REVIEW

Polyurethane (PU) based multifunctional materials: Emerging paradigm for functional textiles, smart, and biomedical applications

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Abstract
The advances in polymer chemistry research have revolutionized the field of smart materials and biomedical. Polyurethanes (PUs) are a versatile polymer class with diverse structure, morphology, and behavior under various conditions deemed suitable for many functional and intelligent responses. Owing to their biocompatibility, biodegradability, excellent mechanical strength, tailorable chemical, and physical forms, PU has drawn significant research attention in recent times for a wide range of applications. Herein, this review paper outlines PU's recent advances in specific applications encompassing functional textiles, intelligent functionality and medical usage. This article contains a comprehensive review of recent developments and research works concerning PU's direct involvement as coatings, 3D objects, or composite parts to add novel purpose to the textile substrates, smart objects, or medical applications. Commencing with PU's application for the waterproof breathable textiles, the review further explores recent research where PU was incorporated as a phase change material and protective clothing. This review further delves innovative functionalities and responsiveness of the polymer triggered by various stimuli. It ends with an inclusive review of PU's different forms of application concerning medical devices and activities. Finally, perspectives on future challenges and new research opportunities are also presented and discussed.

KEYWORDS
biomedical applications, functionalization of polymers, polyurethane, stimuli-sensitive polymers, textiles

1 | INTRODUCTION

PU is a copolymer of urethane moiety that possesses more complex chemical structures in which typically three types of monomers are added: a diisocyanate, macroglycol (an oligomeric macro monomer), and chain extenders.[1–4] In this segmented block copolymers, hard segment-enriched domains, which are composed mainly of the diisocyanate and the chain extender, are dispersed in a matrix of soft segments (macroglycol moieties).[5–9] Wurtz synthesized the first urethane in 1849, but Professor Otto Bayer pioneered the way to successfully synthesize PU as a byproduct of the reaction between a polyester diol and a diisocyanate in 1937 in Germany. From then until today, PU polymers have increasingly been used in many ways, including a coating, elastomer,

foam (flexible or rigid), adhesive, sealant, paint, varnish, and many more due to their versatility, excellent mechanical and chemical resistance properties, comfortability, cost-effectiveness, and environment-friendly nature.[4,10–12] As a result of its versatile properties, it has touched almost every area of material engineering, from shoes to aeronautical coatings.[13,14] However, the notable end-users of PU are sports, medicine, medical devices, biomedical, leathers, footwear, rubbers, films, biomimetic materials, mattresses, building and construction, electronics and appliances, packaging, upholstered furniture as flexible foam, and metal adsorbent. This diversity of applications is due to the tailorable chemistry (low glass transition temperature) and the flexibility of PU-based polymers even at low temperatures.[1,6,7,15–22]

According to the survey of Covestro LLC, a manufacturer of premium, high-tech polymers based in Leverkusen, Germany, in 2019, the global demand for PU was an estimated 18.4 million tons, and it is forecasted to increase 22.5 million tons by 2024. Another report shows that the worldwide consumption of PU was estimated at 60.5 billion USD in 2017, and it was predicted to be over 79 million USD by 2021.[1,3,23]

As flexible substrates, PU has brought a significant breakthrough in functional textile fields for their cost-effective nature, bulk production capabilities, and interesting physico-chemical properties.[24] The PU-coated fabrics possess exceptional electrical and adhesive properties, greater resistance to abrasion, wear, weather, and high resistance to chemicals and water.[25–28] Additionally, the absence of plasticizers makes PU coatings durable for washing and dry cleaning.[29] PU coatings on nylon fabric provide high moisture permeability and water repellency properties to the base fabric. These fabrics are the best substitute for synthetic leather cloths used in foul weather garments for civilians and military.[30] With the incorporation of PU coating, the highly hydrophobic textile surfaces show anti-sticking, anti-contamination and self-cleaning characteristics.[29]

Finally, the application of PU in textile and clothing, especially in technical textiles, is also burgeoning for having their exceptionally balanced and tailorable properties besides their conventional application as elastane/lycra in clothing.[31]

Additionally, PUs have played a dominant role in a myriad of unique medical devices with excellent performance in biomedical applications.[32] Different polymeric materials can be used, either as coating or as plain medical devices, such as polyvinylidene fluoride (PVDF), polyethylene (PE), polypropylene (PP), poly (methyl methacrylate) (PMMA) and silicone.[32] Interestingly, PUs offer one of the most diverse classes of material with unique elasticity, metal-like toughness, and durability.[33–35] Therefore, they are vastly used in medical devices,

coatings, elastomers, adhesives, sealants, and automotive interiors.[33] Additionally, thermoplastic-based PUs can be blended with other polymers, reshaped and modified for targeted applications.[36–38] The outstanding biocompatibility and antimicrobial efficacy, excellent mechanical and adhesion properties as well as low cost have made PU the most promising candidate. Consequently, it has drawn the great attention of researchers to utilize them in a wide range of biomedical applications, including catheters,[39–41] endotracheal tubes,[42] PICC lines,[43] and ureteral stents.[44,45]

Furthermore, PU as a shape memory polymer (SMP) in 4DP is gaining interest in the past few years, as the applications and printing techniques for these materials are changing rapidly.[46,47] These 4D printed PU structures respond to external stimuli, allowing them to change some of their properties. This behavior represents a firm ground for revolutionary advancements in the field of robotics, biomedicine, and industry.[48] Moreover, SMPU has emerged in additive manufacturing (3DP and 4DP) as a versatile technology platform for future advanced manufacturing systems.[49,50] Their applications have expanded dramatically from visual prototypes[51] to tissue engineering,[52,53] electronic devices,[54,55] and high-performance metamaterials.[56–58] The advantages of the PU over conventional polymers are summarized in Table 1.

While numerous research works have been published on using PU in functional and medical textiles, the lack of review in this specific area is the motivation of this present work. Table 2 demonstrates the gap in the literature based on the recently published review articles that are closely aligned to the proposed topic. Numerous review articles on the different paradigms are also found based on functional textiles[4,18,70–72] and protective and smart applications.[17,20,22,23,30,73] However, critical reviews addressing the recent progress of PU-based materials/coatings in functional textiles, smart materials with responsive behaviors and protective materials are yet to be amassed in a single frame. Furthermore, this paper comprehensively reviews the striking and interesting biomedical applications and PU based medical devices. This paper is also likely to feature the state-of-the-art 4DP of PU with reversible shape memory features. Afterward, the future challenges of PU based materials are outlined. The emerging prospects of PU as a key material in next-generation functional applications are also envisioned.

2 | PU COATING IN WATERPROOF BREATHABLE FABRIC

Breathability is a material's ability to transmit moisture vapor or perspiration through it. It is one of the

TABLE 1 Advantages of PU over conventional polymer

Application area	Conventional polymer used	Limitations of conventional polymer	Advantages of PU	Reference
WBF	Animal fat, wax, vegetable oils, silicone, PE or polyvinyl chloride (PVC) polymers	Stiff and uncomfortable	Impart flexibility, adhesion, and abrasion resistance	59
PCMs	Inorganic hydrated salts, paraffin waxes, fatty acids, fatty alcohols, and eutectics compounds	Loss of strength, flammability, ineffective phase change, leakage	Thermal insulation, recyclability, CFC-free, high mechanical and chemical stability, heat absorption capacity, Solid–solid PCMs	60–62
Chemical protective	PVC, PVDC, PTFE, and PE	PVC, PVDC, PTFE are expensive & have very poor heat stability. PE is Flammable and sensitive to stress cracking	Exert decent strength, hydrophobicity, breathability, enhanced visual properties, excellent abrasion resistance, chemical resistance, and low-temperature flexibility	63, 64
Ballistic Protection	Fibrous materials (soft body armor), metal plates, ceramic or plastic materials (hard body armor)	Bulky and limits the wearer's mobility	Improved mechanical properties, optical properties, and lightweight	65,66
Smart application	Alloys and some conventional polymers (polynorbornene, trans-polyisoprene, styrene-butadiene copolymer, crystalline polyethylene, ethylene-vinyl acetate copolymer etc.)	Adjustable mechanical properties of limited range, Poor fatigue property, Nonlinearity of actuation force	Biocompatibility and tunable physicochemical properties. High compressibility, and excellent shape changing ability upon external stimuli	49, 50
Medical application	PVC, Polyester, Polyethylene or Polypropylene	Contains reproductive toxin, flammable	Excellent biocompatibility, Oxygen permeability, easier process ability, barrier properties, exceptional mechanical strength with flexibility, and abrasion resistance	67–69

fundamental performance criteria of modern textile industries as it explains the comfort level of the material and the ability of our body to maintain its core temperature under various conditions and work rates. To be breathable, clothing must be designed to maintain a balanced situation spontaneously under different environmental conditions and body movements without attenuating the evaporation of humidity caused by perspiration. Controlled movement of water vapor and liquid water from the skin's surface to the atmosphere through the fabric helps to prevent perspiration leftover next to the skin. All of these desired phenomena appreciably go under the technical term moisture management.[80–84]

Conventionally, waterproof fabrics are coated with animal fat, wax, vegetable oils, or PVC polymers, but coated fabrics are uncomfortable to wear because of their stiffness and unforthcoming behavior towards escaping perspiration vapor. On the contrary, although the water-repellent fabric is more comfortable to wear, it has transient water-resistant properties. The specialty of WBF is that it should be simultaneously waterproof (impedes water droplets getting into the fabric) and breathable to allow water vapor to escape from the skin through the openings between fibers and yarns.[59]

As worthwhile soft coatings for textiles, the popularity of PU coated WBF has grown due to their perceived qualities and physical properties, such as flexibility, adhesion, and abrasion resistance, which are superior to those of other polymeric materials.[85] A few popular types of PUs have been discussed below.

2.1 | Waterborne PU coating for WBF

The use of waterborne PU (WBPU) coating in WBF has drawn significant research attention because of the

TABLE 2 Recently published review articles on the application of PU

Types of PU	Focusing areas	Reference
PU	Application areas in medical science, automobiles, coatings, adhesives, sealants, paints, textiles, marine industry, and wood composites	4
PU	Biomedical application	7
PU and PU nanocomposites	Application in medicine	19
PU foam	Metal adsorbent	20
PU-based material	Immobilization of enzymes and cells	74
Waterborne PU	Synthesis and application in water-based Ink	75
PU and PU composites	Recycling and recovery	21
PU	Removal of organic dyes from textile wastewater	72
PU composite foams	High-performance applications	76
PU	Structural and infrastructural engineering applications	77
Porous PU scaffolds	Fabrication of scaffold for tissue engineering	78
Shape Memory PU (SMPU)	Shielding of electromagnetic interference, medical bandage development, bone tissue engineering, self-healing, implants development, 4D printing (4DP)	79, 48

growing concern of diminishing potential environmental pollution by reducing low volatile organic compounds generated from the solvent-borne synthesis technique. In addition, the stable nature of the materials, ease of use, and similar performance to solvent-based systems have made it a straightforward alternative to solvent-borne PUs in the field of WBF.[85–87] PU coated WBF requires a balance between water vapor permeability and water resistance, which is achieved by tailoring hydrophilic and hydrophobic segments. Traditionally, poly (ethylene glycol) (PEG) segment is utilized as a hydrophilic soft segment to endow high WVP during PU synthesis due to its hydrophilic nature. This hydrophilic PU increases water vapor penetration and decreases water resistance.[85] Yen et al. examined the effect of the composition of polycaprolactone–poly (ethylene glycol)–polycaprolactone (PCL–PEG–PCL) triblock copolydiol soft-segment on the structure and physical properties of WBPU and showed that higher molecular weight of PEG in soft segment improves the WVP of WBPU-coated nylon fabrics due to its higher hydrophilicity.[88]

WBPUs, prepared from poly propylene glycol (PPG) 2050 (copolymer of ethylene oxide and propylene oxide with —OH end groups) and hydrophobic poly (tetramethylene ether glycol) (PTMEG) achieved a balance of WVP (910–990 g/m^2 × 24 h) and waterproof properties (>10,000 mm H$_2$O) than those prepared from PEG/PPG/PTMEG-based WBPU when used as a textile coating.[89] Apart from hydrophilic and hydrophobic components, several other factors affect the fabric's breathability, such

as type of PU dispersions, coatings, additives, and processing conditions.[90]

In synthesizing waterborne siloxane-containing PU from PTMG and PEG, polysiloxane can be used as a modifier due to its hydrophobic characteristic and high vapor diffusion rate. In the presence of hard segment, hydrophilic chain extender, and neutralization agent, the incorporation of 10% α,ω-aminopropyl polydimethylsiloxane as mixing soft segments with PU results in the highest moisture permeability (2130.15 g m^2 × 24 h) and water resistance (30.0 KPa).[91,92] WBPU coating on electrospun nanoweb membrane laminated taffeta and taslan base fabric improve abrasion resistance and durability of the nanoweb membrane compared to commercial Gore-Tex XCR in terms of waterproof and breathable properties. Nevertheless, the structural properties of the fabric also directly affect the breathability and waterproofing while laminating and coating.[93]

2.2 | Shape memory PU for WBF

Shape memory PU (SMPU) has sensory abilities and can respond to external stimuli (heat, light, electric field, magnetic field, chemical, moisture, pH, etc.) in a predetermined shape. Recently, SMPU has been extensively used for developing WBF due to its superior performance and versatility.[94] Coated/laminated textiles with SMPUs provide high WVP at higher temperatures and low WVP at lower temperatures to uphold user comfort. However, the efficacy of shape memory behaviors of PU largely
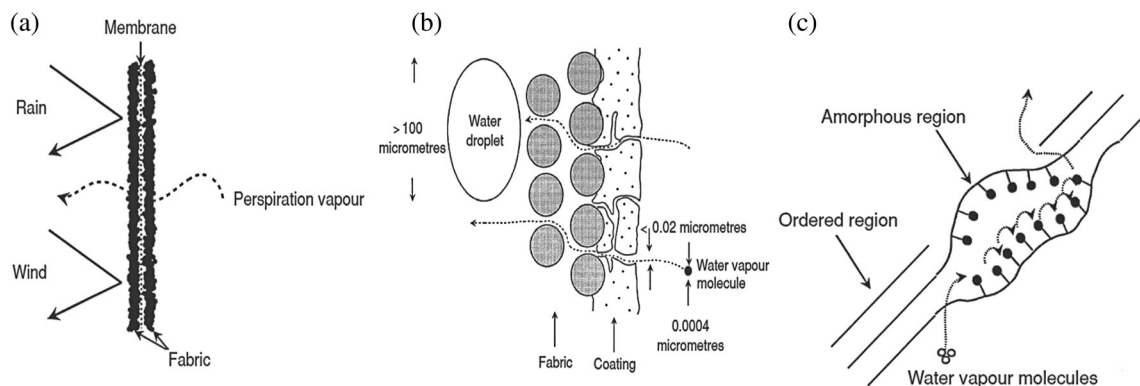
FIGURE 1 Moisture vapor transmission through (a) micro porous membrane (b) micro porous coating, and (c) mechanism of hydrophilic polymer. *Source*: Reused with permission from Reference [101], © Elsevier, 2000.

depends on soft segment crystallinity and the formation of the physical cross-link structure of the hard segments of PU.[95,96] In other words, fluctuation of soft segment's molecular weight, soft-hard segments mole ratio, and the polymerization process control the shape memory behavior of PU.[97,98]

Polyester woven fabric can be turned into water vapor permeable fabrics after coating with SMPU synthesized from poly (tetramethylene glycol), 4,4'-methylene bis (phenyl isocyanate), and 1,4-butanediol. WVP of PU-coated fabrics decreases drastically with the increased concentration of coating solution, whereas only a slight change occurs with the control of PU hard-segment content.[16] Ding et al. investigated the significance of crystal melting of soft segments in case of a dense SMPU film. Crystal melting of SMPU takes place with the increase of temperature from 10 to 50°C and increases amorphous area, resulting in a significant rise in WVP of waterproof fabric.[99] Investigation shows that the presence of ester groups in the backbone of SMPU decreases the WVP. Inversely, the presence of PEG increases WVP due to the increasing hydrophilicity of the SMPU.[100]

2.3 | Hydrophilic PU membrane for WBF

Hydrophilic membranes, a thin film of chemically modified polyester or PU, can be synthesized by incorporating poly (ethylene oxide) in the polymer structure. The inclusion of poly (ethylene oxide) in polymer helps to constitute the hydrophilic part of the membrane by forming an amorphous region in the core polymer system (Figure 1c). This amorphous structure acts as the intermolecular pores to allow water vapor to pass through, whereas the solid nature of the membrane precludes the penetration of liquid water. The only difference between the microporous and hydrophilic material is that water

vapor passes through the permanent air-permeable structure (Figure 1a,b). Contrarily, the vapor is transmitted through adsorption-diffusion and desorption mechanisms in the hydrophilic materials.[101] Improved adhesion on textile substrates and lower cost are other advantages of hydrophilic PU coatings over microporous PU.[102]

The commencement of hydrophilic PU coating minimizes the usage of synthetic leather cloths in unwholesome weather. Nylon fabric imparts distinctive water-repellant moisture-permeable properties while coated with hydrophilic PU, synthesized from ε-caprolactam-4,4'-diphenylmethane diisocyanate and hydrophobic polyols, and finally de-blocked with PEG of different molecular weights at 130°C under nitrogen atmosphere.[103] Additionally, Tsai et al. launched cationic aqueous PU (CAPU) coatings incorporating 4,4-methylene bis (isocyantocyclohexane), N-methyl diethyol amine with various polydiols of different molecular weights (Mn = 1000, 2000, and 3000 g/mole) like PEG, PPG, and PTMG for ether type polyols and PCL for ester-type polyol. Here, ether-based CAPU showed a higher surface tension value, wash durability (80%), contact angle and WVP with a sequence of PEG-PU > PTMG-PU > PPG-PU > PCL-PU. At the same time, the waterproofing followed the order: PTMG-PU > PCL-PU > PPG-PU > PEG-PU and the value rose with the increase of molecular weight of the soft segment.[104] Durable water-repellent textiles can be processed using an ambient temperature and UV curable polydimethyl siloxane-containing PU system.[105,106] Moreover, PU coated waterproof cotton fabric represents improved breathability while micro-cracked and dipped in suitable solvent (ethanol, DMF, acetone) at different ratios and times.[107]

2.4 | Electrospun PU coating for WBF

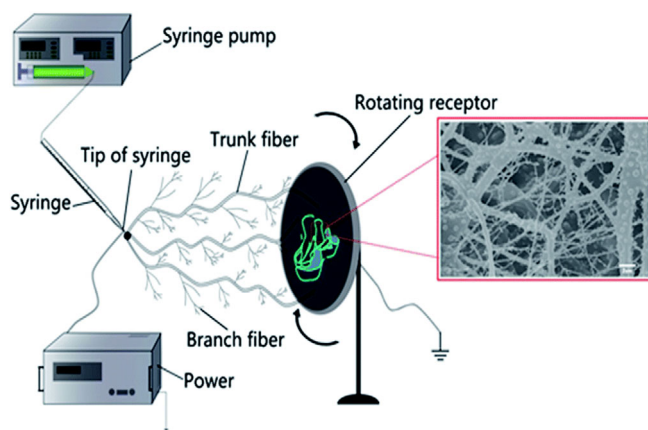Electrospinning, a high voltage-dependent novel fiber-forming technique (Figure 2) of developing waterproof

FIGURE 2 Electrospinning technique. *Source*: Reused with permission from Reference [109], © the Royal Society of chemistry, 2017 [Color figure can be viewed at wileyonlinelibrary.com]

breathable materials, provides an ultrathin interconnected membrane-like web of extremely fine fibers (10–500 nm in diameter) with a very small pore size.[108]

Electrospun PU web based multifunctional WBF provides superior water-proofing and breathable properties to PU resin-coated fabrics regarding air permeability, vapor transmission, and thermal insulation properties. For instance, fabric with solvent-electrospun (using *N*,*N*-dimethylacetamide as solvent) PU web shows greater comfort performance than resin-coated fabrics. However, the water resistance value does not reach that of resin-coated fabrics.[110] Han et al. investigated a facile coaxial electrospinning technique to electrospun nanofibers having PU core and polycarbonate shell materials. They established its feasibility by measuring the water vapor transmission rate of resulting nonwoven mats.[111] More recent relevant research activities concerning PU coating applied on WBFs have been enlisted in Table 3.

3 | APPLICATION OF PU AS PHASE CHANGE MATERIALS

In recent times, applications of thermal energy storage and exchanging materials are gaining popularity and acting as a crucial aspect in energy storage (by thermochemical, sensible and latent heat storage process) due to the demand for developed energy conservation and improved thermal comfort. Among the notable application areas of PCMs, application in solar energy utilization,[116] waste heat recovery,[117] building air conditioning,[118] electric energy storage,[119] temperature-control of greenhouses,[120–122] telecommunications and microprocessor equipment,[123] kitchen utensils,[124] biomedical and biological-carrying systems,[125] food transport and storage

containers,[126] and insulating clothing and textiles for thermal comfort are worth mentioning.[127] The materials used for latent heat storage systems are known as PCMs. PCMs can absorb or release large amounts of heat during phase transitions between two solid states and/or liquid and solid states. A wide range of PCMs with innumerable melting points are categorized as organic (paraffin, fatty acids, and polyethylene glycol), inorganics (salts, salt hydrates, hydroxides, and metallic), polymeric and eutectic combination of organic and/or inorganic materials.[60–62] The incorporation of PCMs into clothing provides not only thermal comfort to the wearer but also improves thermal behavior of the wearer, protecting them against inclement weather.[128] Currently, application of PU-based PCMs in textiles and clothing sector is one of the most important research topics and a plethora of approaches are practiced worldwide like, deposition of PCMs on PU films, incorporation of PCMs in PU films, PU-PCMs composite, and microencapsulation technique.[129,130] Following sections discuss briefly about PCMs incorporation with PU.

3.1 | PCMs embedded PU films

Imparting composite membranes of PU and PCMs as a coating of basic fabric offers commendable thermal comfort of the composite fabric. Yoo HJ and his team prepared coated polyamide fabric by embedding WBPU-urea emulsions with 30% PCMs (hexadecane, octadecane, and eicosane) through in-situ polymerization process and in presence of thickener and hardener (traps PCM in the coated material).[131] Similarly, Hong W arranged WBPU-PCM from hexamethylenediisocyanate and PEG in presence of chain extender, catalyst, and neutralizing agent. Produced WBPU-PCM can provide high-thermal stability and admirable thermal cycling stability. Environment-friendly application and reversible phase change process have turned it into a great prospect in functional textile. In addition, performance can be modulated by varying reaction temperature, reaction time, PEG relative molecular mass and chain extender content.[132,133] Recently, Ke GZ has launched a blended (PU-PEG) porous membrane and established that PU/PEG membrane with porous structural features provides a suitable transition temperature and high-transition enthalpy.[134]

3.2 | PU foams incorporated with PCMs

PU rigid foams are widely used as the ultimate energy savers for thermal insulation and are highly competitive compared to other insulating materials due to their

TABLE 3 Recent works concerning PU coating of WBF

Materials	Function	Application area	PU category	Special characteristics	Ref
Polytetramethylene Glycol based PU	Provide an ideal condition for thermal insulation and vapor transmitting ability according to the body temperature and humidity	Functional textile	Shape memory PU	Moisture sensitive	112
Ether based cationic aqueous PU	Achieved higher surface tension value and contact angle	Packaging, hydrogels, medicals, biosensors	Hydrophilic PU membrane	High-wash durability	104
Hydrophilic membranes based on PEO-PUs	Provide higher hydrostatic head resistant and higher contact angle of water droplets	Sportswear, medical textiles, weather resistant workwear	Hydrophilic PU membrane	Zero air permeability	113
PU modified with Poly ethylene macromer	Provides improved water vapor permeability	Breathable coated fabric	Waterborne PU	Temperature responsive water vapor permeability	85
WBPU having 70% PEG and 0.5% hardener	Provides good water insolubility, low water contact angle, high water absorption, higher water vapor transmission	Breathable coated fabric	Waterborne PU	Good dimensional stability in water	114
Solvent (N,N-dimethylacetamide)-electrospun PU	Provides better thermal insulation and water resistance	Protective garments	Electrospun PU web	Superior air permeability and moisture transport properties	110
Fluorine-free PU nanofibrous membranes	High water contact angle, robust hydrostatic pressure, desirable water vapor transmission rate, good air permeability	Protective textiles	Electrospun PU	Not available any toxic solvent residues	115

recyclability and chlorofluorocarbon-free nature. In addition to heat absorption capacity of PU, the confined air within the wavelike structure provides a passive insulation property of foam. Furthermore, the main advantages of PUs foam are their lowest thermal conductivity, high mechanical and chemical stability at different temperatures, and the ability to form sandwich structures with various facer materials. The chemistry behind the formation of PU foam is the reaction between polyols and diisocyanates obtained from crude oil in presence of a small amount of blowing agent and water. Here, water reacts with isocyanate groups giving carbamic acids, which spontaneously release CO_2 and generate foam bubbles. On the other hand, the blowing catalyst composition provides the resulting PU foam with improved breathability.[135,136]

Although incorporating PCMs in PU foam is problematic because of the violent foaming reaction, it can engineer functional properties in textile and clothing. Sarier N combined paraffin waxes as PCMs (*n*-hexadecane and *n*-octadecane) at different ratios with PU foam to produce thermal insulators equipped with an improved buffering function against temperature changes.[135] Besides, PU foam incorporated with micro PCMs is considered one kind of innovative heat-insulating material which can frequently store and release heat energy.[136]

3.3 | Solid–solid phase transition PCMs–PU composite

PCMs that go through a solid–solid phase transition with the concomitant absorption and release large amounts of heat are known as solid–solid PCMs (SS-PCMs). At a fixed and well-defined temperature (melting point of soft segment) SS-PCMs can change their structure from one lattice configuration to another (crystalline to semi-crystalline or amorphous). However, by adjusting the chain length of soft segments or the rigidity of the backbone, the transition temperature may be tailored. The most significant advantage of SS-PCMs is their leak-proof nature of the process. Hence, no problems are associated with liquids handling and can be processed without additional storage systems. Besides, there is no visible change in the appearance of the PCM due to solid–solid phase change, merely a slight expansion or contraction of the materials. However, the main drawback of SS-PCMs is that these materials require nucleation to prevent supercooling, which is not desired in SS-PCMs.[137]

Alkan C introduced PUs as SS-PCMs for thermal energy storage using three different diisocyanate molecules (*N,N*-dimethyl formamide, hexamethylenediisocyanate, and isophoronediisocyanate) and PEG at three different molecular weights.[138] PU systems with hyper branched polymers are combined to add multifunctional properties. A newly developed material from novel hyper branched PU copolymer that uses hyper branched polyester as chain extender shows excellent polymeric solid–solid phase change heat storage capacity.[139] Recently, a novel binary shape-stabilized composite PCM (CPCM) was developed using a crosslinked PU copolymer with a solid–solid phase transition to load additional PEG. The developed composite shows excellent thermal stability, good reusability, and large heat storage density. Their dual-phase transition (solid–solid phase transition of the PU matrix and solid–liquid phase transition of free PEG) during heating and cooling process possesses outstanding heat storage density of the CPCMs.[140] Research also showed that PU-based SS-PCMs generated by crosslinking of reduced graphene oxide with hexamethylenediisocyanate biuret and PEG segments show high latent heat, proper phase change temperature, effective light-thermal conversion and remarkable thermal stability and reliability, and hence, pave the way for energy storage and conservation.[141]

3.4 | Microencapsulated PCMs-PU composite

In microencapsulation technique, a tiny particle of active agent or core material is surrounded by coating or shell, where core material can be encapsulated in solution, dispersion or emulsion form.[142] The first attempt to use microencapsulated PCMs (micro PCMs) in PU foams was developed to improve their thermal performance in the 1990s.[143] As renewable and clean energy storage materials, research interest in micro PCMs is accelerated to extend their application in thermal energy fields like heating and air conditioning of buildings, medical products, footwear, automotive interiors, and thermal insulation materials, mostly in thermal adaptable fibers, fabrics, coolants, and coatings.[144–146] Incorporation of PCMs can improve thermal regulation capacity of the PU foam matrix.[147] Enhanced thermal energy storage capacity of PU foam has been investigated with the addition of thermo-regulating microcapsules with polystyrene (PS) or PMMA shell and hence minimizing the addition of extra fillers.[148] Additionally, inorganic supported (silica) PU foam with PCM composite increases 27%–30% compressive strength to alternate conventional materials. Reduction in cell size in PU/inorganic support-PCM leads to a significant reduction of thermal conductivity and improves doped foams′ insulating capacity.[149] Recently, Kim EY set a microcapsule by blending WBPU (aqueous phase) and octadecane (organic phase) as PCM in presence of emulsifiers using homogenizers and
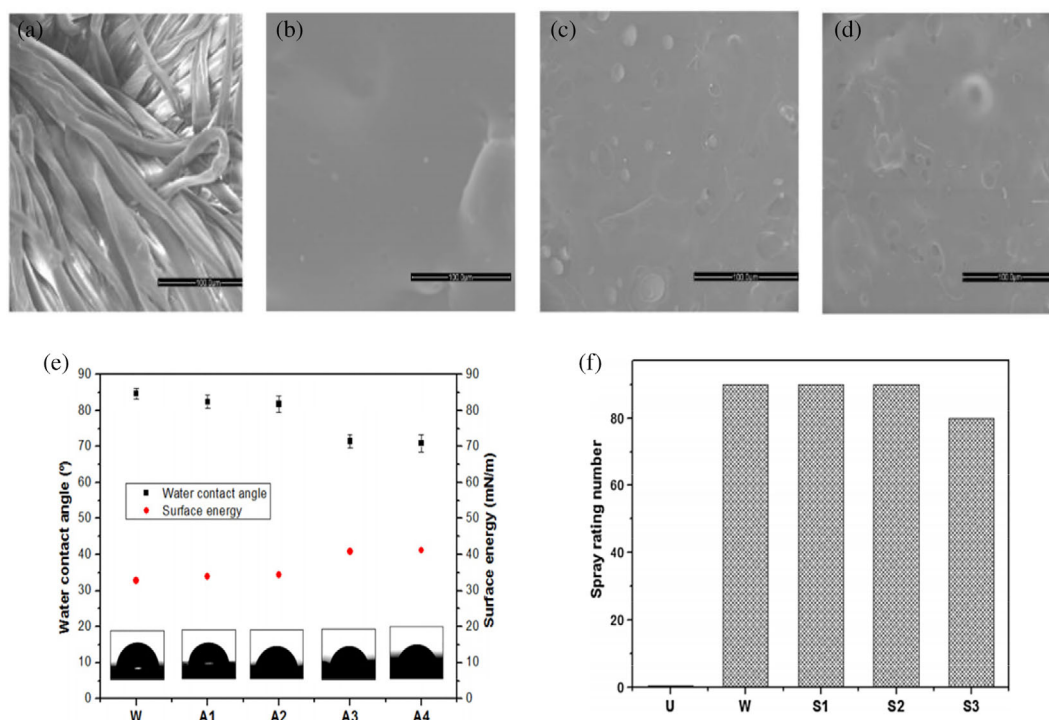
FIGURE 3 SEM micrographs of (a) uncoated cotton (b) PU-coated (c) PU–SAP-coated (1.5%) fabric before chemical treatment (d) after chemical treatment, (reused with permission from Reference [159] © springer nature, 2019), (e) water contact angle and the surface energy of PU and PU-aerogel coated fabrics; W-PU, A1-PU + 0.5% aerogel, A2-PU + 1.0% aerogel, A3-PU + 2.0% aerogel, A4-PU + 3.0% aerogel (f) water repellency of cotton fabric coated with PU and PU–SAP at different concentrations; U-uncoated cotton fabric, W-coating with WPU binder only, S1-0.5% SAP coated fabric, S2-1.0% SAP coated fabric, S3-1.5% SAP coated fabric, (reused with permission from Reference [158] © Elsevier, 2019) [Color figure can be viewed at wileyonlinelibrary.com]

formulated coating materials from WBPU/octadecane/emulsifier blends, thickeners, and hardeners. Addition of thickener and hardener plays a vital role in trapping microencapsulated octadecane within WBPU. Later, the prepared coating mixture was used to coat nylon fabric and finally investigated the thermal behaviors of coating materials at various conditions and established that WBPU/octadecane-coated nylon fabric shows higher heat transfer properties than those of the control and WBPU-coated nylon fabric.[150] However, the fabrication process of PCMs-PU composite deals with complexities that include the possibility of de-foaming action of micro PCMs with co-polymer (melamine–formaldehyde) shells and the rapid rise of the mixture viscosity, caused by the reaction between isocyanate and hydroxyl groups on the shell surface.[144,151]

ability to protect in various harsh environments rather than its esthetic features.[152] Materials for such barrier textiles are selected based on the type of hazards, environments, and materials available for selection.[153] Various types of protective textiles are available based on their specific application areas, for example, industrial, military, civilian, agriculture, medical, sports, space, etc.[153,154] These areas can be associated with two main barrier functionalities: chemical and ballistic protection. While the performance of the chemical protective clothing is determined by its permeation, penetration[155] and degradation characteristics, high strength and stiffness to weight ratio indicate for the ballistic protective cloths.[156,157] PU has gained popularity because of its ability to exert decent strength, liquid penetration resistance, and breathability.[64]

4 | PU FOR PROTECTIVE CLOTHING

Protection is mandatory for the personnel who are continually at the risk of being exposed to hostile environments. The efficacy of protective clothing is scaled by its

4.1 | PU incorporated chemical protective textile materials

Chemicals protective textile materials are frequently expected to be designed to defend the wearer from different toxic and precarious chemicals reducing the threat of
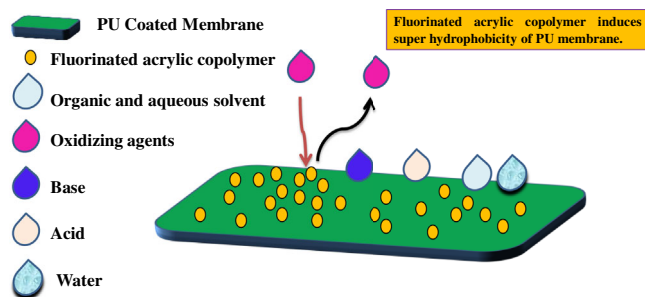
FIGURE 4 PU coated membrane with fluorinated acrylic copolymer. *Source*: Idea taken from References [162,163,165] [Color figure can be viewed at wileyonlinelibrary.com]

injury and illness of human body.[153] Researchers have worked on a wide range of chemical protective textile materials by incorporating coated surfaces on a base fabric through lamination or coating techniques where various polymers like PU, PVC, PVDC, PTFE, and PE were successfully introduced.[63]

Integration of PU binder to the hydrophilic natural fiber provides hydrophobicity of the composite and demonstrates the greatest resistance against the penetration of water and liquid chemicals like acetone, ethanol, DMF, toluene, acetic acid, formic acid, n-decane, hexadecane. Bhuiyan and his research group utilized PU binder and porous silica aerogel on the cotton fabric surface to investigate the thermal comfortability and protective performance of the composite specimen. PU-aerogel-cotton composite exhibited excellent water resistance properties (Figure 3e) due to hydrophobic PU, silica aerogel, and the roughness of the silica aerogel coated fabric at different concentrations.[158] After applying the PU coating, the cloth surface was significantly covered by the binder film (Figure 3b), leaving no fibers exposed as shown in the Figure 3a. However, addition of sodium poly acrylic acid as superabsorbent polymer (SAP) in PU-cotton composite imparts insignificant negative impact on the repellency of the fabric reducing the water contact angle (Figure 3f) since they have the propensity of absorbing and retaining water or aqueous solutions. However, the unaltered surface morphology PU–SAP coated fabric after chemical treatment (acetone, ethanol, DMF, *n*-decane, hexadecane, acetic acid, 85% formic acid, and 40% caustic soda) suggests the durable resistance against liquid chemicals (Figure 3c,d). Owing to their hydrophilic nature, they improve the air permeability of the composite materials to some extent and present simultaneous protection with thermo-physiological comfort to the wearer by providing improved heat and vapor transmittance.[159]

Interpenetrating polymer networks (IPNs) are the combination of two or more polymers where a partial interlacing on the molecular scale is present in the matrix. However, there is no covalent bonding, and therefore, the polymers cannot be separated unless the chemical bonds are broken.[160] Conversely, semi-interpenetrating polymer networks (SIPNs) are hydrophilic polymers-based solutions having two independent components, where one is a crosslinked polymer and the other one is noncrosslinked polymer. In SIPNs, crosslinked polymers constitute between 10% and 90% of the semi-interpenetrating network composition. IPNs and SIPN in PU can provide a surprising effect on the resistance properties of PU coated composite.[161]

For example, alkali-treated bamboo (*Bambusa balcooa*) fibers while coated with PU/PS (50/50) SIPN shows a higher degree of resistance against water and different solvents (acid, base, oxidizing agents, and organic and aqueous solvent) even more than the PU coated one (Figure 4). This is because both the polymers are water repellent and between the two polymer coatings, PU/PS coating has the edge over PU. On the contrary, PU/polyacrylonitrile (PAN) SIPN coated composite materials showed the reverse consequence. The reduced chemical interaction between PU coated fiber and chemical reagents can be attributed to the absences of more polarity polymers like PAN.[162,163] Besides, PU/acrylic latex based IPN coating shows commendable resistance against stain, water, microorganism, weather, light, and high temperature when applied to the synthetic woven fabrics in presence of a crosslinking resin and an organic fluorochemical. This coated fabric shows durable outdoor applications, especially as sun awnings, lawn and patio umbrellas, boat covers, and the likes.[164]

In addition to chemical protection, electrospun PAN/PU/TiO$_2$ nanofibrous membranes impose multifunctional properties (UV resistance and waterproofness) when modified with 2-hydroxy-4-*n*-octoxybenzophenone (UV531) and fluorinated acrylic copolymer (FAC). UV531 absorbs light of UV region directly to convert it into heat energy and their benzene ring imparts hydrophobicity to stir up waterproofing nature of the base fabric. FAC ensures super hydrophobicity of the pristine membranes. The prepared composite membranes have diverse possibilities of using in high-altitude garments, protective clothing, covering materials, self-cleaning materials, and other medical products.[165]

PU coatings based on acrylic polyols and trimer of isophoronediisocyanate are resistant to acids, alkalis, and solvents.[166] A very thin layer of PU nanofiber web can significantly improve barrier performance for challenging liquids that adds a wide range of physicochemical properties.[167] In addition, functionalized PU nanofiber by *N*-chlorohydantoin can decontaminate demeton-*S*-methyl, a simulant for V-type nerve gas.[168] The two-component PU coatings are widely used in automotive interior and
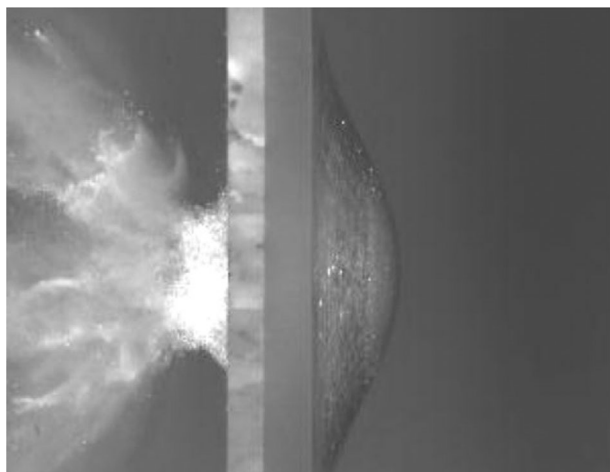
FIGURE 5 Deflection of PU backing due to the impact of a 0.44 FMJ bullet (400 m/s). *Source*: Reused with permission from Reference [73], © springer nature, 2011

electronic device applications due to their high durability and excellent tactual sense. These newly designed multifunctional polyol systems have improved chemical resistance.[153]

4.2 | Ballistic protection

Developing advanced body armor has become an emerging field with increasing international and civilian conflicts. Ballistic protection has become an indispensable requirement, especially for law enforcement officers and military personnel.[169,170] Since the early history of humanity, protection against ballistic material has been a matter of concern. The very first armor was developed from animal skin to protect against the smack of a club. Before developing effective armor in the 1850s by the Australian "bushranger" Ned Kelly, leather, metal plates, and silk were used as protective materials against ballistic threats.

The ballistic threats have increased with the development of small arms ammunition and fragmenting munitions.[171] Different types of bullets include full metal jacket (FMJ), hollow point, soft point, open tip match, plastic tip, armor-piercing, tracer, armor-piercing incendiary, frangible, shotgun ammunition, have different ways of release and penetration mechanisms. Therefore, ballistic protection material differs from the type of ballistic hazard encountered. For example, bomb disposal suits protect the whole body, whereas body armor is designed to preserve significant body organs. This section of this review paper will cover the previous research work done with PUs to develop transparent and nontransparent armor, including soft and hard body armor.

4.2.1 | PU for transparent armor

Transparent armor is mainly produced from bulk polymers. Transparent armor or bullet-resistant glass is a layered composite of glass sheets where elastomers such as polyvinyl butyral, PU, and ethylene-vinyl acetate films bind the glass layers.[73] Goggles and visors are made as transparent polymer sheet armor for using against a low level of ballistic threats.[172] For the higher ballistic resistance, multilayer and thicker glass sheets are used. Carton and Broos proposed innovative transparent armor concepts using PU.[173] They developed two transparent armors-glass-clad PU and glass-pellets in PU. Among these two concepts, the glass-pellets in PU concepts showed decent multihit capacity through lacked optical properties. Considering the optical properties, the glass-clad-PU concept exhibited both excellent optical properties and ballistic resistance as well.[173]

For the glass-clad PU concept, the researchers developed a 5 mm thick float glass panel with a 20 mm thick layer of PU on the rear side of the material. The specimens resisted a 9 mm FMJ bullet at 0° NATO impact at 400 m/s, which indicates that their developed material from the glass-clad concept allowed sufficient ballistic protection. Figure 5 shows the deflection of PU backing due to the impact of a 0.44 FMJ bullet (400 m/s).

Furthermore, Ekeren and Carton investigated the potentiality of PU to use as a backing material for transparent armor.[73] They tested their specimens by using differential scanning calorimetry (DSC) and dynamic mechanical analysis (DMA). The analysis of these tests showed that elastomeric material like PU performs well when the material is in its glass transition range.

4.2.2 | PU for nontransparent armor

Body armor is usually nontransparent. Hard body armor and soft body armor are two different types of body armor.[169] Hard body armor is made from metal plates, ceramic or plastic materials. Reinforced plastic armor, ceramic composite material, etc., are also used to produce hard body armor. Hard body armor is designed to give ballistic protection from powerful projectiles. According to the National Institute of Justice Standard (NIJ), type III hard armor shall withstand 7.62 × 51 mm FMJ and steel jacketed projectile.[174] On the contrary, soft body armor is made from fibrous materials, which make it lightweight with increased comfort, durability, and reliability.

For the hard armors, ceramic and metallic materials have been widely used in the past.[65] Though the metallic materials provide excellent strength and toughness for

hard body armor, its high density makes it bulky and limits the wearer's mobility. To make the hard armor in lightweight, various kind of ballistic ceramics such as, alumina (Al_2O_3), boron carbide (B_4C), silicon carbide (SiC), and titanium-diboride are incorporated, which can provide sufficient hardness and mechanical characteristics to resist the ballistic impact.[175–177] However, the ceramic is an inherently brittle material that cracks after the first impact and makes the ballistic armor ineffective for protection. Therefore, to keep a balance between protection and weight of the body armor, researchers have developed hard body armor from ceramic and metallic back layered composite material.[178,179] Nevertheless, the weight of these composites is still a concern that has been overcome by incorporating organic polymer into body armor. Furthermore, the polymer is easy to process and economically feasible.

Among different polymers, the PU layer is applied to improve the ballistic protection performance of the body armor. For instance, a relevant study found thin pre-layer of elastomeric PU on steel armor increases the ballistic limit against bomb fragments.[180] They reported that this happens because of the impact-induced transition mechanism of elastomer to the glassy state. The principle is that the kinetic energy of the projectile fragments turns to the plastic deformation of the steel. Here, the elastomeric PU layer on the striking face increases the energy dissipation, subsequently improving ballistic protection.

Again, the resin has a vital role in polymer matrix composite ballistic material. Among different types of polymer resin, thermosetting polymers such as ester, epoxy, and phenolic are generally used for ballistic armor.[181,182] Besides these traditionally used matrix resins, researchers also use different resins for ballistic protection. For instance, Kevlar™ reinforced poly(BA-a)/PU composite was developed for ballistic protection applications.[183] Researchers reported, 30 plies and 50 plies of Kevlar™ reinforced 80/20 poly(BA-a)/PU composite can resist the penetration of ballistic impact equivalent to levels II-A and III-A based on NIJ Standard, respectively. Here the addition of 20% PU provided the maximum penetration resistance as only the neat poly(BA-a)-Kevlar™ composite exhibited the complete penetration of projectile. In another research work, the researchers used glass and aramid fiber reinforced polybenzoxazine/PU composites for developing ballistic protection.[184] They used type S and type E glass fiber. They found S glass composite backed by aramid fiber reinforced 80/20 polybenzoxazine/PU composite can resist the level III ballistic impact based on NIJ standard. The finding of their works implies that their developed laminated polymer composite can be used for advanced protection.

Colombo, Zordan, and Medvedovski invented ceramic-polymer composites for lightweight ballistic protection using thermosetting (crosslinked) and elastomeric PU.[66] The conducted study also designed composite layers for the body armor. The front layer was made of a ceramic faceplate, which works to dissipate the impact energy of the bullet and the backing layer absorbs impact energy and stops the bullet from penetrating further. The researchers developed the backing layer from PU infiltrated silicon carbide (SiC) ceramic foams. Among these two types of PU, the thermosetting PU showed better mechanical properties, which could resist the penetration of 5.56×45 mm SS109 and 7.62×51 mm NATO Ball full metal jacket projectiles. This research also implies that the PU in-filtered ceramic foam can replace the expensive polyaramid or polyethylene backing layer.

However, layered composites are delaminated under the influence of shear or transverse load. To overcome this issue, Lyashenko-Miller and Marom interleaved ultra-high molecular weight polyethylene (UHMWPE) fibers/PU matrix composite with thin PU films reinforced carbon nanotubes (CNT).[185] The study revealed that CNT-reinforced PU at the midplane improves the interlaminar fracture toughness, which provides better ballistic protection.

Bullet or bomb fragment penetrates with high pressure and heat. Therefore, a material with suitable elastic property and thermal stability shows the possibility of working well. In that case, PU has both mentioned properties. Also, as per the review done in this section, researchers have worked on the application of PU and found PU as an effective polymer to use for manufacturing ballistic resistant material. Table 4 accumulates some recent research works based on PU in protective applications.

5 | PU IN SMART APPLICATION

Developing innovative and smart materials is one of the most demanding areas for establishing future technology. Smart materials can respond in a pre-programmed or manual system to communicate externally upon applying a stimulant.[189] The usual existing systems mostly involve attaching small electronic devices like sensors, circuits, actuators, etc., into products. In case of wearables, lacing or looping conductive one-dimensional (1D) wires is another strategy. As a result, the functionality of such smart products greatly depends upon these electronic components rather than the product itself acting as the smart device. This sort of attachment contains serious risks, especially for those that stay in our body contact. One way to reduce the use of these devices is by using a

TABLE 4 Recent research works based on PU in protective applications

Materials	Base material	Main function	Additional features	Negative aspect	Application area	Reference
Integrating silica aerogel with PU binder as a coating agent	Cotton	Better chemical resistance	Moisture vapor transfer, air permeability	Chemicals adsorbed due to silica aerogel but no penetration through the fabric	Protective clothing	158
PU and PAN electrospun nanofibrous coated with polyvinylidene	100% polypropylene (PP) spun bonded nonwoven	Barrier performance against pesticide liquids	Air permeability and water vapor transmission	Air permeability decreases with increasing polyacrylonitrile	Agro textiles	186
Electrospun PU fiber web	Spun bonded nonwoven	Barriers to liquid penetration	Thermal comfort	Air permeability decreases with increasing electrospun web area density	Protective textiles for agricultural worker	167
Glass supported by PU backing layer	-	Bullet resistance	Perform best in glass transition temperature	Showed smaller shear modulus at low temperature	Transparent armor	73
Polybenzoxazine/PU reinforcement	Aramid and Glass fiber composite	Improved penetration resistance	Lower weight, less bulkiness	-	Nontransparent armor	184
Glass-pellets in PU	-	Ballistic resistance	-	Lacked optical properties	Transparent armor	187
Polybenzoxazine (BA)/PU alloys	Kevlar composite	Ballistic resistance	-	Char yield decreases with increasing PU amount and leads to poor thermal stability	Ballistic armor	188

functional polymer which will make the product itself act as the driver for smart application.[190–193]

Over the past two decades, numerous research works have been conducted worldwide to turn daily usable products, especially textile-based materials, into smart and multifunctional materials.[194] Figure 6 accumulates some significant areas for smart applications concerning polymers.

PU is one of the materials that show versatility in the field of smart polymers. One of the most prominent application areas of PU is coating for different smart applications. The targeted areas are typically protective clothing, work wear, hygiene and care applications.[200] However, the potential application areas burgeon well beyond the textiles. In the following sections, the authors described the research works carried out in recent times on the specific areas of smart products where PU was involved. The categorization is based on different smart functionalities that PU can display.

5.1 | Stimuli-responsive PU

External stimuli are external factors that might trigger any changes to the subjected polymers.[201] Several stimulations are responsible for numerous changes in the physical, mechanical, thermal, biological properties, etc. Depending on the responses of the stimuli, the end products made from such polymers also display similar property changes afterward, which may be in the form of smart response.[202] PU can carry out the purpose of these stimuli sensing sensors to respond and adapt accordingly by giving proper response in the form of property, behavior or shape changes.[202–204]

PU-based products which can change their flexible structure in response to a stimulus fall under this category.[204] They can undergo significant morphological modifications or changes in functionality with the variation in surrounding environmental stimulation. Thus, unique characteristics can be given to different
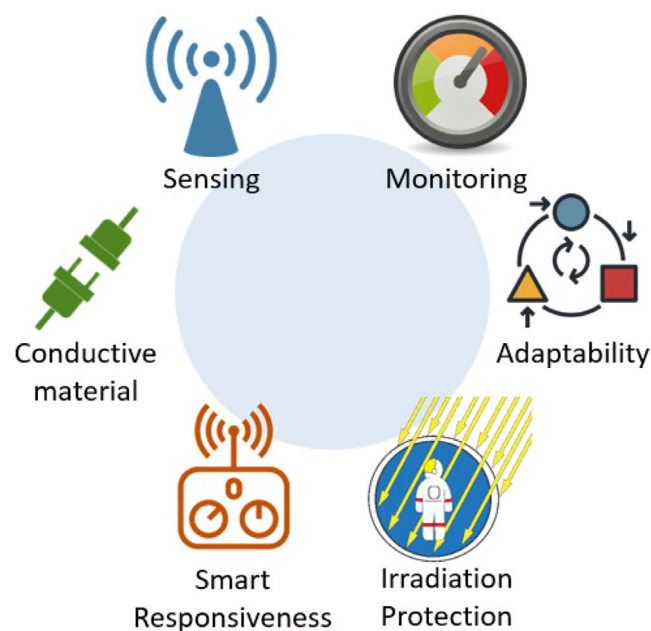
FIGURE 6 Active research on polymer based smart application areas. *Source*: Based on the References [190,191,195–199] [Color figure can be viewed at wileyonlinelibrary.com]

commodities, such as textile products, by integrating stimulus responsive PU with them.[205] The major and most common stimulants are temperature, water, pH, and light. In case of temperature responsive PU, functionality of the smart product can be activated and regulated because of fluctuations in surrounding temperature. This feature is being applied in many areas, from smart wearables, intelligent medical devices, and heat shrinkable packages to technologies as advanced as self-deployable spacecraft structures. Shabaani et al. worked on a PU-based thermos-responsive scaffold for tissue engineering, capable of self-healing (~95%) at a temperature similar to body temperature (40°C).[206] Gao et al. has worked on a temperature-regulated PU core based fiber mat for intelligent drug discharge.[207] Xiao et al. synthesized a temperature responsive PU. They incorporated it to prepare a nanohydrogel with a substantially improved uniformity in crosslinking and demonstrating temperature stimulated coil to globule transition in aqueous medium.[208] Yi et al. reported a three-dimensional PU sponge that showed temperature stimulated smart super wettability applicable in the separation of water–oil.[209] In case of water-stimulated PUs the functionality can be triggered by water, moisture or any aqueous medium due to the plasticizing effect of the water. This results in shape modification and this phenomenon can be accelerated even more with the help of a water soluble ingredient.[210,211] Recently, Li et al. has reported an exciting water responsive behavior of PU. In wet conditions, PU showed a reversible change in water absorption behavior

with potential application to minimize the risk of skidding or slipping on wet surfaces.[212] Han, Hu, and Chen developed an artificial skin penetrating WPU in collagen nonwoven network that shows water triggered shape fixation and recovery.[213]

Further, recently another group came up with water responsive composite PU exhibiting excellent shape memory performance. They highlighted "Action of a Mechanical Hand" as a potential application area of such smart responsiveness.[214] Some PU-based polymers show sensitivity at various pH values in surroundings. Typically, the response is related to swelling or de-swelling of the PU. Kim and Co. used a pH sensitive PU to develop a switchable nanoparticle releasing intravaginal ring (IVR) for on-demand nanocarrier discharge operation.[215] Liao et al. synthesized a PU-based self-assembly micelle capable of controlled drug release triggered by pH,[216] whereas, Chen et al. fabricated a smart surface with pH responsive PU coating on textile surface that exhibited a high oil–water separation efficacy. The super hydrophobic surface showed switchable transition to super oleophobicity at different pH.[217] Similarly, Xi and Jiang reported a self-fluorescent PU multiblock copolymer with reversible ultraviolet absorption property at changing pH and thus established its potential application as pH controlled fluorescent probe.[217] PU-based photo-chromatic materials have the ability to be stimulated by light because of difference in light absorption.[218,219] A porous $Ag_3PO_4@AgBr$ loaded PU and negative ion composite film was reported to demonstrate light responsiveness with potential application in organic pollutant dissemination and germ protection.[220] Ren et al. synthesized a zwitter ionic PU micelle capable of detecting bacteria via near infrared bio-imaging functionality paving way for antibacterial operation potentiality.[221] More recent research on smart responses involving PU have been enlisted later in Table 6.

5.2 | Conductive PU

Some polymers are referred to as "synthetic metals" due to their inherent conducting ability. These conducting characteristics can be incorporated into various applications, including power supply, monitoring, sensing, etc.[190,191,194–197] The method of attaching the conductive element might be either in the form of electricity stimulating electrodes in the post treatment or as conductive fillers in the primary step. Electrospinning is a popular fabrication technique. Among post-treatment techniques, coating, resin processing, electroplating, etc., are worth mentioning.[193,222] PU is a great prospect for such applications. Liu et al. reported a fiber-based flexible conductive electrode sprayed with silver/WPU composite for

wearable electronics applications.[223] Ding et al. electrospun a stretchable yet conductive PU fibrous nonwoven structure applied in the form of sheet or yarn as conductor.[224] Wan et al. prepared a conductive and elastomeric porous PU foam in combination with polypyrrole (PPy) intended for pressure sensing application.[225] Later, study reported elastic knitted fabric treated with conductive polymer poly (3,4-ethylenedioxythiophene) (PEDOT)—polyelectrolyte poly (styrene sulfonate) (PSS) and PU for wearable electronics applications.[226] Guo et al. investigated a conductive electrode formed of PU/polypyrrole/graphene for potential applications in wastewater treatment.[227] Some more recent works with PU with its conductivity properties are mentioned later in Table 6.

5.3 | PU with sensory characteristics

The properties of getting deformed by external stimuli can be incorporated in measuring some critical physiological parameters, particularly in human body.[205,228,229] Numerous applications of PU can be corresponded in sensing common readings like heart rate, respiratory rate, body temperature, and various gestures (Figure 7).[205,229,230] Therefore, a stretchable and body attachable skin like multifunctional sensor made of multiwalled CNT/polyaniline coated PU for e-skin and health monitoring application was reported by Hong et al.[231] PU-based strain sensors for human motion and health related strain sensing were also reported.[232,233]

Scientists have conducted a detailed study on making foams capable of sensing pressure. For example, a piece of PU foam soaked with the mixture of PPy and Naphthalene Disulphonic Acid (NDSA) or Polyaniline (PANI) showed revolutionary results demonstrating pressure sensing ability of PU foam. The principle is to generate different resistance at different weights measured by my multimeter and send the value in a signal form. Figure 8 represents schematic of such an arrangement. In addition to good environmental stability, they show good sensitivity. The application area is not being confined to pressure sensing but also covers foreseeable applications throughout rechargeable batteries, bio and chemical sensors, transducers, Electromagnetic Interference shielding, etc.[190,204,233–237] Huang et al. studied conductive thermoplastic PU porous foam with carbon nanotubes (CNT) for sensing pressure.[238] Other PU foam-based pressure sensors triggered by compression deformation were reported by various scientists.[239–246] Among different types, PU fiber-based[247] and hybrid structures[248,249] were also reported as pressure sensors.

The application of segmented PU (SPU) as an optical device is possible because this material can act as a chromogenic copolymer. SPU along with copolymers like acetylene can be incorporated as a thermos chromic element. It is suitable for temperature sensor application. In general, the optical fiber is coated with cladding layer of SPU copolymer to get desired result as an optic sensor. Already the application of such research is visible in military uniforms, where optic fibers coated with SPU-diacetylene

FIGURE 7 Physiological parameters sensing ability of PU-based smart products. *Source*: Developed based on References [205,229,230] [Color figure can be viewed at wileyonlinelibrary.com]
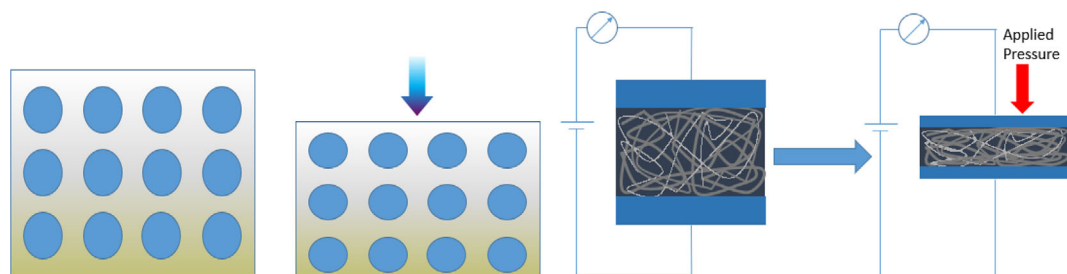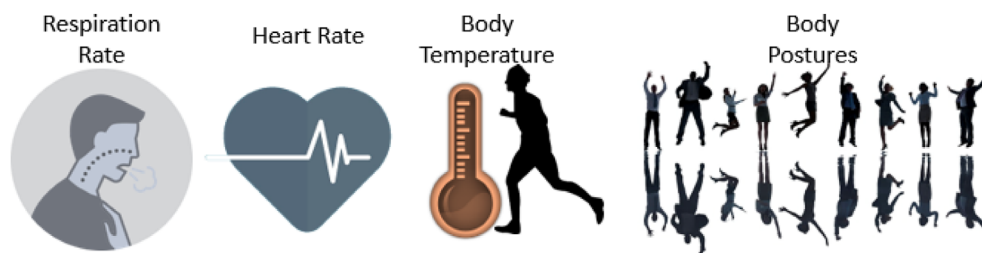

FIGURE 8 Schematics of pressure sensing PU foam relaxed state versus compressed state. *Source*: Recreated based on the References [204,250] [Color figure can be viewed at wileyonlinelibrary.com]
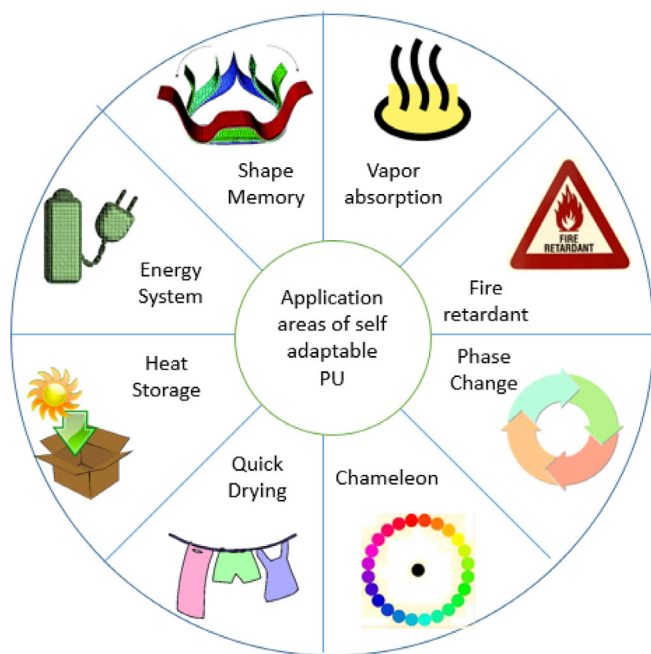
FIGURE 9 Some areas of characteristics for application of self-adaptability [Color figure can be viewed at wileyonlinelibrary.com]

temperature sensing film were used to get protection and warning in advance on the battlefield under different circumstances.[191,251–254] Using SPU with diacetylene increases the possibility of cross polymerization rather than random cross linking, which may occur unevenly throughout polymer chain. The segmentation in the SPU limits this random distribution of cross linking, providing a more or less uniform distribution. It helps to expose the optical characteristics on a better standard.[252,255,256]

Shape memory principled PU-based temperature sensor was reported by Zhang et al. for effective and safe application in fire alarm systems.[256] A PU/carbon composite for fabricating dual emission temperature sensing applications was reported by Jiang et al.[257] Jasmi et al. fabricated a PU/graphene nanocomposite based fiber bragg grating (FBG) device for temperature sensing.[258]

5.4 | Self-adaptability of PU

Self-adaptive polymers are modern building blocks for smart and intelligent products that display specific, distinct, predictable, and smart responses when triggered by small external stimuli such as pH, temperature, ionic strength, chemical, and light.[201,259–261] The self-adaptability characteristics are connected to shaping the memory property of PU. This can be applied to make innovative products, for instance, wearables that will adapt to customers' providing improved comfort sensation.[262–264] Typically, these characteristics can be incorporated into

functional PU with specific composition, blending PU with specific molecular additives or modifying PU via various physical/chemical treatments.[201] Applications of self-adaptable PU encompass energy systems, chameleon products, vapor absorption, phase change composites, shape-memory applications, heat storage, fire retarding commodities, quick-drying, etc. (Figure 9).[201,262] The coating of SMPU can add dimension to some smart abilities as finishing applications. Researchers have found if hot air is blown over cotton fabric finished with SMPU, it can retain its original flat shape within a matter of time. The main idea is to reduce –OH groups on cotton fabric surface by pretreatment of the fabric. Then anti-wrinkle finish can be introduced using WBPU acrylate under UV radiation, where click reaction occurs. Also, hydrophilic PU can mitigate discomfort if used in conjunction with existing formaldehyde based resin finish.[262,265–267] Research concerning PU's vapor absorption for specific application areas like packaging materials,[268] omniphobic surface coating,[269] volatile organic vapor detection,[270] etc.

5.5 | Smart wettability

Coating of PU on any surface can provide subjected surface with intelligent wettability. This effect conventionally can be imparted by providing hydrophobic surface where required characteristics can be obtained with micro or nanosized pattern with homogeneous intervals. PU shows different shrinkage effects when subjected to heat contact, mainly because of the phase transmission. This allows the PU coating to control the movement of the water on the coated surface, which may be integrated with programs to provide intelligent wettability, with most potential use in the oil and water separation process. This smart wettability is possible due to combined sliding and contact angles control. These, in combination, dictate the behavior of water repellency to water adhesion, also referred to as membranes with switchable wettability.[263,271,272,273] Guo et al. prepared a PU-based sponge with smart wettability for efficient oil/water separation.[274] In a similar study, Xu et al. exhibited a PU-based carbon foam with pH responsive smart oil recovery application.[275] Zhang et al. recently reported a super hydrophobic PU film for electrically triggered smart capture or release of liquid or solid.[276] Table 5 represents some of the recent research on smart application of PU.

5.6 | 4D printed SMPU

4DP, a relatively modern concept, has gained popularity since it was first introduced in 2013. This technology seeks

TABLE 5 Recent research works based on smart application of PU

Responsive stimuli	PU type/ component	Function	Smart features	Potential application area	Reference
Temperature	Pyridine-type zwitter ionic SMPUs (PyZPUs)	Dual-shape memory effects (SMEs), triple-SMEs and quadruple-SMEs upon application of temperature	Superior biocompatibility (when pyridine section >21%)	Biomedical	277
Temperature	DHIR and PU hydrogels	Actuators based on bi-layered (DHIR-PU) thermo-morphic material	Large temperature range of application	Advanced manufacturing, soft robotics, medical devices	278
Water	PyZPUs	Moisture sensitive SMEs	Superior biocompatibility (when pyridine section >21%)	Biomedical	276
Water	PHP	PEG provides water-responsiveness	Prompt shape recovery (>10 s)	Biomedical	279
pH	PU-CAP coaxial fiber	Capable of discharging drugs regulated by pH of bio fluids like semen	Elevated mechanical strength (13.27 ± 2.32 Mpa)	Intravaginal drug delivery	280
pH	PECU based SMPU	Shape memory functionality at various pH	Enhanced mechanical features	Smart biomaterial, sensors, and actuators	281
pH	BES-Na based PU micelles	pH tunable drug release, Discharging rate—79.17% (at pH 6.8) and 89.83% (at pH 5.0) within 24 h	Superior drug release efficiency at acidic condition	Smart drug delivery nanocarrier	282
Light and radiation	PEG based PU hydrogel	UV irradiation can instigate and tune the swelling properties of the PU hydrogel consequently affecting the content amount and content ratio inside the PU network	Superior mechanical and elastic properties	Biocompatible drug career	283
Light and radiation	Graphene-based PU	Shape memory (90%) and self-healing (76%) properties in infrared light	Flame retardancy and water resistance	Functional composite polymers	284
Conductivity and EMI shielding	ZnO/silver-based waterborne-PU (WPU) thin film	Uniform composite structure provides highest conductivity (1.27×10^4 S/m)	Tunable EMI shielding performance (92% retention of shielding capacity after 1000 folding cycle)	Wearable and portable electronics	285
Conductivity and pressure sensing	MWCNT and graphene coated PU sponge	Great conductivity due to MWCNT networking in PU matrix	Very low density and higher compressibility (75%), enhanced pressure	Health monitoring, wearable electronics, motion detection	286

(Continues)

TABLE 5 (Continued)

Responsive stimuli	PU type/ component	Function	Smart features	Potential application area	Reference
			sensing (up to 5.6 kPa)		

Abbreviations: DHIR, dipole–dipole and hydrogen-bonding interaction reinforced; PEG, polyethylene glycol.
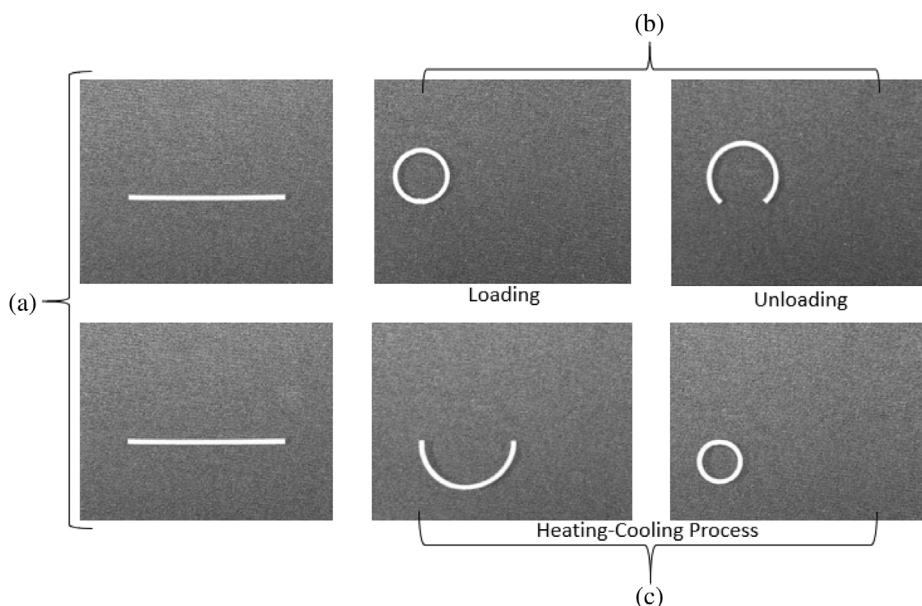

FIGURE 10 Illustration of PU-based SMP via 4D printing; (a) beam after printing, (b) beam after loading and unloading by cold programming, (c) heating–cooling process. *Source*: Reused with permission from Reference [288], © IOP Publishing Ltd., 2018

to incorporate 3D printed materials with additional stimulus responsiveness and programmable architectural deformation. Shape memory polymers like PU is a great candidate for this 3DP-based smart technology.[286] This multifunctional polymer can be deformed and retained back the original shape by applying external stimuli, mostly temperature.[287] As a result, they can be shape programmed while fabricating and depositing on the targeted surface. Thus, the necessity of post modification to achieve desired architecture can be readily avoided.[288] Hot and cold temperature triggered programming are popular mechanisms.

For instance, Bodaghi et al.[288] used the hot-cold programming approach in 4DP technology to demonstrate an adaptive structure with the triple shape memory effect. The authors developed a PU-based triple SMP for mechanical and bio-medical devices. The effectiveness of the developed polymers was investigated in both low and high temperatures by performing elastoplastic and hyperelastic thermos mechanical characterization. They used different speeds (10, 30, 40 mm s^{-1}) to print beams with cold programming (23°C) and bending (360° circle) by a 4DP. Figure 10a shows the configuration of the beam after printing, whereas Figure 10b illustrates the configuration of the beam after cold programming. After the cold programmed printing, they subjected the curved beams

to a hot temperature (85°C) and then again cooled down to 23°C to complete the heating–cooling process. Figure 10c illustration of the heating–cooling configuration of the beam after 4DP. As demonstrated in Figure 10c, the hot-cold process can successfully be applied to PU-based 4D printed SMP to impart a self-bending feature to it.[288]

Therefore, considering the effect of hot-cold temperature in SMP, understanding the shape transition caused by shape memory performance is crucial for 4DP technique.[290] The shape memory performance depends on the difference between elastic modulus and yield point. Glass transition temperature (T_g) plays a vital role in this regard. Generally, the yield point is high below T_g and low above T_g. For applied strain cycle, loading increases temperature and oppositely, unloading decreases temperature. This phenomenon of temperature changes caused by material deformation depends on materials, condition and applied strain rate. Greater strain rate causes greater stress and temperature change. Pieczyska et al. further investigated the mechanical behavior of shape memory PU (SMPU) under 10 tension cyclic loading and unloading.[291] They observed temperature increase for loading and vice versa and this deformation of SMPU directly influenced the strain rate.

TABLE 6 Recent research works regarding medical applications of PU

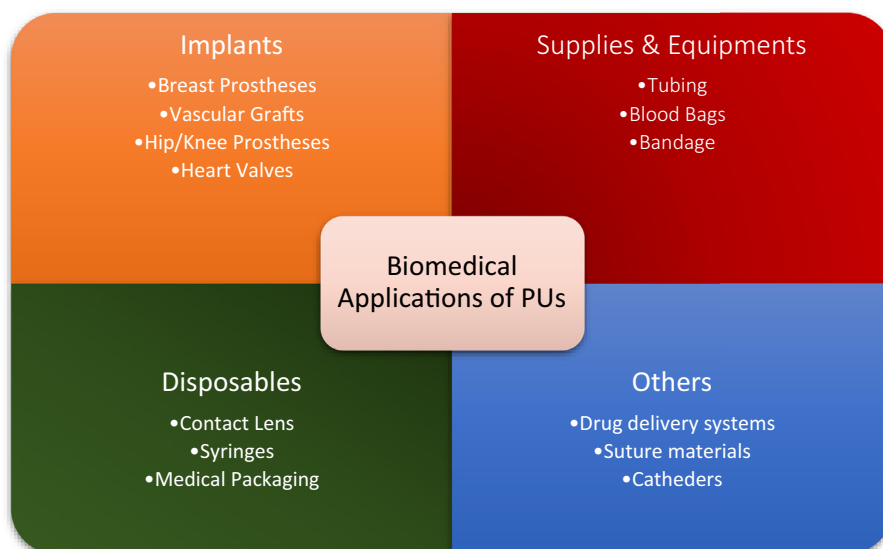| Components	PU nature	Biocompatibility	Structure	Fabrication technique	Feature	Application	Ref
PU and murivenna oil blend	Thermoplastic	Yes (tested with blood)	Electrospun nanofiber mat	Electrospinning	Improved water attraction and wettability	Wound dressings scaffold	358
PU, corn oil and neem oil blend	Anticoagulant	Yes (nontoxic to RBC and HFC)	Electrospun nanofibers	Electrospinning	Enhanced mechanical strength (PU/corn oil—11.88 MPa and PU/corn/neem oil—12. 96 Mpa)	Bone tissue engineering scaffolds	359
Collagen/PU blend	Bioactive	Yes (tested with MG-63 osteoblast-like cells)	Bio artificial Coating	Salination technique	Exhibited good compatibility in both SBF and PDPC environment	Bone tissue engineering	360
HA nanoparticles combined PU	Thermoset	Yes (tested using human lung fibroblasts)	PU-HA composite foam	Ultrasound mixing and polymerization	Improved fibroblast adhesion of connective tissue without instigating inflammation	Tissue engineering	309
HAP/GG/PU sponge	Bioactive	Yes (tested with MDCK cells)	Circular scaffolds	Polymeric replication technique	Tunable porous structure, performance (79.3%) with cell generation support	Bone tissue engineering	361
PU/zinc nitrate nanocomposite	Thermoplastic	Yes (tested with blood)	Nanofiber scaffold	Electrospinning	Enhanced hydrophilicity (84° ± 4.041°), mechanical strength (15.98 MPa), surface roughness (277 nm)	Wound dressings	362
PU/ginger extract composite	Thermoplastic and bioactive	Yes (tested with RBC and HDFC)	Textile scaffold	Electrospinning	Developed wettability (80.33° ± 4.51) and roughness of surface (389 nm); improved proliferation rate; faster blood clotting	Skin tissue engineering	363
PU/copper sulfate (CuSO$_4$) nanofiber blend	Antimicrobial	Yes (tested with blood)	Membrane	Electrospinning	Enhanced hydrophilicity, tensile strength (18.58 MPa) and cell viability rate	Wound healing	364
Ag/Cu nanoparticle incorporated PU	Biologically active and antibacterial	Sample did not exhibit toxicity up to 14 days	Metal integrated nanomaterials	Electron beam evaporation and vacuum deposition	Bactericidal characteristics (gram positive, gram negative bacteria and fungi)		
Implants, drug delivery, antibacterial coating and packaging	365						
PPC/TPU blend	Thermoplastic SMPU	Yes (tested with blood and cells)	Spiral shaped stent	Melt blending	Enhanced shape recovery ratio ~95%, rapid response at 37°C	Biomedical stent	366

(Continues)

TABLE 6 (Continued)

Components	PU nature	Biocompatibility	Structure	Fabrication technique	Feature	Application	Ref
Regenerated silk fibroin/PU composite	Nondegradable	Yes	Tubular mesh	Electrospinning	Avoiding after processing phase separation for clinical applications	Tissue engineering	366
TPU carrying antibiotic (levofloxacin)	Thermoplastic and bio active	-	3D printed mesh	Hot melt extrusion	Bacteriostatic activity in both *Staphylococcus aureus* and *Escherichia coli* cultures	Vaginal mesh implants	368
WPU/Carbon and metal fillers composite	Conductive	-	Film and foam	Transfer coated	Capable of operating as skin electrode of medical devices	Medical orthoses and postural devices	369
G-HTPB (g-COOH)-PEG triblock PU	Thermoplastic	Yes (tested with normal epithelial cells)	Micelle	Polymerization and dialysis	High drug loading capacity (pH dependent) over various concentrations	Nanocareer for drug delivery	370

Abbreviations: PPC, propylene carbonate; RBC, red blood cell; HFC, human fibroblast cell; HA, hydroxyapatite; HAP, hydroxyapatite; GG, gum ghatti; MDCK, Madin–Darby canine kidney epithelial cell; HDFC, human dermal fibroblast cells; WPU, water based PU; PCL, poly(ε-caprolactone); SBF, simulated body fluid; PDPC, human periosteal derived precursor cell.

Soft polymeric hyper-elastic and elasto-plastic features of PU enhance its technical periphery to be used with 4DP. Dual material based meta-structure with reversible characteristics is one of the practical examples of such behavior. These structures are responsible for introducing some exceptional features like variation in softening/hardening, thermal effectiveness, unusual dynamic performance, etc. The targeted energy absorption performance of such meta structures is instigated by converting kinetic energy into others via elastic/plastic deformation, structural breakdown, zero/negative poison's ratio, etc.[291] Negative poison's ratio characterized by 3DP auxetic structures or behaviors tends to show better stiffness and energy absorption.[289] The most effective among the aforementioned energy absorption techniques is via plastic deformation. The repeatability of the unit cells characterizes the materials absorption behavior to a great extent. Cyclic loading-unloading experiment can reveal information about materials repeatability. If repeatability declines with dimension, the material may fail during loading-unloading and plastic deformation.[292] Bodaghi et al. also reported about significant deformation responses of soft 3DP hyper elastic metamaterials. Such metamaterials demonstrated anisotropic and unstable mechanical responses under both tension and compression. Typically, factors like unit cell shape, direction and mechanical loading intensity considerably impact such behavior.[293] Additionally, advantages concerning lower cost, lighter weight, flexible design, biocompatibility, etc., have helped PU gain advantage over others.

Bodaghi et al. utilized SMPU filament to fabricate shape adaptive, reversible thermo-mechanical constructions via fused deposition modeling (FDM) 3DP technique. The SMPU 4DP structure demonstrated phase-dependent elastoplastic, hyper elastic responses and temperature depending on geometric programming. The potential application areas can be self-deployable biomedical stents, adaptive surface structures and memory devices.[289] Similarly, Damanpack et al. studied the PU filament based 4DP beam structure's behavior under compact/impact loadings. The experiment revealed that the 4DP PU beam could successfully recover large residual plastic deformation caused by high/low velocity-low energy upon heating.[294] In another study, Bodaghi et al. printed SMPU to fabricate an adaptive metamaterial equipped with self-foldable functionality with FDM. Such 4DP smart architecture can potentially contribute to making self-tightening surgical products, self-conforming objects, structural/dynamic switches, self-coiling stents, etc.[295] Another 4DP PU structure with similar behavior was reported by Hu et al., demonstrating potential application in designing smart materials and architectures. Stimulation in heating could successfully transform 3DP

Biomedical Applications of PUs

Implants
- Breast Prostheses
- Vascular Grafts
- Hip/Knee Prostheses
- Heart Valves

Supplies & Equipments
- Tubing
- Blood Bags
- Bandage

Disposables
- Contact Lens
- Syringes
- Medical Packaging

Others
- Drug delivery systems
- Suture materials
- Catheders

circular/square flat plate shape into curved shell architecture. This thermo-mechanical smart self-bending ability was pre-programmed during printing.[288] Bodaghi et al. also worked on a triple SMPU based 4DP structure. While traditionally dual SMPs are capable of remembering only one shape, more recent and advanced triple SMPs are known for recovering the original shape from two temporary deformations. The target of the study was to reaffirm FDM 4DPs capacity and introduce necessary computational tools for future smart material design with smart polymers like triple SMPU.[287] Another research work by Bodaghi et al. introduced the 4DP meta-sandwich structure for reversible energy absorption application. FlexPro-PU as soft-hard filaments combination was fabricated as dual material lattice shaped energy absorbers. The auxetic structure could develop a stiffness range and dissipate energy due to elasto-plastic or hyper elastic nature of the constituents. Potential application areas for such performances are protective packaging, crash impact mitigation in vehicles, airplanes, etc. and personal protective equipment.[291]

6 | APPLICATION OF PU IN MEDICAL

PU polymer has some spanking properties which are controllable in various ways. This is the reason they are considered one of the suitable candidates for bio medical applications. Besides, they contain high biocompatibility as well as excellent mechanical properties along with design flexibility, lower cost, and processability. Their most significant characteristic is their capability to undergo changes chemically with the possibility of limitless modifications structure and function wise.[297–300,301] Thus, PU can effectively cover a wide range of medical areas by controlling common properties, including mechanical properties, oxygen permeability, and barrier properties.[67–69,302–304]

6.1 | PU in bio-medical components

Bio-stable and degradable PUs are being used as different ingredients and structures in artificial organs and medical devices. PUs are popularly being incorporated in drug delivery systems and inlay materials for tissue repair. Furthermore, with the advancement in membrane development, it has become quite pragmatic to obtain artificial skin, bandages for wounds, angioplasty balloons, neural connectors, etc.[296] Figure 11 encompasses the important biomedical application areas of PUs.

Excellent biocompatibility, easier processability, exceptional mechanical strength with flexibility, flexural endurance, and abrasion resistance have made PU one of the versatile and ideal polymers to be used as biomedical devices.[67,296,306,307] Medical components and their applications range throughout as adhesives, coatings, medical equipment, synthetic coverings, wound dressings, etc.[79,296] Biodegradability is one of the crucial requirements for making them environment friendly. PU is considered a biodegradable polymer mainly because of its chemical structure consisting of segmented polymer. It contains monomers carrying two or more hydroxyl groups (soft segment). The ─OH groups react with monomers bearing two or more isocyanate groups (hard segment). Thus, degradability is the consequence of the eruption of hydrolytically sensorial soft segment bonding.[308]
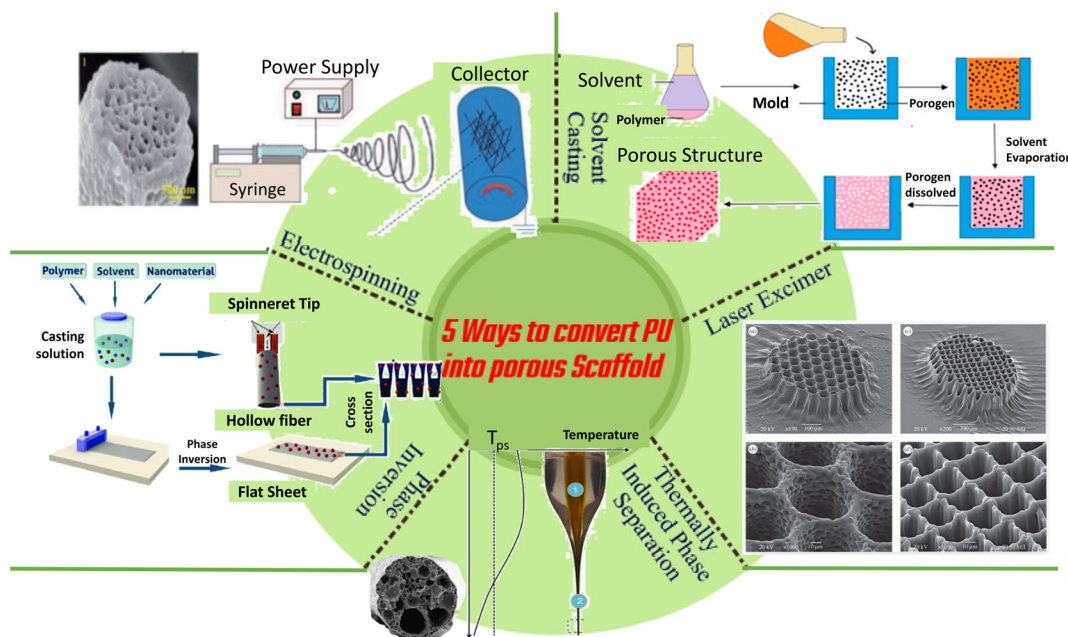
FIGURE 12 Different ways of converting PU into porous scaffold. *Source*: Recreated based on ideas from References [307,316–321] [Color figure can be viewed at wileyonlinelibrary.com]
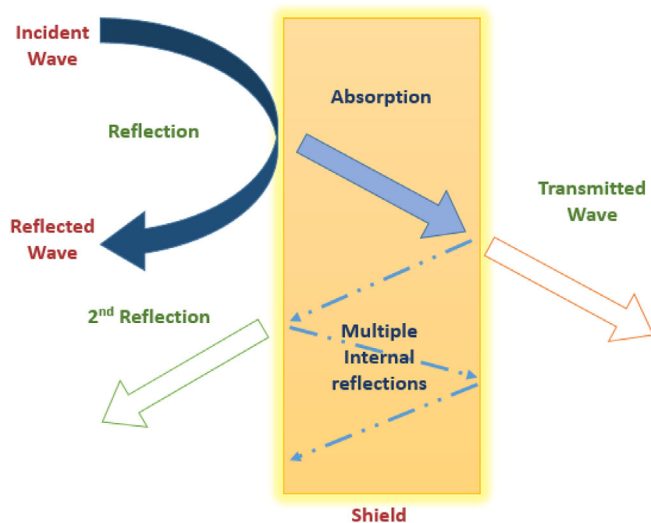

FIGURE 13 EMI shielding principle. *Source*: Recreated from Reference [327] [Color figure can be viewed at wileyonlinelibrary.com]

intended end-use. Evolvement and characterization of matters (fibers, films, membranes etc.) for such applications are being ensued depending on structural, thermal, and biological behavior of PU.[297,310,311] For instance, in tissue engineering, bioactive composites ensure improved scaffold properties and reduced degradation of the structure.[312–314] In order to have desired biological repercussions like migration, maturity, proliferation etc., it is naturally quite significant to govern the micro environment at cellular level inside scaffold.[315]

A porous structure performs better than a smooth surface to attain intended goals. There are several ways to convert PU into scaffold with porous structure (as shown in Figure 12).

6.2 | Shielding electromagnetic interference

Technology is advancing day by day. In the age of high-speed data flow, the term "communication" is of great essence. Fast communication requires higher frequency. It may range from microwave to millimeter waves. The electromagnetic waves that circulate around when different electronic equipment are subjected to use may have hostile effects on the performances of other equipment and can cause damage to the human body. The ever generating electric and magnetic signals from daily used devices like computers, microwave ovens, and radio

Many recent studies have been conducted on PU combined along with other elements applied as scaffolds to act as a three-dimensional matrix for cell adhesion, proliferation, differentiation and tissue ingrowth though imitating natural extracellular matrix. These scaffolds may be applied in the form of diversified features such as foams, sponges, fibers, membranes, tubes, films, or as injectable hydrogels etc. mostly depending on the
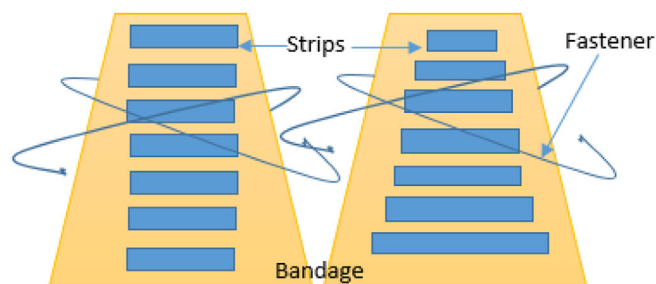
FIGURE 14 Pressure bandages equipped with SMPU strips. *Source*: Recreated from References [79,330] [Color figure can be viewed at wileyonlinelibrary.com]

transmitters can contact other signals to produce amplified and unpredictable electromagnetic waves. It may have complex and adverse effect on human health as well. This phenomenon is known as Electromagnetic Interference (EMI). To minimize these effects, polymers with some special characterizations are being researched and developed every day; SMPU series signs up as one of them.[79,322,323]

EMI Shielding refers to the reflection, absorption, and adsorption of the Electromagnetic radiation by any surface, thus not permitting the 100% penetration of such radiation. Although metal seems more suitable for the intended purpose, it has limitations like heavyweight, corrosion, and physical rigidity. On the other hand, PU has gained popularity due to its large recoverability, superior molding, and lightweight. Such shielding property can be acquainted with aspect ratio, that is, ratio of length to diameter of the filler present in the polymer. Vargas et al. developed a PU-based composite in combination with montmorillonite/PPy-dodecylbenzenesulfonic acid (MMt-PPy. DBSA) for potential electro-magnetic application.[324] This can be applied briskly in medical devices. The EMI attenuation in shielding is mostly due to three reasons, (a) reflection of wave from the shield, (b) absorption of wave as it passes through the shield, (c) multiple reflections due to various surfaces or interfaces within the shield. This characteristic of PU is applicable in medical equipment to ensure shielding.[79,322,325,326] Figure 13 illustrates the schematics of EMI shielding.

6.3 | Use of PU as pressure bandage

Several studies suggest that difficulties in treatment of diseases like ulcers can be alleviated in multiple ways where exterior pressure exertion can be of assistance.[328] This external pressure can be employed by driving fluid using external compression using vascular and lymphatic compartments.[329] It is possible to impart sufficient

pressure with the help of pressure bandage. Shape memory characteristics of polymers like PU are put to provide adequate pressure. This pressure comes from the deformation energy that is stored in the SMPU and released when recovery is being performed and triggered by some external stimuli.[330–332] This strength release can impart pressure on any circular object. A bandage with pre-stretched strips of temperature responsive Shape Memory Polymers was recently invented. The bandage with strips can be wrapped around a body part that will gradually shrink by applying an external stimulus in the form of heat.[79,330] The underlying mechanism is shown in Figure 14.

6.4 | Self-healing

In an engrossing attempt to imitate nature, many studies have been conducted to construct such a material that will be capable of healing or repairing on its own.[333] The general characteristics of polymer describe its inability to regain initiatory strength or protective capabilities after getting deranged by any phenomena.[79,334,335] Self-healing property is directly stringed with the Shape Memory characteristics of the polymer.[336] So, PU has emerged as class of polymer having great potential in this aspect by healing invisible micro-cracks. Self-healing might be propagated by micro capsule with liquid healing agents, solid-phase catalysts or micro vascular network. Over the past decades, scientists worldwide have been exploring this topic and have developed multiple processes to synthesize PU containing self-repairing properties. The self-healing polymer formation and synthesis principle fall into two major classes, extrinsic and intrinsic self-healing. In case of extrinsic self-healing, the healing depends heavily on microcapsule or pipeline containing healing agents. When a crack is formed, the capsule/pipeline will somehow break or leak to release the self-healing agent to activate the process. Dynamic reversible covalent or ionic bonds take charge of the whole process in intrinsic self-healing. Studying the analogy of both principles, one can easily state that intrinsic self-healing will be much faster than extrinsic one as the latter relies on the availability of correct healing agent. The ultimate strategy most scientists used was to use the "Close Then Heal (CTH) or Self Memory Assisted Self-Healing (SMASH) concept".[337–340] Likewise, meltblowing techniques have been used to cross-link PU and polycaprolactone with MWCNTs, which showcases both shape memory and self-healing ability. The mixtures of MWCNTs allow the product to react against near-infrared radiation (NIR) signals. It is quite possible to control the self-healing and shape memory capability from a distance.[79]
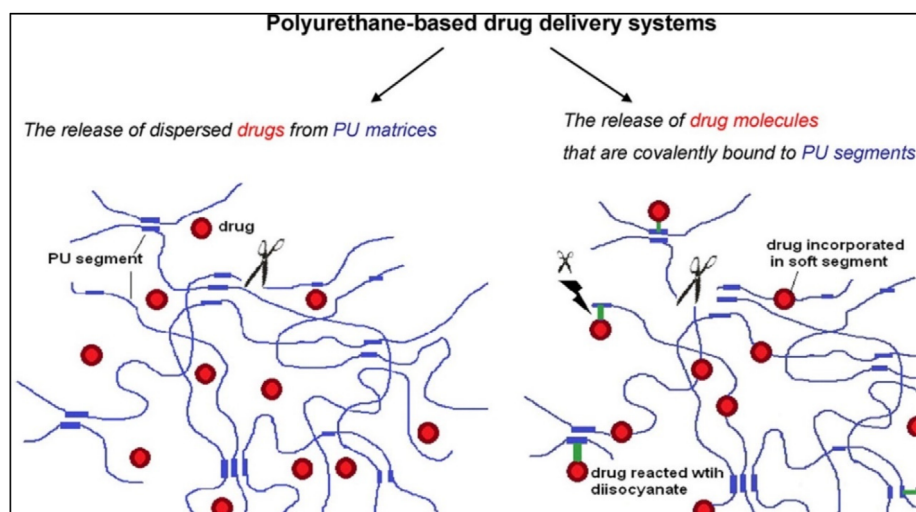
6.5 | PU in biomedical applications

6.5.1 | Drug delivery

A modern concept in the field of biomedical is the potential use of PU nanocomposite (PUNC) as a career for controlled drug delivery. The drug-carrying materials must be insured safe, biocompatible and nontoxic; PU has been found suitable for the job not only because they contain above characteristics but also, they show better compatibility with tissues and blood. Also, the successful implementation is highly dependent on the interactional behavior between PU and the drug.[298] The fact that blocks of PU copolymers show decent cytocompatibility and expose cell enhancement level.[341] Additional requirements such as sufficient mechanical strength, large surface area and highly porous structure are also requisite in this case. Some of the devices are catheters, drug eluting stents (DES), drug eluting balloons (DEB) and guidewires.[67,298] For instance, as a part of cancer therapy, the Polymer Nanovehicles in the forms of polymer liposomes, vesicles, and micelles are being used.[341]

According to a thesis conducted on the drug release behavior of PUNC, the drug size has an essential impact on release behavior. In the case where lower molecular weight drug release is an option, experiment showed that, release of hydrophobic drugs was more perpetuated than the release of hydrophilic drugs. In these circumstances, PU can be engaged both as film or coatings. When applied as coatings, PU can provide with additional advantage of anti-inflammatory or anti-thrombotic agents to minimize host response.[298] The enhanced permeability and retention (EPR) effect helps the careers to increase the drug's gathering at intended areas of application.[341–344] PU has already been examined as carrier for anti-tumor drugs.[298] Figure 15 represents schematic of such a drug delivery system.

However, there are some difficulties in long term petition that includes sudden release of drugs initially, referred as the "Burst Effect." Also, once coalesce with release passage, the carrier tends to lose a large portion of the drug carried in a short time, thus causing irregular drug release.[298]

6.5.2 | Tissue engineering

In the current field of medical research, one of the most studied segments is to find a suitable alternative to counter the lack of organ transfers due to either organ loss or organ failure. The constant lack of organ donors has led to this research.[315] Also, traumatic damages and futile healing after injuries lead to damage of different body parts.[79] PU can be considered for this important application in the biomedical field, that is, tissue engineering (TE). The applications range throughout fabricate several formations for blood transfusion, skin reparation, nerve reform as well as bone growth.[67] The most effective in reconstructing organs, it is found that the most usual step is to grow cells on a scaffold to make necessary three-dimensional tissue.[346] In this regard, nanoclay that is organically deformed can also be used to develop composites.[67,347] Moreover, the recent development in the concept of bioactive composites brings together the benefits of multiple materials to improve intended actions.[312,348] TE consists of three fundamental principles, forming a template for tissue growth, which is generally a porous three-dimensional scaffold containing biomaterials, cells for tissue growth, and factors that influence growth.[79] Tissue growth might be boosted by fibroblast growth factor and scaffold to be used.[349,350]

One way can be the development of various types of organically modified clay in the form of nanoparticles

used to make composite with PU. PU/Nanoclay composites are frequently used for cell viability, proliferation, and adhesion.[67] Hydroxyapatite contains a decent amount of bone minerals, so it has good compatibility with PU for numerous TE applications.[309] Also, one of the recent development is organically modified graphene capable of being used on TE applications.[67] Furthermore, recent studies also showed potential of PPy to be used as a nanoparticle with PU as they have good electric conductivity. So the combination is a hybrid scaffold with electric conductivity and mechanical resilience. Here base material is PU with PPy nanoparticles providing network for increased electrical conductivity. Other than these, the composite also demonstrates the ability to entrap and let go bio molecules with decent control, can undergo doping that is reversible and easy to deform.[351] The concept of elastic scaffolds might be incorporated where soft tissue growth is crucial. To fulfill the requirement during tissue development elastic scaffolds, show eligibility as ductile against mechanical perturbance.[307,352]

Several organs can be repaired or reconstructed through TE. One of the most common applications is bone tissue growth. Studies have recorded more than 4.5 million reconstructive surgeries have been conducted on bone injuries.[79] The bone structure is very complex as they contain all nanostructure, sub nanostructure, micro structure, sub micro structure, macro structure in a very nonoriented and order less formation.[311,352] PUs are also significantly used in synthetic bone manufacturing. As synthetic bone provides advantages like cheap cost, availability of manufacturing material, and easy processability, Polymers are best suited for the job. Among the polymers, PU demonstrates tunable mechanical properties, lower degradation in the presence of noncytotoxic breakdown materials, better inject ability and local diffusion-controlled release of bio molecules.[353] Although the supposed elements to form a scaffold in synthetic bone tissue engineering are ceramics or metals, they are limited by their weight, cost, inability to be deformed, etc. PU and its composites, laced with shape memory characteristics, can hold a temporary formation structure and have the ability to recover as per necessity after being embedded in the bone.[79]

6.5.3 | Implant devices

Nowadays, the popularity of the usage of Pus has gone up in medical applications. Along with the other crucial applications, they show outstanding potential as medical implants with some unique properties. One of the many advantages of using PU as a biomedical implant is that

TABLE 7 Commercialization of PU based materials[372–374]

Application area	Products	Brand and manufacturers
WBF	Work jacket JKT001	Apparel supply solutions (Ireland)
	Work vest Bilbao	Dassy (Belgium)
	Work brace overall DREMBIB	Guycotten (France)
	High-visibility pants 01743 N	Industrial starter S. P.A.(Italy)
	High-visibility jacket 204x series	PLANAM Arbeitsschutz Vertriebs GmbH (Germany)
	Waterproof jacket MADOF 1	SEANA textile (Spain)
PCMs	Still in lab scale	
Chemical protection	Work coveralls Proflair, Work coveralls L0064	PPSGB (UK)
	Work apron Buckle, KARRO TITAN, TITAN, CHAINMAIL, SHOULDER STRAP, Work over sleeve	Top Dog Manufacturing (Canada)
	Work coveralls FR61, ST50, Work jacket FR79, Work pants FR78, Work glove A310, A400	Portwest Clothing Ltd (UK)
Ballistic Protection	Ballistic Nylon (PU Coated Fabrics)	DuPont (USA)
Smart application	Auxetic PU foam	Smart materials (Malta)
Medical application	Medical Film—Baymedix®, Dureflex®, Platilon®, Texin®, Desmopan®	Covestro (Germany)
	PU hose MUT series	Beswick Engineering Co, Inc. (USA)
	PU hose VULCANO 04 ET	IPL SPA (Italy)
	Compressed air tube PUT series	Laminar Technologies (India)

the di-isocyanate and di-ol groups contained by the polymer can easily be replaced with other monomers, which opens doors to a lot more variable properties per requirements. Moreover, it is also quite possible to introduce the
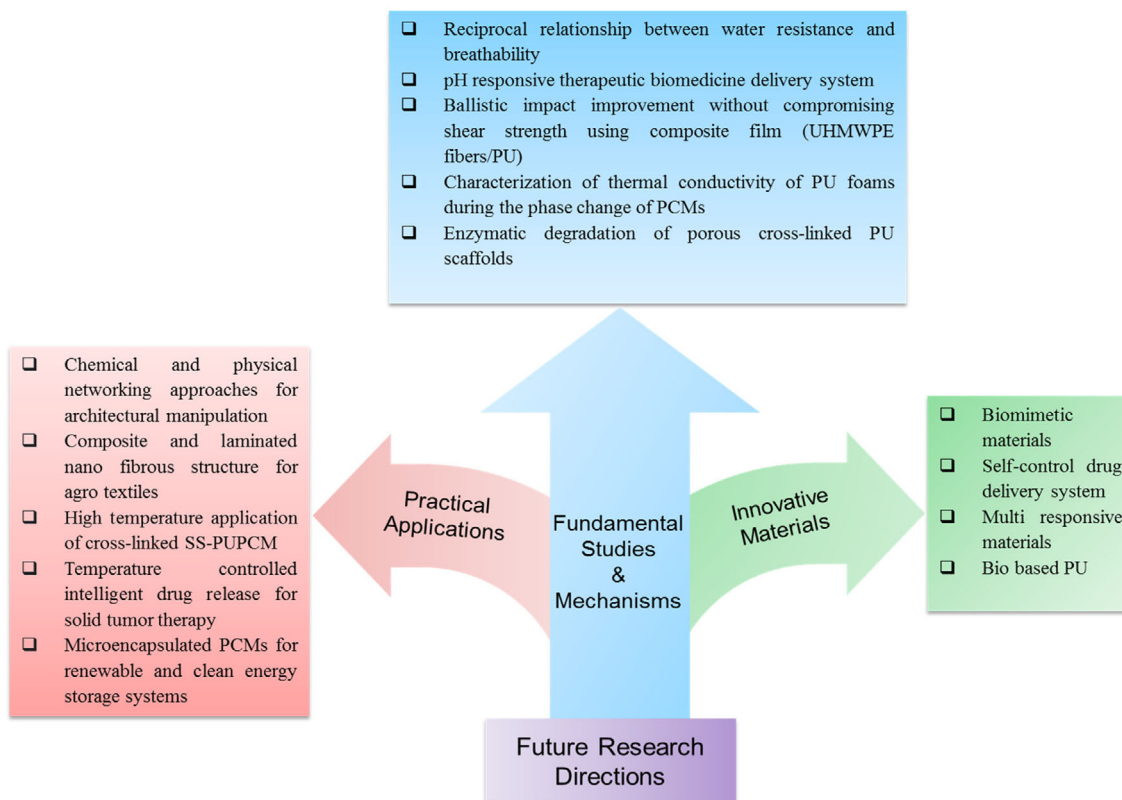
❑ Reciprocal relationship between water resistance and breathability
❑ pH responsive therapeutic biomedicine delivery system
❑ Ballistic impact improvement without compromising shear strength using composite film (UHMWPE fibers/PU)
❑ Characterization of thermal conductivity of PU foams during the phase change of PCMs
❑ Enzymatic degradation of porous cross-linked PU scaffolds

❑ Chemical and physical networking approaches for architectural manipulation
❑ Composite and laminated nano fibrous structure for agro textiles
❑ High temperature application of cross-linked SS-PUPCM
❑ Temperature controlled intelligent drug release for solid tumor therapy
❑ Microencapsulated PCMs for renewable and clean energy storage systems

Practical Applications

Fundamental Studies & Mechanisms

Innovative Materials

❑ Biomimetic materials
❑ Self-control drug delivery system
❑ Multi responsive materials
❑ Bio based PU

Future Research Directions

FIGURE 16   The graphic envisages the future research directions using PU. [Color figure can be viewed at wileyonlinelibrary.com]

widest range of morphological possibilities by commencing diamines to develop thermoplastic elastomers. Furthermore, it is also possible to choose whether to develop permanent medical implants for long term use or make disposable and degradable scaffolds for application like tissue regenerating.[354,355]

Similarly, PVC, polyester, polyethylene, or polypropylene may also be used to develop various medical implants and might give an advantage regarding costs. However, PU may still ace because they are most suitable where complex mechanical, biocompatibility and blood compatibility are concerned.[356] PUs show comparatively better hemocompatibility as they have lower thrombogenicity alongside brilliant mechanical properties.[7,357] Medical implants must be mechanically strong enough to withhold the environment in which they will be incorporated. The implants should help good proliferation properties with nontoxicity. They must be easily removable, surface modifiable, and bio absorbable with decent surface property, superior bio stability and substantially biocompatibility.[7,67]

Although PU-based materials show potentialities while used as medical implant devices, they have some challenges in encapsulating different biological entities. Encapsulation may occur either in tissue generation or as calcification while incorporated in the biological environment inside the human body. PU implants could lead to calcification while being used as heart valve. Another challenge that may appear due to contact with blood is that PU may stem blood clotting, leading to serious health issues. Also, when it degrades, potentially toxic diamines may get released inside the body, which may initiate health complexities. Even after having such challenges, scientists hope to overcome these by deforming the typical structure or adding new biocompatible monomers as composite.[355] Table 6 enlists some recent research works regarding medical applications of PU.

7 | CONCLUSION AND FUTURE OUTLOOK

Over recent years, PU polymer in the form of coating and hard material has played an increasingly important role, especially in functional textiles, high-quality materials, smart devices, biomedical fields, and other fields of great interest. Nevertheless, guided by environmental concerns and legal obligations, PUs are now being sought as a more environmentally friendly and safe alternative to other polymers.[371] Owing to PU materials' excellent properties and versatile nature, it was necessary to conduct a review on PU functional polymer focusing on its different

aspects. Hence, this review presents and explores the latest advances in PU-based materials/coatings regarding functional textiles, smart materials, and protective materials. This review also outlines the recent developments in the field of smart PU materials based on their responsive behaviors triggered by various stimuli (P^H, Temp). Additionally, we explicitly concentrated on the inclusive review of PU's different application forms concerning medical devices and activities.

So far, the trend of commercialization of PU based functional materials in these specific areas is burgeoning (Table 7). But still, most of the literature covered in this review is merely in the preliminary stage. Therefore, extensive future work is needed to investigate the performance and feasibility of the materials on the industrial scale and the primary step could be the pilot-scale production.

As discussed above, the integration of PU has made a notable impact on functional textiles, smart devices, and biomedical applications with novel and dynamic features. However, there are still much broader areas to delve further in PU than those investigated so far (Figure 16).

Though PUs have fascinating mechanical and tunable physical properties, several shortcomings, including low flexibility, and poor stability, lack of biocompatibility, carcinogenicity, have been revealed, which constrain the subsequent advances of PU for next-generation applications.

A great deal of work is being conducted to establish the PU as a suitable coating for WBF. Yet, more exploration is needed to address the reciprocal relationship between water resistance and breathability.[90] There can be more research towards diversifying the PU coated surfaces by integrating different resins with different PU amounts and chemical compositions, which might bring a remarkable breakthrough in breathable waterproof research through diverse functionalities.[93]

There have been several facile approaches to designing PU-PCM to induce desired functionalities, but some are ineffective for user satisfaction. Hence, it is also essential to explore more facile and robust techniques to avoid problems like inhomogeneity in terms of microbeads distribution,[56] high-temperature stability for application in extreme heat,[375] and low-conversion efficiency.[141] Deterioration in protective performance with PU superabsorbent polymer coating,[159] decontamination application,[168] introducing perforation in PU wearable ballistic application,[184] improvement in shear strength of PU laminated substrate,[185] enzymatic degradation of PU porous scaffolds,[138] etc. are some of the areas needs further comprehensive exploration. Regarding PU foam's pressure sensing application, the sensing range is relatively small

(0–100 N/m^2), which could be another area to explore further in-depth.[148]

Diverse research directions in medicinal and material chemistry have opened more opportunities to design/fabricate highly functional and biocompatible PU-based coatings and bulk materials in numerous smart medical applications. These include antibacterial surfaces and catheters, drug delivery vehicles, stents, surgical and wound dressings, tissue engineering scaffolds and nerve generation, cardiac patches and breast implantations.[35] However, more advanced and rapid drug delivery systems[208] and stimulant responsive therapeutic operations[217] are yet to be investigated.

PUs were carefully designed for implantable biomaterials to undergo several degradations once they contact living tissues in biomedical applications. Thus, degradation may lead to toxicity. Several studies demonstrated a general PU cytocompatibility, biocompatibility at usage and degradability. However, such studies are nascent and need to be intensified on an explicit characterization of the degradation products and their corresponding toxicity levels. Finally, future trends should focus on developing fully biobased smart materials (shape-memory PUs or covalent adaptable networks such as vitrines) with biomimetic and tunable dynamic covalent bond properties. Also, these smart structures indeed need to be investigated for advanced and adaptable properties in terms of mechanical resistance, degradability and recyclability compared to current systems.[376]

AUTHOR CONTRIBUTIONS

Partha Sikdar: Writing – original draft (equal); writing – review and editing (equal). **Tanvir Mahady Dip:** Writing – original draft (equal); writing – review and editing (equal). **Avik K. Dhar:** Writing – original draft (equal). **Maitry Bhattacharjee:** Writing – original draft (equal). **Md. Saiful Hoque:** Writing – original draft (equal). **Solaiman Bin Ali:** Writing – review and editing (equal).

CONFLICT OF INTEREST

To the best of knowledge, the authors have no potential conflicts of interest.

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.