagnetic interference shielding, shape memory, and biomedical materials, underscoring the need of integrating these macromolecular sponges on industrial level environmentally friendly designs. Future research must be intended to resolve key challenges related to manufacturing and applicability of polyurethane nanocomposite foams. In particular, material design optimization, invention of low price processing methods, appropriate choice of nanofiller type/contents, understanding and control of interfacial and structure-property interplay must be determined.

Keywords: polyurethane; nanocomposite; foam; manufacturing; properties; radiation shielding; shape memory; biomedical

1. Introduction

Polyurethane forms an important contribution to the thermosetting, thermoplastics, or elastomeric type of polymers due to the range of intrinsic physical features and advanced utilizations [1]. These polymers have flexibility of backbone variations by altering soft or hard units and probable hydrogen bonding between the segments [2]. Worth mentioning application areas of polyurethanes (as coatings, fibers, sponges) expand from defense and devices to medical sectors [3]. Moreover, advancements in the field of polyurethane materials can be seen in the form of nanocomposites with inorganic or carbon nanoadditives [4,5].

Abundant literature reports have been noted on preparation, physical aspects, and technical significance of polyurethane sponges or foams [6]. Similarly, polyurethane foams filled with different types of nanofillers have also been investigated for designs and applied attributes [7]. In this regard, most important types of nanofillers have been noted as graphene and carbon nanotube [8–10]. These hybrid foams have been manufactured by using variety of self foaming, free rising, foaming agent, freeze drying, in situ, solution, and chemical methods [11]. Consequently, nanocomposite foams own low density, flexibility, mechanical/compression strength, thermal features, other high tech features [12]. The high performance nanocellular polyurethane architectures have been applied for important applications concerned to radiation shielding, stimuli responsiveness, and biomedical sectors [13].

This up-to-the-minute review is planned to cover almost every physical and practical facet of polyurethane nanocomposite foams, for first time in the literature so far. In this concern, basics, synthesis, categories of polyurethane hybrid sponges (polyurethane/graphene nanocomposite foams, polyurethane/carbon nanotubes nanocomposite foams), and applications (radiation shields, shape memory, tissue scaffolds) have been conversed. As per reported knowledge, future of polyurethane nanocomposite foams simply relies upon overcoming field challenges of facile processing, design and property optimization, ecofriendliness, and large scale processing.

2. Polyurethane and polyurethane foams

Polyurethane is a versatile polymer with thermoplastic, thermosetting, or elastomeric backbone structure [14]. Basically, a polyurethane main chain consists of carbamate or urethane links [15]. In the case of segmented polyurethanes, prepolymers with isocyanate functionalities have been developed to further react with diamine, dihydroxyl, or similar short chain bifunctional compound [16]. Consequently, segmented polyurethanes have two types of segmental units, i.e., isocyanate based hard segments and polyol based soft segments [17]. It is important to mention that secondary interactions or crosslinking may exist between polyurethane chains due to the presence of amine (-N-H) and carbonyl (-C=O) functionalities in the main chain [18]. Notable features of polyurethanes can be listed as mechanical strength, thermal stability, thermal conductivity, electrical conductivity, nonflammability, anticorrosion, chemical resistant, and so on [19]. Subsequently, applications of these remarkable macromolecules have been reported for thermal insulating materials, foams, gaskets/seals, packaging, building, electronics, and transportation to name a few [20].

One of the outstanding behaviors of polyurethanes have been noted as the formation of polymeric sponges or foams [21]. Polyurethane foams may have close or open cell microstructures [22]. These polymeric foams usually have the elasticity, low density, heat stability, heat conductivity, and nonflammability characters [23]. Practical uses of polyurethane foams have been observed for aerospace automobile interiors, industrial packaging, insulating materials, furnishing, and other areas [24–26]. For synthesizing polyurethane foams, numerous facile routes have been practiced, as per literature reports so far. Usually, the synthesis of polyurethane foam may involve reactions of isocyanate and polyols, as shown in **Figure 1**. An initial attempt by Saint-Michel et al. [27] reported the polyurethane foam fabrication using 4,4'-diphenylmethanediisocyanate and polypropylene triol in the presence of dibutyltin dilaurate (as catalyst). In this process, in situ produced carbon dioxide from polyisocyanate caused self foaming process [28]. The microstructural analysis revealed close shell cell nanostructures. Consequently, fine electrical conductivity and mechanical properties were observed form these polymeric sponges. Advancements in the field of polyurethane foams led to the development of nanoparticle reinforced hybrid materials [29,30]. **Figure 1** shows most probable reactions of isocyanate functionalities involved in the formation of polyurethane foams [31]. Herein, in situ production of carbon dioxide (key agent for self foaming) can be seen as a result of reactions between isocyanate groups and water [32]. **Figure 2A** shows scanning electron microscopy images of polyurethane foam having varying isocyanate index (0.88–1.1). With increasing values of isocyanate index, strength/integrity of cell walls seemed to be enhanced and porosity was decreased due to increasing viscosity, crosslinking, and foam reactions of the system. **Figure 2B** depicts relationships of tensile strength with isocyanate index and compressive strength vs. isocyanate index at 50% deformation and 20% deformation of foamed samples. According to these results, the linear relationships between the properties were observed due to enhancements in integrity of the cellular foam structure with rising isocyanate index. **Figure 2C** illustrates glass transition temperature vs. isocyanate index of polyurethane foam. Hither, glass transition temperature was found linearly dependent upon the isocyanate index of polyurethane foams, which may also affect their mechanical properties. It can be suggested that optimal temperature around ~23 ° must be used to attain desirable mechanical properties of these sponges.

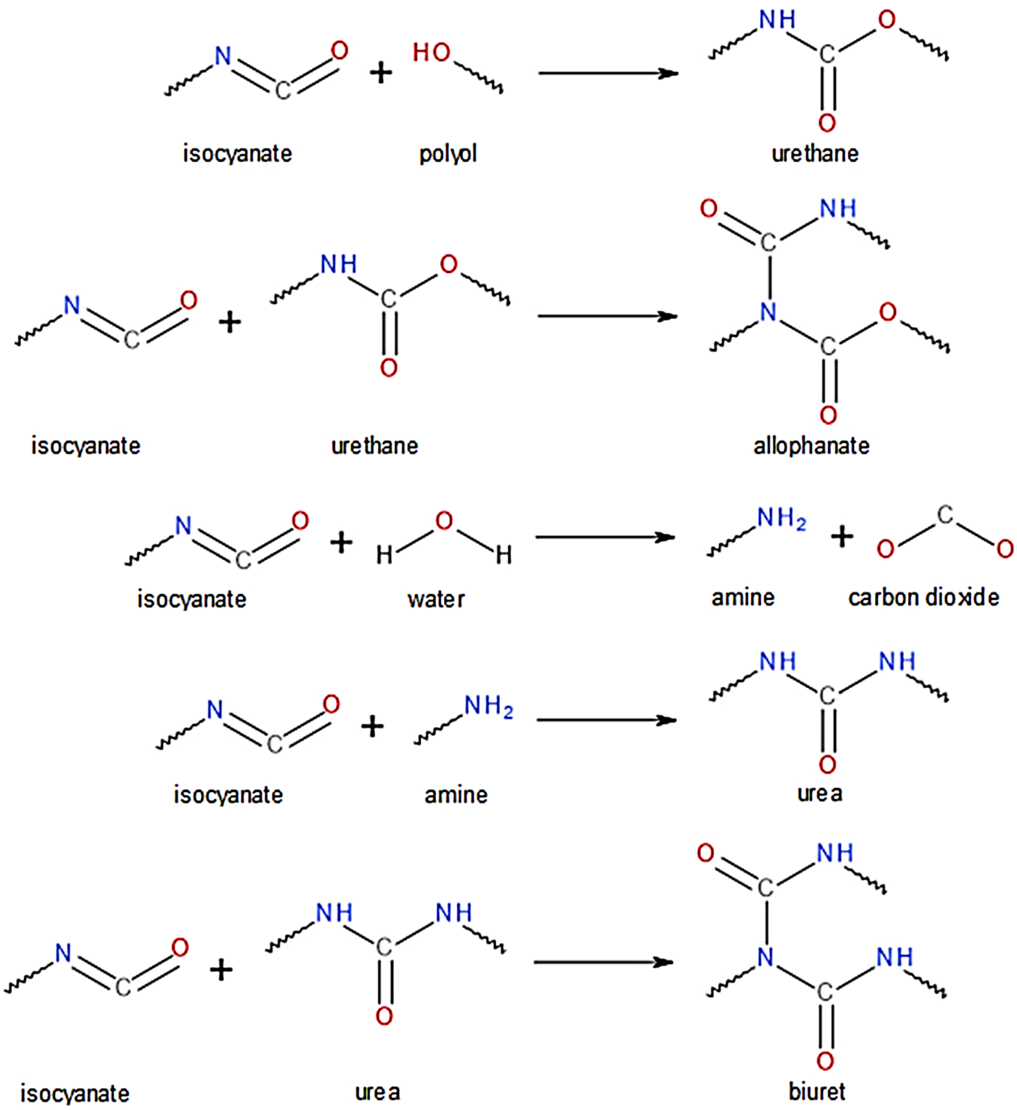

Figure 1. Common reactions involved in polyurethane foam manufacturing via isocyanate reactions [31]. Reproduced with permission from MDPI.

Besides, waterborne polyurethanes have been considered as an environmentally friendly type of polymers with solvent-borne backbone units [33,34]. These polymers have been studied for valuable thermal, mechanical, anticorrosion/antichemical, barrier, permeability, and other characteristics [35–37]. Consequently, waterborne polyurethanes have developed in the form of nanocomposites, foams, nanofibers, and other industrially viable materials been and their foams have been reported for advanced applications [38–40].

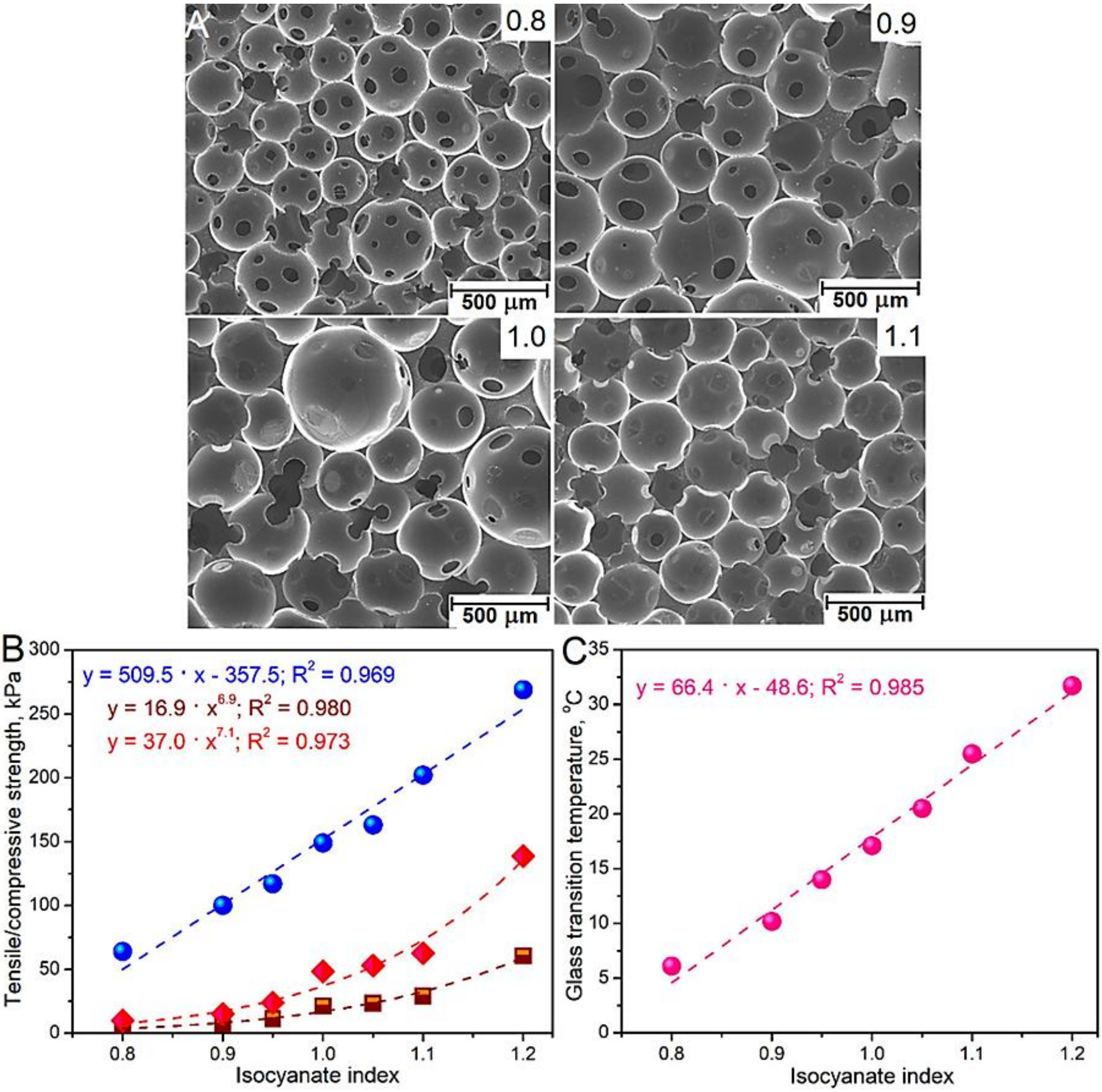

Figure 2. (**A**) Scanning electron microscopy images of polyurethane foam having varying isocyanate index; (**B**) tensile strength vs. isocyanate index (blue circles) and compressive strength vs. isocyanate index at 50% deformation (red squares) and 20% deformation (brown squares) of foamed samples; (**C**) glass transition temperature vs. isocyanate index of polyurethane foam [31]. Reproduced with permission from MDPI.

3. Polyurethane foams with carbonaceous nanoreinforcements

3.1. Graphene nanoreinforced polyurethane nanocomposite foam

Name of graphene appears first among the most remarkable nanocarbon discoveries [41]. Graphene occurs as a nanosheet of hexagonally organised sp^2 hybrid carbon atoms [42]. According to structural specifications, graphene is believed as a single layer out of a stacked graphite structure [43]. Since discovery, countless bottom up or top down strategies have been adopted to form two dimensional graphene nanostructure, including exfoliation, hydrothermal, vapor deposition, plasma/laser, and chemical or electrochemical routes [44]. Subsequently, scientific explorations on graphene unveiled a range of notable attributes, such as superior surface area, Young's modulus (~I TPa), thermal transport (~2000-5000 W/mK), electrical conduction (~200,000 cm2V-1s-1), and other valued characteristics [45].

Amid high-tech applications, worth of graphene has been noted in the fields of space/defense, energy devices (solar cells, fuel cells, capacitors, batteries), electronics (sensors, diodes), civil engineering, textile, environmental remediation, and medical areas [46,47].

Technical implications of graphene have been further enhanced in the form of polymeric hybrids using varying matrices [48]. In this regard, polyurethanes have also been applied as valuable matrices for graphene and derivative nanofillers [49]. Several high performance polyurethane/graphene nanocomposites have been designed and examined for physical properties and advanced industrial uses from energy and environment to biomedical devices [50]. Similar to polyurethanes, hybrid foams or sponges have been prepared with graphene reinforcements [51,52]. Among early attempts, Hodlur et al. [53] reported coating method for graphene deposition on polyurethane foam. The hierarchical sponges depicted fine percolation and electron conduction behavior under low applied pressures, e.g., ~0.5 atmospheres. Chen et al. [54] used curing method for the formation of polyurethane/graphene nanocomposite foam. Adding 5–20 phr graphene contents to polyurethane foam matrix exhibited significantly higher electrical conductivity (1.5×10^{-3} to 1.3 S cm^{-1}), than the unfilled foams (1.0×10^{-11} S cm^{-1}). These superior conductivity properties of hybrid foams seemed to be due to the formation of consistent three dimensional networks in these materials. Kim et al. [55] preferred catalyst foaming strategy to form polyurethane/graphene nanocomposite foam. These spongy nanomaterials revealed notable sound absorption properties. Herein, including 0.5 phr graphene nanofiller to polyurethane foam caused 7 times higher sound absorption coefficient than the unfilled foams.

Patole et al. [56] prepared a system based on polyurethane/poly(dimethyl siloxane)/graphene foams. **Figure 3A** shows a facile resin infiltration technique for the formation of hybrid foams. In this regard, initially polyurethane/graphene foam was formed using carbonization process. Later, poly(dimethyl siloxane) was impregnated on the nanocomposite foam to form polyurethane/poly(dimethyl siloxane)/graphene foam hybrids. **Figure 3B** illustrates scanning electron microscopy micrograph of the hybrid foam, where graphene can be observed with a defect free lattice structure. Such morphology confirmed the effectiveness of synthesis techniques applied to form these polymeric sponges. Moreover, the hybrid foam had electrical conductivity of ~2.9 S m^{-1}, due to the presence of three dimensional graphene architecture. **Figure 3C** shows functioning and resistance vs. time plot of a pressure sensor based on polyurethane/poly(dimethyl siloxane)/graphene foam. Pressure was applied using fingertip and resistance variations were measured with a multimeter. The resistance behavior was found directly related to the applied pressure over repeated cyclic process. Such pressure or strain sensors based on polyurethane foams can be useful for future soft robotics applications.

Zhong et al. [57] fabricated polyurethane/graphene oxide and polyurethane/reduced graphene oxide nanocomposite foams. For this purpose, a commercial polyurethane foam (40 × 40 × 30 mm^3) was coated with graphene oxide through continuous solution dipping plus squeezing processes (**Figure 4A**). The as prepared polyurethane/graphene oxide hybrid foam was treated with hydrazine hydrate (reducing agent) to form polyurethane/reduced graphene oxide nanocomposite sponge.

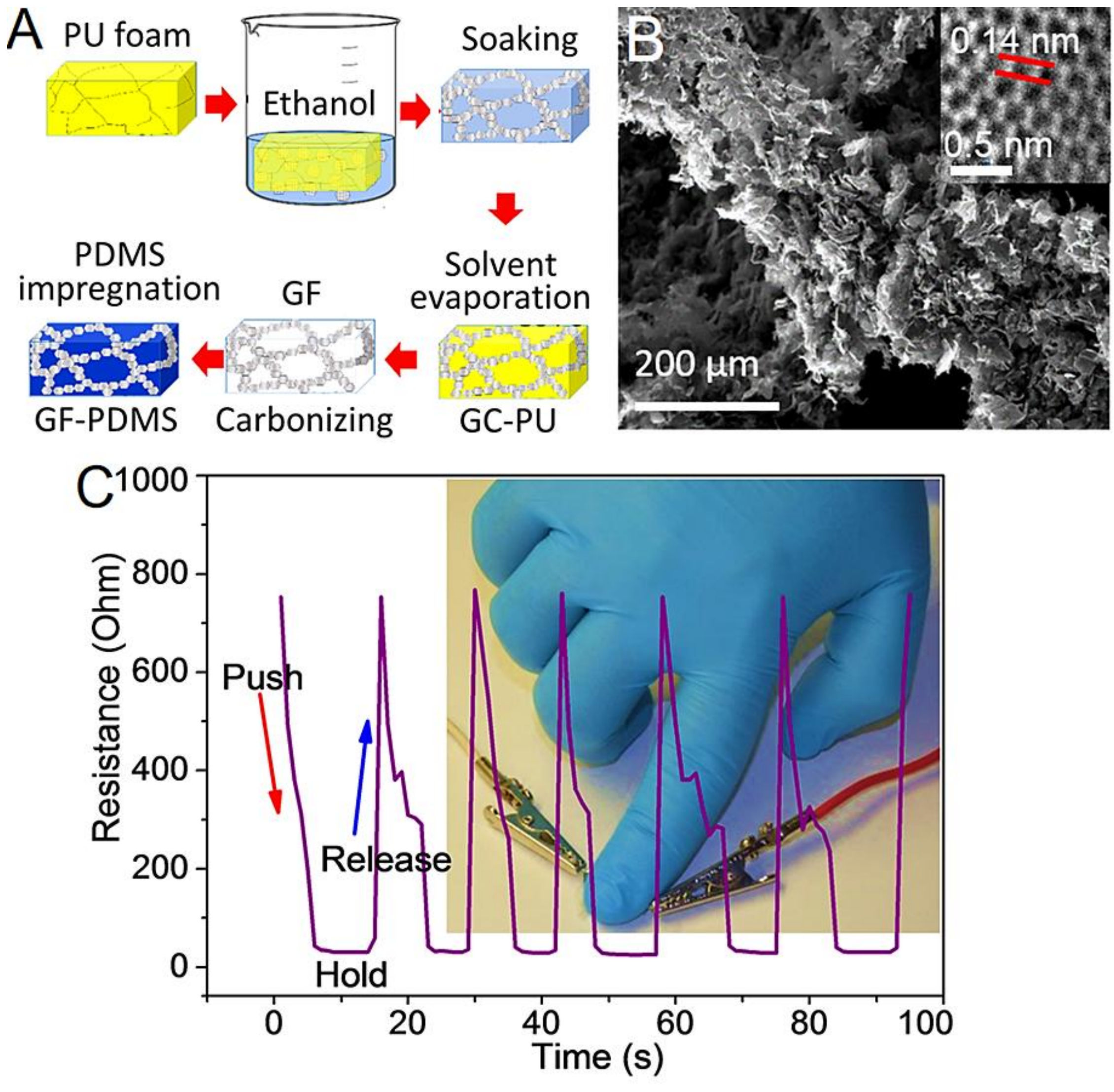

Figure 3. (**A**) Step wise fabrication of polyurethane/poly(dimethyl siloxane)/graphene foam; (**B**) scanning electron microscopy image of hybrid foam, inset: atomic-resolution image of the same with graphene crystal lattice; (**C**) resistance vs. time plot for polyurethane/poly(dimethyl siloxane)/graphene foam, inset: experimental setup for hybrid

based pressure sensor with fingertip for applying pressure [56]. PU = polyurethane; PDMS = /poly(dimethyl siloxane); GF = graphene foam; GF-PDMS = graphene foam-poly(dimethyl siloxane); GC-PU = graphene crystal-polyurethane. Reproduced with permission from ACS.

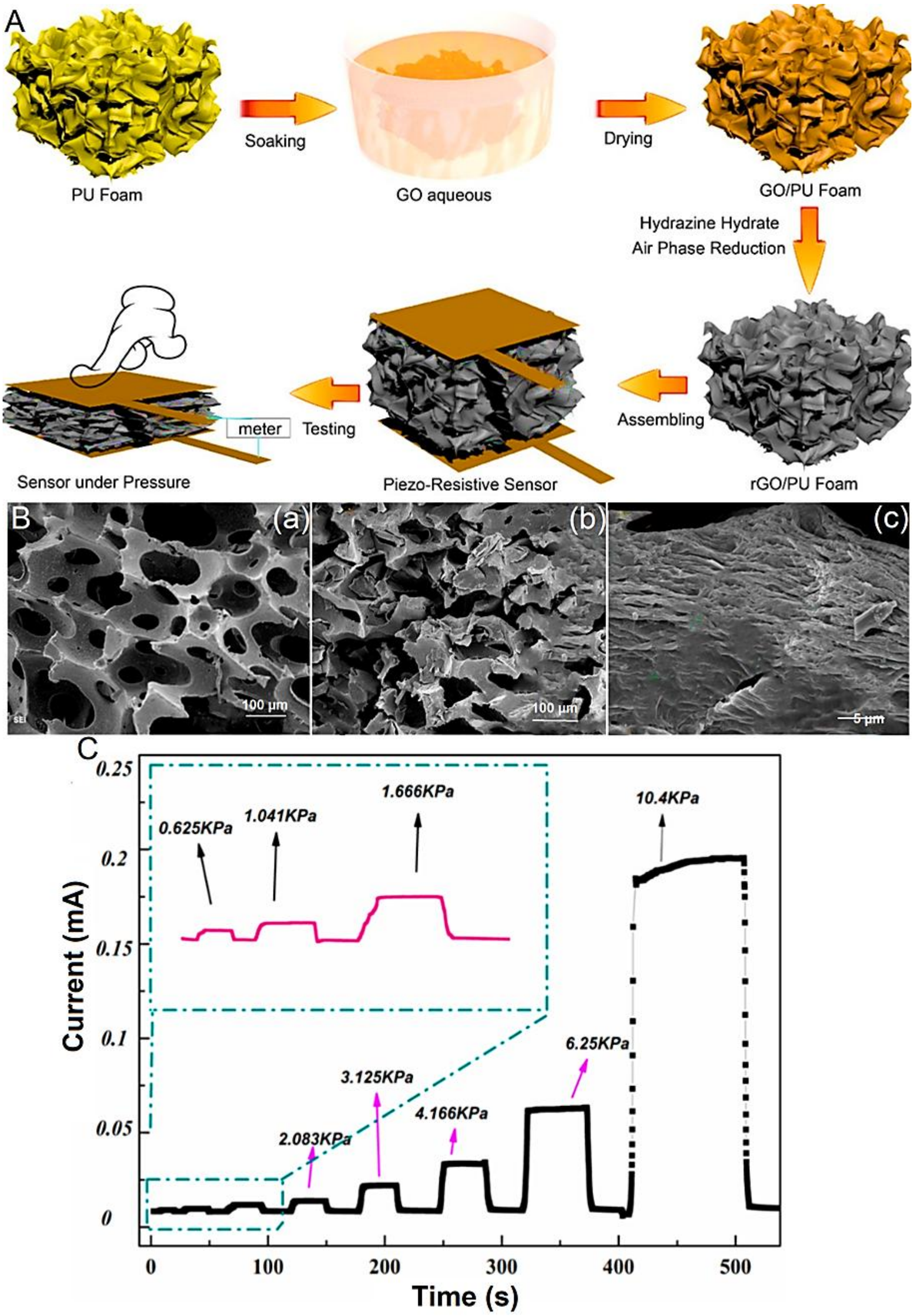

Figure 4. (**A**) Schematic of the formation of polyurethane and reduced graphene oxide based foam and derived pressure or piezo-resistive sensor; (**B**) scanning electron microscopy images of (a) pristine polyurethane foam; (b, c) reduced graphene oxide based polyurethane foam with different magnifications; (**C**) current vs. real time scan of polyurethane and reduced graphene oxide based nanocomposite foam under different applied pressures [57]. GO =

graphene oxide; PU = polyurethane; GO/PU = graphene oxide/polyurethane; rGO/PU = reduced graphene oxide/polyurethane. Reproduced with permission from MDPI.

Figure 4B a–c present scanning electron microscopy micrographs of pristine polyurethane foam and reduced graphene oxide filled polyurethane foam. In the case of pristine foam, uniform porosity and surface roughness was observed. This surface roughness was found beneficial for the adsorption of graphene oxide in the foam architecture. Consequently, polyurethane/reduced graphene oxide nanocomposite foam revealed typical graphene like wrinkled surfaces. Furthermore, **Figure 4C** shows a relationship between current and real time for polyurethane/reduced graphene oxide hybrid foam, with increasing applied pressures (0.62–10.4 kPa). It was observed that increasing pressure on the hybrid foam caused continuous rise in current stages due to signal-to-noise ratio and recyclability of the piezoresistive sensor.

3.2. Carbon nanotube filled polyurethane nanocomposite foam

Carbon nanotube is one of the most remarkable discovery (1991) in the field of nanocarbon nanoallotropes [58]. It is a one dimensional hollow cylinder shaped carbon nanotecture which is composed of sp^2 hybrid atoms [59]. This cylindrical nanostructure may exist as single walled or possess overlapping cylinders to form double walled, or multi walled carbon nanotubes [60]. The diameter of carbon nanotube can be as small as few nm, whereas length has been reported in the range of 100 nm to few µm [61]. Among common synthesis tactics, arc discharge, chemical vapor deposition, laser ablation, catalytic, and chemical approaches have been applied to form carbon nanotube [62]. The precisely designed nanocarbon nanostructures have superior aspect ratio, chirality, optical, electronic, electrical, magnetic, and thermal attributes [63,64]. Subsequently, an endless potential of carbon nanotube can be noted for defense/space, energy/electronics, coatings, construction, textile, sports, and biomedical areas [65–67].

Besides, carbon nanotube can form the most valuable type of nanocomposites with different polymeric matrices [68]. In this concern, notable scientific attempts can be seen regarding polyurethane and carbon nanotube derived nanocomposites [69]. Consequently, carbon nanotube reinforced thermosetting, thermoplastics, or biobased polyurethanes exhibited countless structural, thermal, mechanical, and tribological features; therefore leading to high end commercial acceptance [70]. Along the similar lines, carbon nanotube has also been reinforced in polyurethane foams to form high performance next level industrial hybrids. As compared to polyurethane/carbon nanotube nanocomposites, the derived hybrid foam revealed exceptional advantages of strength-to-weight ratio, mechanical firmness, flexibility, electrical percolation, thermal transport/stability, and other beneficial properties [71]. Therefore, polyurethane/carbon nanotube foams have been found promising for numerous industrial applications, where polyurethane nanocomposites were found least efficient [72]. Among initial scientific attempts, You et al. [73] used free rise foaming technique (cyclopentane as foaming agent) to form polyurethane/carbon nanotube hybrid foams. The resulting spongy nanomaterials developed efficient matrix-nanofiller links and percolation effects leading to reasonable electrical conductivity of about 0.2 Scm^{-1}.

Later, Zhai et al. [74] adopted facile water blowing practice to form carbon nanotube filled polyurethane foam. These hybrid foams revealed valuable compression based stress-strain features due to load transfer effects of increasing carbon nanotube contents. Espadas-Escalante et al. [75] applied blowing agent based free foam rising practice to design polyurethane/carbon nanotube foams. These spongy hybrids were tested for compressibility, heat conduction, and flame resistance attributes. Accordingly, adding carbon nanotube contents (0.1–2 wt.%) to polyurethane foams enhanced the flame stability by reducing the flame propagation speed. Huang et al. [76] adopted an innovative direction dependent freezing process for the formation of carbon nanotube reinforced thermoplastic polyurethane foams. **Figure 5A** a–c show complete steps, equipment, and mechanism for ice crystal growth involved in the freezing process applied for the formation of thermoplastic polyurethane/carbon nanotube foams.

Herein, use of direction dependent freezing led to the formation of aligned hybrid foam architecture. **Figure 5B** a–c depict scanning electron microscopy micrographs for pristine thermoplastic polyurethane sponges and thermoplastic polyurethane/carbon nanotube hybrid foams. These nanocomposite sponges revealed unique consistently aligned architectures due to the effectiveness of the manufacturing technique used. Hence, polyurethane/carbon nanotube hybrid foams formed unidirectional stairs like nanoarchitectures. Besides, **Figure 5C** displays a reversible compression behaviour of aligned (freezing method) and irregularly grown nanocomposite foams. As expected, aligned polyurethane/carbon nanotube hybrid foams revealed superior shape reattaining behavior after compression due to structural integrity and synthesis technique used. On the other hand, irregularly grown hybrid foam was suggested to have distorted cell structure and meagre shape recovery on compression cycles.

Guo et al. [77] formed pristine thermoplastic polyurethane and thermoplastic polyurethane/carbon nanotube nanocomposite foams using fused filament fabrication based three dimensional printing technique. **Figure 6A** demonstrates scanning electron microscopy micrographs of pristine thermoplastic polyurethane and thermoplastic polyurethane/carbon nanotube nanocomposite foams with 1 and 4 wt.% loading level. Relative to the unfilled foam, adding nanofiller contents reduced the cell sizes and enhance the number of cells in the hybrid foams. This effect was observed due to heterogeneous nucleation caused by the nanocarbon nanoparticles in the polyurethane spongy matrix. **Figure 6B** displays actual compression loading and release processes applied on the hybrid foam at varying compression rates. Accordingly, **Figure 6C** present relative current vs. time scan of 4 wt.% carbon nanotube filled thermoplastic polyurethane foam. Herein, a constant current changes over different applied compression rates were observed. Similarly, **Figure 6D** A shows a polyurethane/carbon nanotube nanocomposite foam based wearable sensor for gait recognition (linked to a multimeter). The changes in current were found directly linked to the variations in human gait.

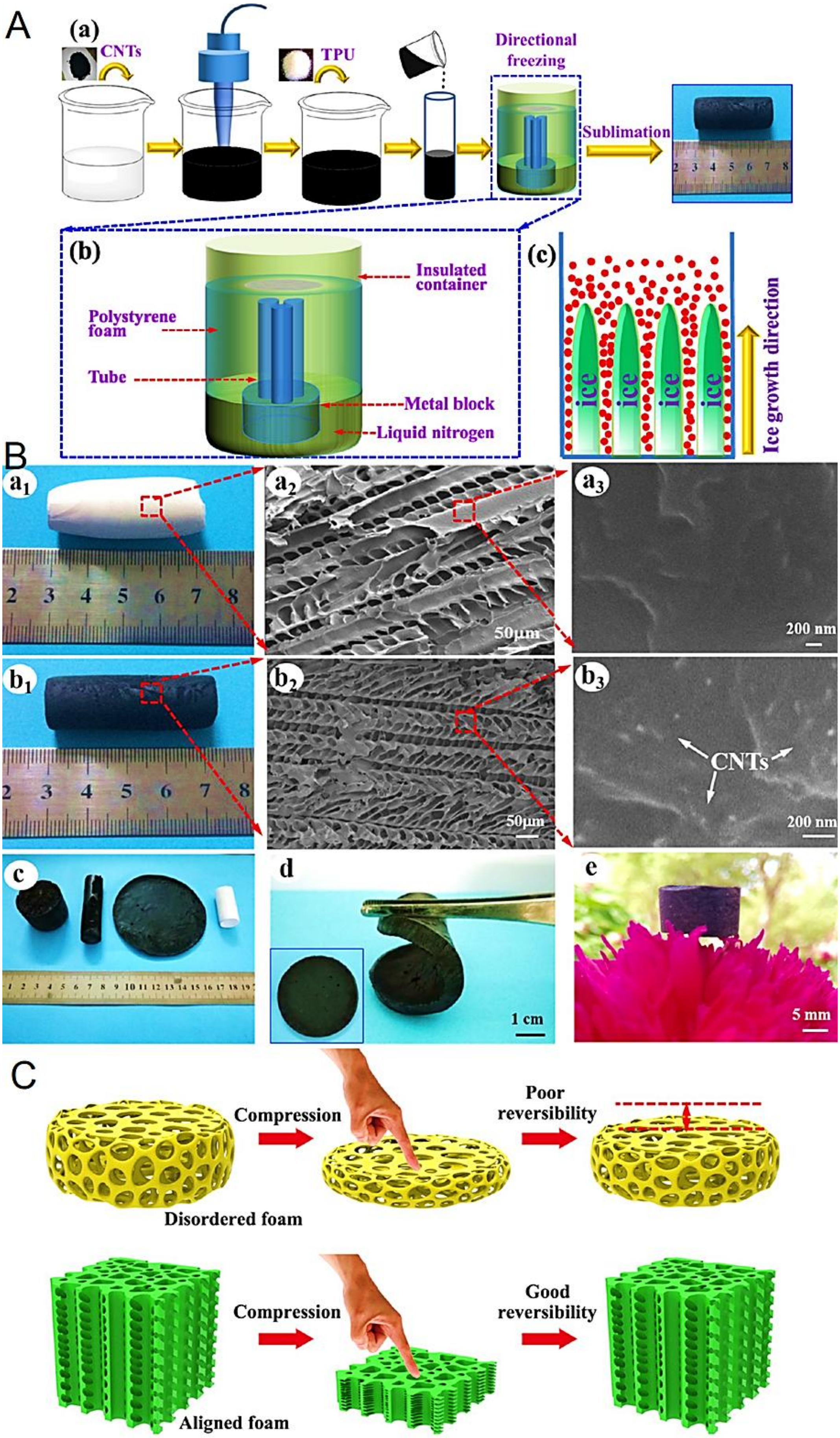
A
(a)
CNTs
TPU
Directional freezing
Sublimation
(b)
Insulated container
Polystyrene foam
Tube
Metal block
Liquid nitrogen
(c)
ice
ice
ice
ice
Ice growth direction
B
a1
a2
50μm
a3
200 nm
b1
b2
50μm
b3
CNTs
200 nm
c
d
1 cm
e
5 mm
C
Compression
Poor reversibility
Disordered foam
Compression
Good reversibility
Aligned foam

Figure 5. (**A**) (a) Manufacturing of thermoplastic polyurethane/carbon nanotube hybrid foam by freezing technique; (b) freezing equipment used; (c) a schematic of process showing directional freezing and growth of ice crystals; (**B**) scanning electron microscopy images of ($a_{1–3}$) unfilled thermoplastic polyurethane foams; and ($b_{1–3}$) thermoplastic polyurethane/carbon nanotube foams; (c–e) as prepared samples of conducting thermoplastic polyurethane/carbon nanotube foams; (**C**) comparative models showing reversibility processes for aligned and disordered thermoplastic polyurethane/carbon nanotube nanocomposite foams [76]. CNTs = carbon nanotubes; TPU = thermoplastic polyurethane. Reproduced with permission from ACS.

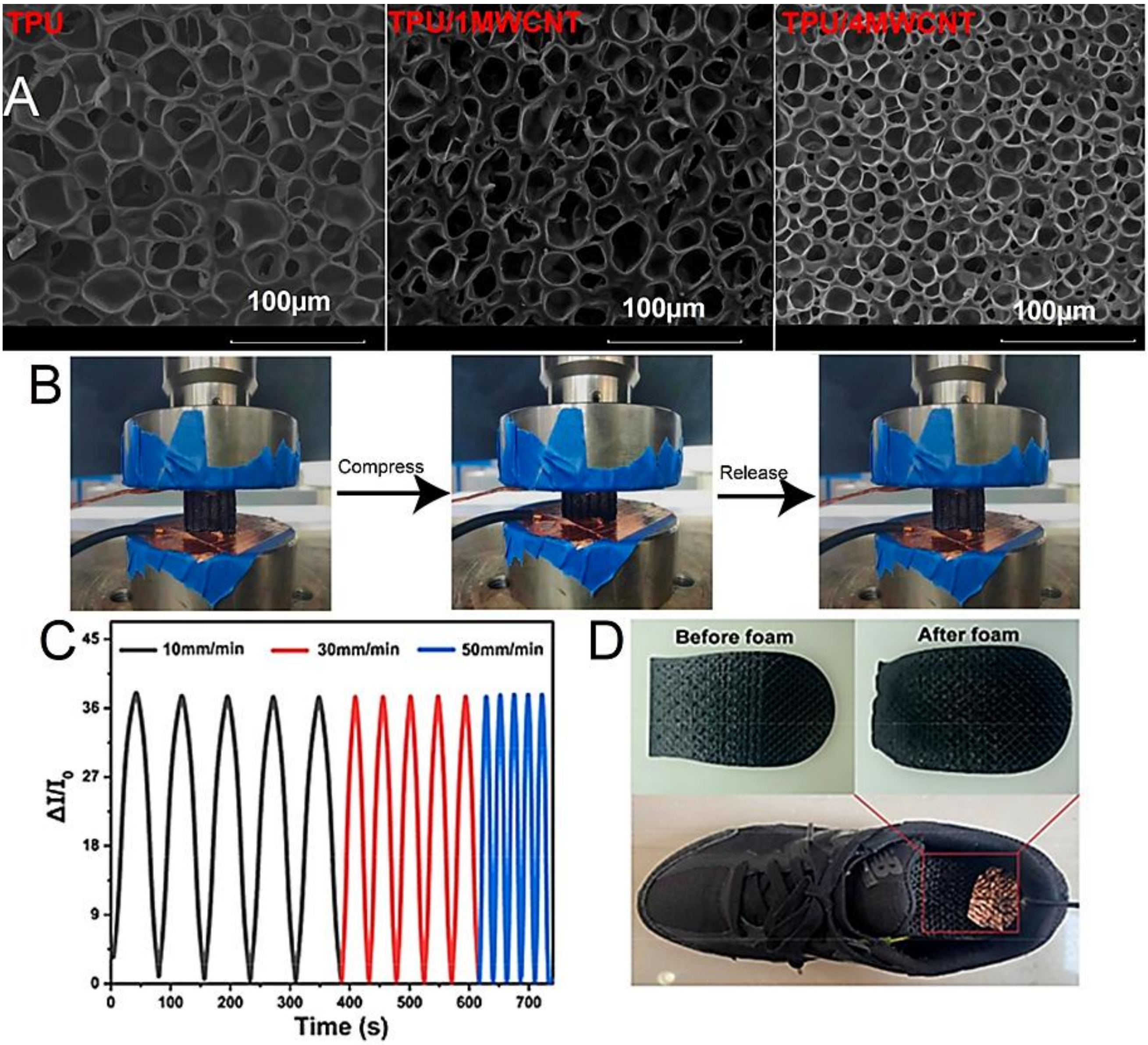

Figure 6. (**A**) Scanning electron microscopy images of pristine TPU foam and TPU/MWCNTs nanocomposite foam (1 & 4 wt.%), left to right, respectively; (**B**) compression loading and releasing stages of the hybrid foam; (**C**) relative current vs. time plot of TPU/MWCNTs at varying compression rates; (**D**) TPU/MWCNTs nanocomposite foam based plantar wearable sensor for gait recognition [77]. TPU = thermoplastic polyurethane; TPU/MWCNTs = thermoplastic polyurethane/multiwalled carbon nanotubes. Reproduced with permission from MDPI.

4. Technical significance of polyurethane/carbonaceous nanocomposite foams

4.1. Radiation shielding

Hazardous effects of continuously rising radiation pollution generated by functional electronics and other devices have been observed for the entire ecosystem (human beings, animals, vegetation, electronic systems) [78,79]. To cope the damaging influences of electromagnetic radiations, several solutions have been proposed, including the use of high performance materials/nanomaterials shields [80,81]. In this regard, polymers as well as derived nanocomposites have gained enormous worth to design high tech radiation shields [82]. For polymeric nanocomposites, carbonaceous nanoreinforcements like graphene or carbon nanotubes have attained scientific curiosity to deal with the environmentally interfering radiations [83,84]. Furthermore, polyurethane has been studied as an important matrix material to deal with the challenges of electromagnetic, gamma, or nuclear rays [85]. Particularly, polyurethane foams and derived nanocomposite foams have been noted for low weight, flexibility, facile synthesis, and valuable electrical conductivity and dielectric properties [86]. However, EMI shielding competency of polyurethane nanocomposite foams seemed to be reliant upon polymer backbone, nanoadditive type/content, dispersion, matrix-nanofiller links, and manufacturing route applied [87].

As per literature reports so far, nanocarbons such as graphene, graphene derivatives, carbon nanotubes, carbon nanofibers, carbon black, etc., have been recurrently applied as nanoreinforcements for polyurethane foams [88]. Li et al. [89] designed polyurethane filled foams with carbon nanotube nanofillers using latex approach. These polyurethane/carbon nanotube sponges exhibited fairly high electrical conductivity (>360 Sm^{-1}) and radiation shielding efficiency (~25 dB). The radiation shielding performance was suggested to be because of the formation of percolation network supporting electron transfer and radiation shielding performance of the hybrids. Jiang et al. [90] used reduced graphene oxide as nanofiller and CO_2 foaming process for polyurethane foams. These nanomaterials revealed lower conductivity (2.5×10^{-1}) than carbon nanotube filled foams, however had reasonable EMI shielding effectiveness (22 dB). In this concern, Gavgani et al. [91] reported on a outperforming polyurethane and reduced graphene oxide derived foams by adding foaming agents (Voranol/tin). These nanocomposite foams had electrical conductivity of ~4 Sm^{-1} and enormously high radiation shielding efficiency (>253 dB). Such performance of polyurethane/reduced graphene oxide foams seemed to be because of the effectiveness of synthesis method used for developing hierarchical and inerfacially connected three dimensional porous nanostructures. Oraby et al. [92] manufactured polyurethane/iron(II,III) oxide/reduced graphene oxide based nanocomposite foams using facile solution sonication and curing routes. These hybrid sponges were investigated for microstructural, mechanical, and radiation absorption properties. Accordingly, **Figure 7A** a–c show transmission electron microscopy micrographs of iron(II,III) oxide/iron(III) oxide nanoparticles, pristine reduced graphene oxide nanosheet, and iron(II,III) oxide/reduced graphene oxide hybrid nanoparticles, respectively.

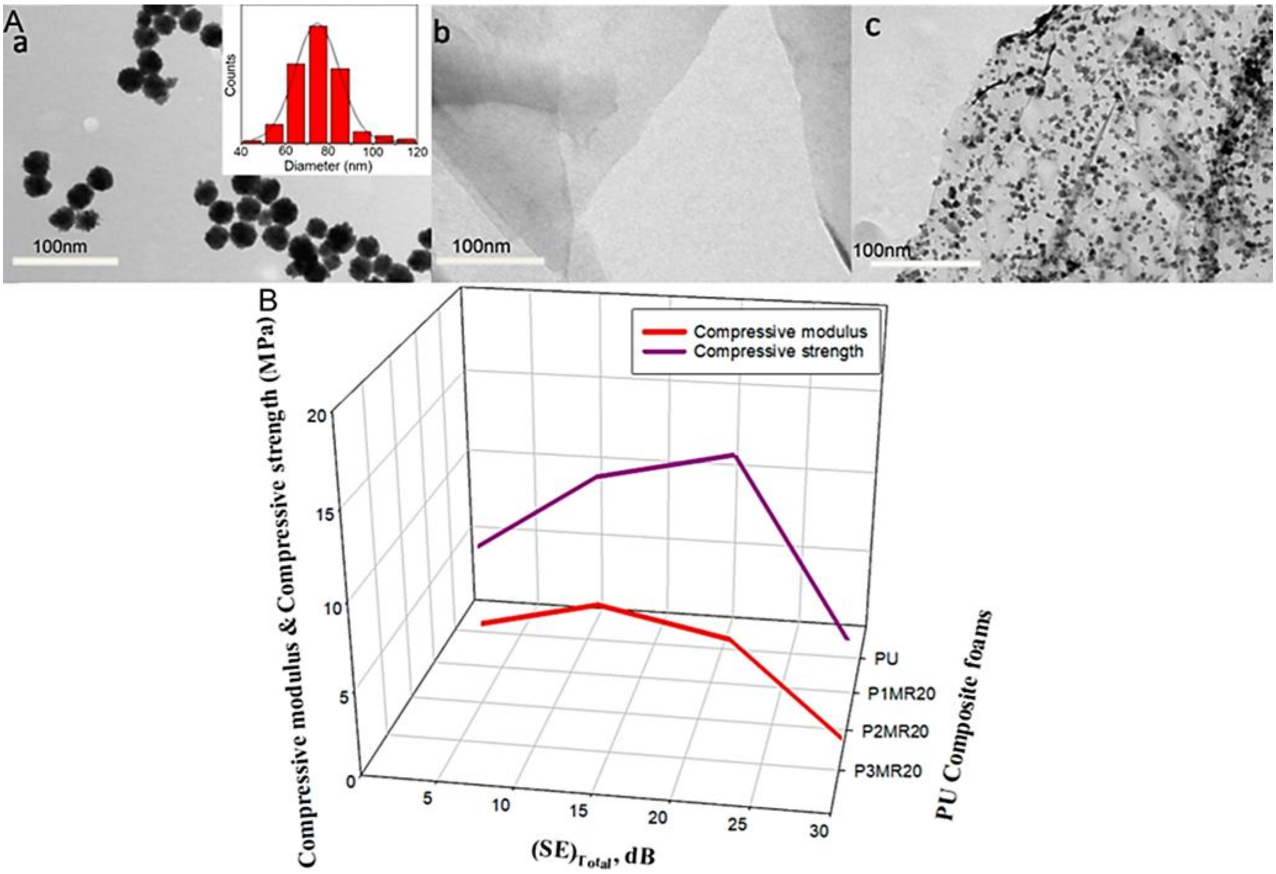

Figure 7. (**A**) Transmission electron microscopy images of: (**a**) iron(II,III) oxide/iron(III) oxide (Fe_3O_4/Fe_2O_3) nanoparticles, inset: particle size distributions; (**b**) reduced graphene oxide (rGO) nanosheets; (**c**) iron(II,III) oxide/reduced graphene oxide (Fe_3O_4/rGO) hybrids; (**B**) mechanical properties vs. shielding effectiveness (SE) and polyurethane foam with filler loading [92]. Reproduced with permission from MDPI.

The iron(II,III) oxide/reduced graphene oxide hybrid had fine dispersion of tiny nanoparticle (~70 nm) over thin transparent graphene surface. In addition, **Figure 7B** illustrates the effect of increasing iron(II,III) oxide/reduced graphene oxide nanofiller contents as well as compression strength and modulus on shielding effectiveness of the nanocomposite foams. As per results, adding nanoparticle contents (up to 35%) caused notable shielding effectiveness of ~33 dB. This effect was attributed to the formation of continuous percolation network of reduced graphene oxide and iron nanoparticles in the polyurethane foams, so leading to valuable electrical conductivity and radiation absorption properties. Similarly, reasonably high compressive strength and modulus of around 15.6 and 5.3 MPa, respectively, were attained for the hybrid foams. Superior mechanical properties of polyurethane foams reinforced with iron(II,III) oxide/reduced graphene oxide hybrid nanoparticles were visibly linked to the integrity of three dimensional nanoarchitectures due to mutual interfacial compatibility.

Into the bargain, polyurethane foams and polyurethane nanocomposite foams (whether open cell or close cell) have been employed in space sector owing to their capabilities towards efficiently attenuating fast moving neutron, γ-rays, and electromagnetic interfering radiations [93]. In addition, these foams have low densities and nonflammability properties to be employed as promising radiation shields for electronics, energy devices, communication equipment, and defense system of aerospace industry [94]. Hence, using high performance polyurethane foam based radiation shields may open invaluable ways for deployments in advanced future space architectures.

For a better literature analysis, **Table 1** shows some significant polyurethane nanocomposite foams applied for electromagnetic interference shielding purposes.

Table 1. Electromagnetic interference shielding (EMI) shielding effectiveness of polyurethane nanocomposite foams.

Foam matrix	Nanofiller	Fabrication	Electrical conductivity (Scm^{-1})	EMI shielding effectiveness (dB)	Ref
Waterborne polyurethane	Carbon nanotube	Latex technology	362	25 dB	[89]
Polyurethane	Reduced graphene oxide	Supercritical CO_2 foaming	2.5×10^{-1}	3.17 vol.%; 22 dB	[90]
Polyurethane	Reduced graphene oxide	Tin catalyst and Voranol foaming agent	4.0	253 dB	[91]
Polyurethane	Fe_3O_4 functional reduced graphene oxide	Sonication; curing	-	25 wt.%; 23 dB	[92]
Polyurethane	Graphene oxide	Solution, heating, casting	3.0	20 wt.%; 17-24 dB	[95]
Polyurethane/polydopamine	Graphene	Dip coating; ultrasonic; compression heating	-	~ 60 dB	[96]
Polyurethane	Graphene nanoplatelets	Supercritical CO_2 foaming	-	1 wt.%; 16-18 dB	[97]
Polyurethane	Graphene	Catalyst; Foaming agent	-	Acoustic performance	[98]

4.2. Shape memory applications

Shape memory (stimuli active) polymers own intrinsic ability to change their shape reversibly, when exposed to light, heat, electricity, or any environmental effect [99]. Innumerable shape memory polymers (thermoplastics, thermosets, rubbers, etc.) have been reported in the literature to date [100]. In this concern, polyurethanes have been studied for proficient shape reversibility behaviour [101]. Accordingly, stimuli responsive polyurethane may display one-/two-/or multi-way shape changing phenomenon [102]. As per literature, uses of shape memory polyurethanes can be seen in smart coatings, textiles, and medical appliances [103]. In nanocomposite form, polyurethanes filled with carbon nanoparticles have been investigated for shape memory effects [104]. Mostly studies reported on the thermoresponsive stimuli responsive effects of polyurethane/nanocarbon nanocomposites [105]. Consequently, these smart polyurethane hybrids revealed notable potential for engineering materials, electronics, defense, and medical areas [106]. For example, graphene has been used as an efficient nanofiller to support the stimuli sensitive behavior of polyurethanes [107]. Zarghami Dehaghani et al. [108] used solution condensation method to form polyurethane from poly(tetramethylene ether) glycol, α,ω-dihydroxy(ethylene-butylene adipate), 1,4-butanediol, and methylene diphenyl diisocyanate. Adding

0.25 wt.% graphene resulted in >92 % enhancement in thermos responsive shape memory effects. Wu et al. [109] filled carbon nanotube in a commercially available thermoplastic polyurethane using solution method. These nanocomposites depicted water sensitive shape recovery in ~120 s. Similarly, few other reports available for nanocarbon filled shape memory polyurethanes [110].

As per literature reports, polyurethane foam materials have stimuli sensitivity towards photo, thermal, current, pH, and water effects [111]. An earlier effort by Singhal et al. [112] mentioned the formation of polyurethane via condensation of 2,2',2"-nitrilotriethanol, N,N,N',N'-tetrakis(2-hydroxypropyl)ethylenediamine, and 1,6-diisocyanatohexane. Later, foaming agent method was applied to form polyurethane foams having glass transition temperature up to ~50–70 ºC. Moreover, thermomechanical shape retrieval of 97–98 % was attained. Moreover, In an earlier attempt, Kang et al. [113] also applied blowing agent technique to form polyurethane foams of polypropylene glycol and 2,4/2,6-toluene diisocyanate with carbon nanotube additives. These nanocomposite foams were tested for thermomechanical shape memory effects. According to results, adding 5 wt.% carbon nanotubes in polyurethan foam caused up to 85% shape recovery properties. Later, Kim et al. [114] preferred microwave heating technique to form stimuli responsive polyurethane/carbon nanotube foams. These thermoresponsive spongy nanomaterials revealed shape fixity and shape recovery ratio of ~95% and 84%, respectively. Kumar et al. [115] performed pressure sensitivity studies on shape memory polyurethane foams. In this regard, **Figure 8A** presents schematic of probable volume changes in shape fixity/recovery of shape memory polyurethane foams. Such changes usually occur around glass transition temperature of the polymer and external pressure was applied in this study. **Figure 8B** shows Tekscan F scan pressure system used to analyze the effect of applied pressure (male heel) on the shape memory polyurethane foams.

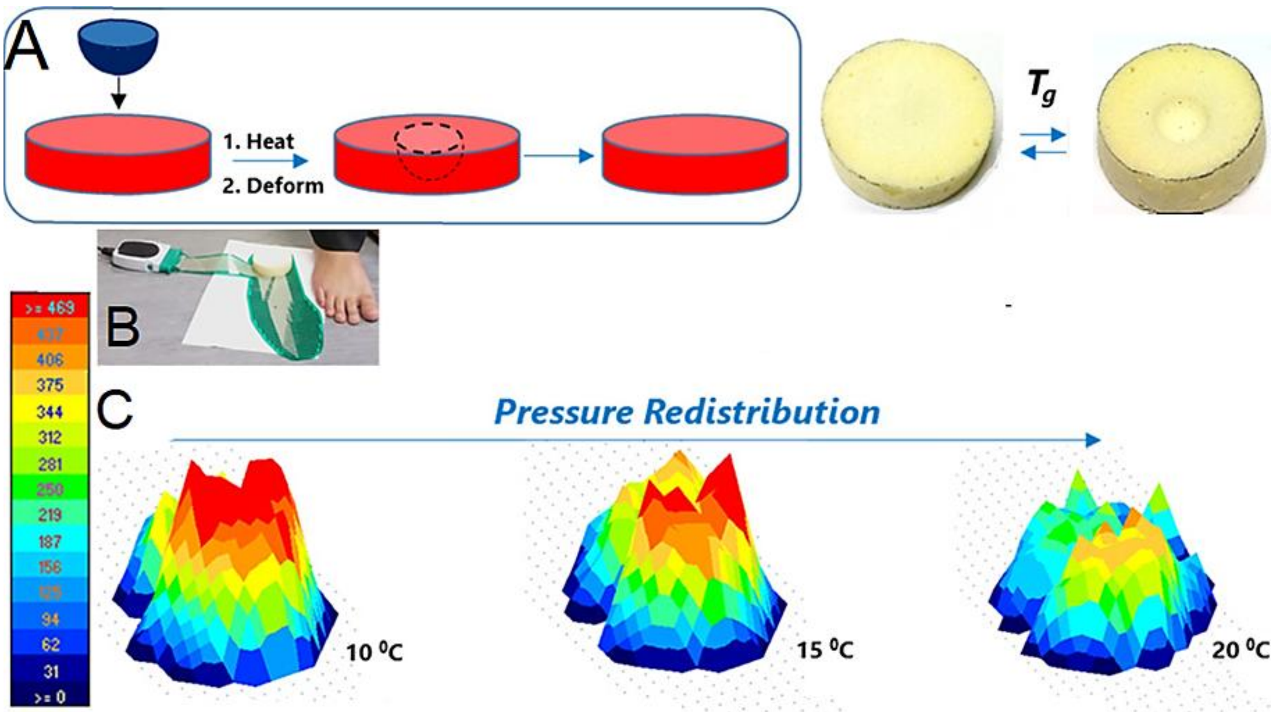

Figure 8. (**A**) Schematic of volume changes in shape fixity and shape recovery processes of shape memory polyurethane at glass transition temperature (T_g) with strain; (**B**) Tekscan F scan pressure analysis using male heel; (**C**) areal pressure distribution of shape memory polyurethane foams under varying surface temperatures and applied force [115]. Reproduced with permission from ACS.

Consequently, **Figure 8C** depicts areal pressure distribution under variable surface temperatures for shape memory polyurethane foam (static force). It was observed that increasing temperature up to 20 °C effectively distributed the applied force and had low modulus due to polymer backbone softening. Contrarily, lower temperatures (10–15 °C) did not efficiently distribute the pressure (concentrated red pressure peaks in **Figure 8C**) due to rigidity of polyurethane foam. It can be suggested that temperature changes along with the applied pressure play important role in shape memory behavior of polyurethane foams.

4.3. In biomedical sector

Polyurethanes have been noted as significant macromolecules for biomedical purposes [116]. In this concern, polyurethanes have countless valuable attributes including optimum physiological features, biodegradability, biocompatibility, prolonged in vivo stability, nontoxicity, and so on [117]. Looking at the medical applications of polyurethanes, a myriad of uses has been reported for tissue scaffolds, bioimplants, drug delivery, coatings, wound healing, smart devices, etc. [118–120].

Polyurethane foams have been designed and tested for *in vitro* and *in vivo* conditions for biomedical uses [121]. Consequently, these spongy materials depicted fine biocompatibility and long term biosustainability during desirable applications in living systems [122]. Among earliest attempts on biocompatible polyurethane foams, Guelcher et al. [123] performed condensation of poly(ε-caprolactone-*co*-glycolide)triol, lysine methyl ester diisocyanate, and tertiary amine. The resulting polyurethane foams were applied as injectable tissue scaffolds [124]. Later, Schreader et al. [125] explored polyurethane foams reinforced with hydroxyapatite nanoparticles for biocompatibility and bone tissue engineering. Furthermore, an olden attempt by Zawadza et al. [126] disclosed the use of electrophoretic deposition to coat polyurethane foam with carbon nanotube nanofiller. The resulting polyurethane/carbon nanotube hybrid foams were tested for bone tissue engineering. In this concern, growth, compatibility, noncytoxicity, and hydroxyapatite growth have been studies for the nanocomposite foams. Besides, Shin et al. [127] formed polyurethane nanocomposite foams with graphene and graphene oxide and studied for skeletal tissue rejuvenation due to biomimetic effects. These polyurethane/graphene nanocomposite foams had minimum cytotoxicity and optimum porosity (~300 μm), which were suitable skeletal cell growth.

Hence, both the polyurethane/carbon nanotube and polyurethane/graphene hybrid sponges have been studied for biocompatibility/non cytotoxicity effects towards biomimetic injectable scaffolds or hydroxyapatite growth for bone or skeletal tissue engineering. Future studies must focus on more design combination, long term *in vivo* stability, and other biomedical uses like drug delivery, bioimaging, etc.

5. Conclusive remarks and future opportunities

In summary, polyurethane, being a multiuse polymer, has been studied for variety of physical and practical probabilities. Among well practiced forms of polyurethanes, spongy materials have been manufactured for strategic features and applications. In the form of foam materials, polyurethanes own specific cell sizes, distribution, and open/close structures, so contributing to valuable characteristics. As compared to pristine polyurethanes, development of nanocomposite foams using valued nanocarbons, graphene and carbon nanotube, revealed notable surface area, cellular nanostructures, nanoparticle dispersion, electron and heat transportation, flexibility retaining integrity, barrier, noncytoxicity, biocompatibility, and other beneficial attributes towards high end uses. According to research efforts to date, application areas discovered for polyurethane nanocomposite foams include electromagnetic radiation shielding, stimuli responsiveness, and medical related uses (**Figure 9**). In polyurethane/graphene nanocomposite foams, polyurethane/carbon nanotube nanocomposite foams, and all the applied fields, adding nanoparticles type, contents, scattering, and interfacial specifications directly influence the materials properties and applied contours. Moreover, feasibility and effectiveness of processing techniques may affect the implication of ultimate spongy architecture.

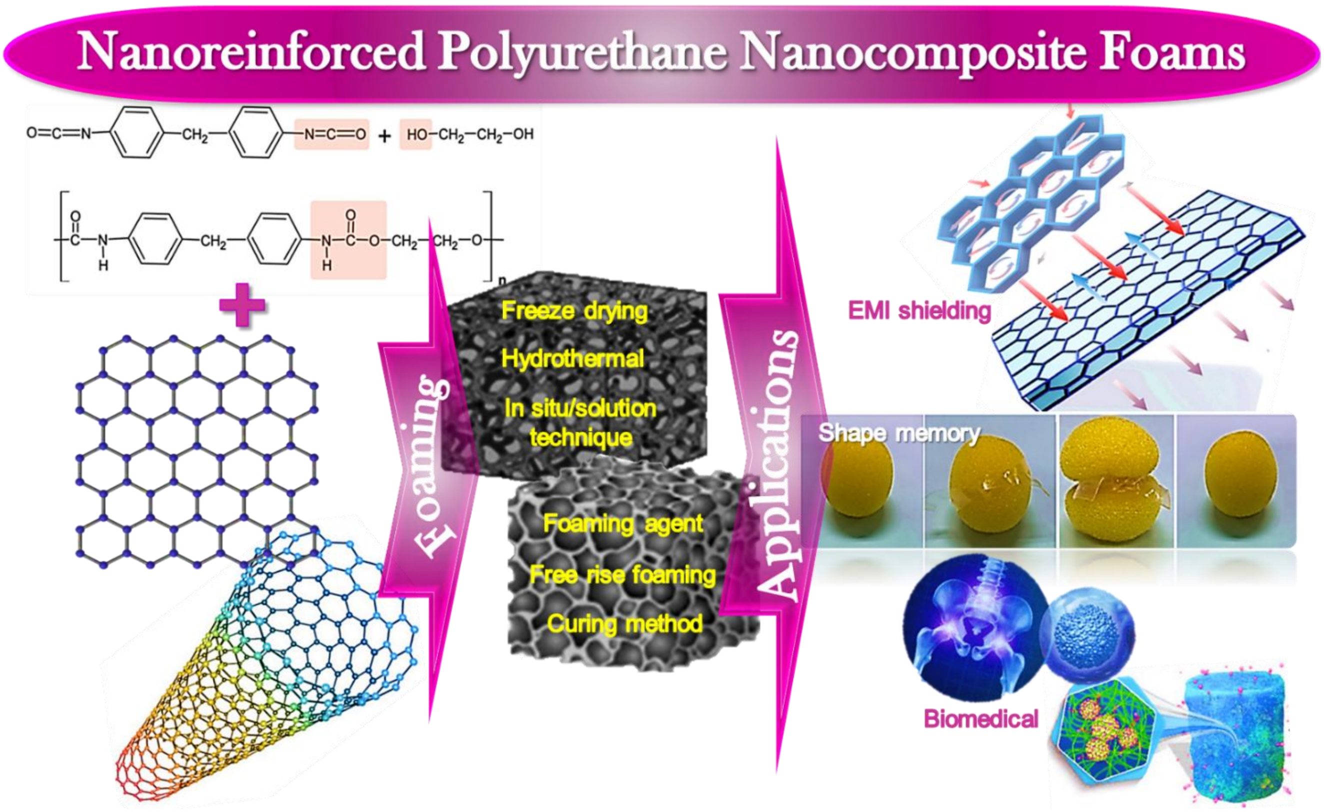

Figure 9. Prospects of multifunctional polyurethane foams.

Looking at the valuable properties of polyurethane nanocomposite foams, we can suggest several future applications of these spongy materials. Especially due to thermal conductivity properties, polyurethane hybrid foams can be used to substitute commercial panels and interiors in aerospace and automotive vehicle structures.

Similarly, such materials can be practiced for advanced construction and civil engineering utilizations. Another side of these nanocarbon filled hybrid sponges not discovered yet seemed to be the smart wearable devices and e-electronics. In addition to radiation absorption, these nanocomposite sponges can be used for encounter sound and acoustic effects in relevant fields. Due to limited research so far on medical sides, comprehensive efforts may reveal application of polyurethane nanocomposite foams in smart drug/gene delivery and smart tissues and artificial muscles.

Concisely, further applied breakthroughs of polyurethane hybrid aerogels can be protracted by explorations of key mechanisms for ultimate cellular structure and interfacial relationships. In addition, scalable manufacturing of polyurethane nanocomposite foams by achieving global sustainability and environmental demands seem indispensable for future commercial modules in high tech industries, from energy to medical.

Conflict of interest: The authors declare no conflict of interest.

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