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Recent Advances in Femtosecond Laser Fabrication: From Structures to Applications

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ABSTRACT Femtosecond laser processing is fast becoming a pervasive method for fabricating micro/nanostructures because it can be used to produce micro/nanostructures on myriads of materials with high precision and resolution, requires little control over environmental conditions, and is simple to implement. Here, we review recent developments in the use of femtosecond lasers for the fabrication of micro/nanostructures through ablation and two-photon polymerization (TPP). Moreover, the applications of some of the fabricated micro/nanostructures are also discussed. We highlight the advantages of femtosecond laser processing by explaining the underlying principles of laser ablation and TPP. We also show the use of this method to fabricate new devices with outstanding performance in several application realm, such as sensors, optical devices, microfluidic chips, and soft robotics.

INDEX TERMS Femtosecond laser, micro/nanostructures, laser ablation, two-photon polymerization (TPP).

I. INTRODUCTION

Fabricating highly efficient and versatile miniaturized devices, such as biomedical [1], chemical [2], and optical sensors [3] and flexible electronics [4], has considerably progressed in recent years. Laser processing is considered to be an appropriate method for fabricating these devices due to its highly desirable performance characteristics, such as non-contact and low environmental requirements, while producing high resolution and precision structures. However, common laser processing is not a high-precision or high-resolution technology when compared with conventional lithographically based methods. A way to improve the precision of laser processing is to increase the pulse energy and reduce the pulse width. Chirped pulse amplification theory for obtaining short and strong laser pulses was first proposed by Donna Strickland and Gerard Mourou and was demonstrated experimentally in 1985 [5].

The femtosecond laser was designed based on this theory and self-mode-locked theory [6] and has an ultrashort pulse, ultrahigh peak power, and ultrahigh electric field intensity. Since then, femtosecond laser processing began to develop rapidly. Among the various processing methods of femtosecond laser micromachining, ablation processing of the material surface (including surface modification) and two-photon polymerization (TPP) have attracted considerable attention from researchers and have many practical applications. The ultrashort laser pulse with high power provides femtosecond laser ablation with rapid and precise energy deposition onto nearly any material. Elimination of thermal diffusion and minimization of the heat affect zone, which are unique advantages of femtosecond lasers, have important implications for material processing in numerous applications, such as sensors, optical devices, microfluidic chips, and flexible

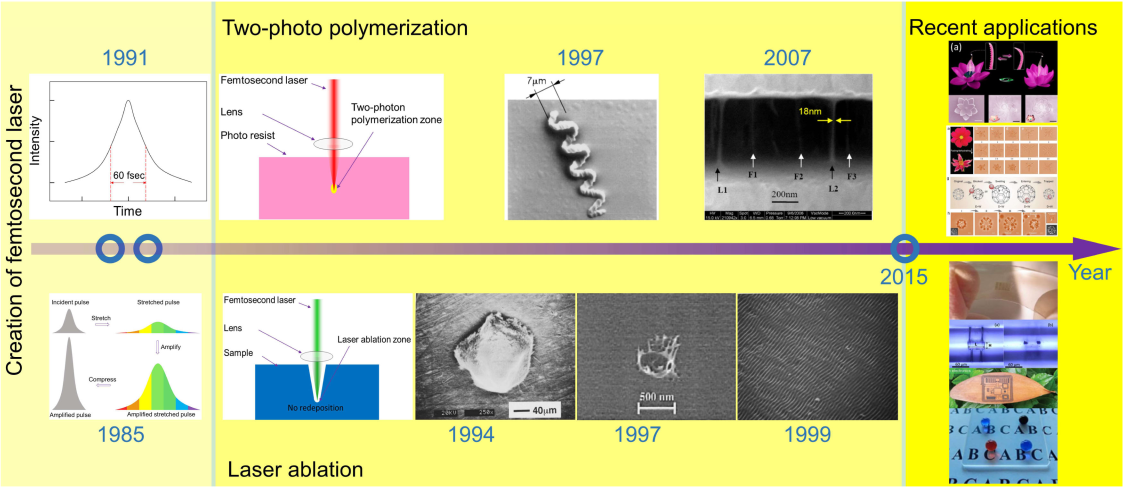

FIGURE 1. The historical development of femtosecond lasers. The femtosecond laser is based on two important technologies, one of which is the Chirped Pulse Amplification proposed in 1985, the other of which is the self-mode-locked laser; the first laser pulse with a femtosecond scale pulse width of 60fs was constructed in 1991. From then on, applications of femtosecond lasers developed quickly. The two major applications of femtosecond lasers are two-photon polymerization and ablation. In 1997, the first 3D structure was fabricated by TPP (with permission from © The Optical Society), and the resolution of TPP broke through the optical diffraction limit for the first time in 2001. In 2007, a structure with a resolution of 1/43 wavelength was formed (with the permission from © AIP Publishing). Femtosecond laser ablation was used on polymers in 1994 (with the permission from © AIP Publishing), the advantages of femtosecond laser processing were demonstrated in 1996, and submicron structures on metals and semiconductors were fabricated by femtosecond laser in 1997 (with permission from © Elsevier); laser induced periodic structure was observed in experiments in 1999 (with the permission from © AIP Publishing).

electronic devices. Owing to the combination of high optical performance systems and the threshold effect of femtosecond lasers, many types of micro/nanostructures with ultrahigh resolution unachievable with conventional laser processing techniques have been fabricated on different material surfaces by femtosecond laser ablation. On the other hand, TPP is another important application of femtosecond lasers and is distinct from conventional photopolymerization. It allows for the fabrication of 3D micro/nanostructures with resolutions smaller than the diffraction limit. Thus, femtosecond-laser-based TPP has become one of the most active areas of research recently. The historical development of femtosecond lasers processing by ablation and TPP is summarized in Fig. 1. TPP could be applied to process 3D structures in micro/nanoscale [7], [8]. Ablation, known as "cold machining" [9], could ablate sharp, well-defined patterns in polymers [10], metals [11], and other solid targets with special structures on micro/nanoscale, including laser-induced periodic structure [12]. The micro/nanostructures fabricated by femtosecond lasers have shown special properties that could be applied in new sensors, biological treatment, and various industries. In 2001, Kawata and Sun fabricated nanoscale sculptures of a bull with a spatial resolution of 120 nm using femtosecond two-photon photopolymerization, which highlights the potential of this technique for fabricating nanorobots within blood vessels [13]. In fact, various micro/nanoscale functional devices with complex 3D structures have been fabricated by femtosecond laser TPP [14]–[16]. In bionics, the structures in nature such as the surface of flowers [17] and artificial muscles [18] can also be processed by femtosecond lasers. In 2019, Saha *et al.* demonstrated scalable sub-micrometer additive manufacturing by spatially and temporally focusing a femtosecond laser to realize projection-based layer-by-layer parallelization; this way increases the throughput by up to three orders of magnitude at a resolution of 175 nm [19] and making femtosecond lasers commercially viable. Femtosecond lasers are also being used to fabricate soft robots, intelligent sensors, and arrays of micro-optical devices, as well as in high-throughput surface modification methods, bionics, and other fields because of their high-resolution 3D processing capability. Thus, femtosecond laser processing has become one of the most versatile micro/nanofabrication technologies.

In this review, we summarize the recent advances in applications that benefit from improved structure performance by utilizing femtosecond laser processing. The applications are divided into two categories by processing methods, namely, ablation and TPP. In the ablation section, the applications are focused on new sensors, optical devices, microfluidic chips, flexible electronic devices, and surface modification. In the section concerning TPP, the applications are focused on magnetically driven microrobots and soft robotics. The advantages and applications of femtosecond laser machining structures will be discussed by reporting the advances of these research fields.

II. ABLATION USING FEMTOSECOND LASER

A. NONLINEAR ABSORPTION OF FEMTOSECOND LASER

The characteristics of the femtosecond laser are related to its unique reaction with materials. The interaction of light with materials is generally linear, which means that the transmittance and absorptivity of light do not change with its intensity. However, in the case of the femtosecond laser, the pulse duration is of the order of femtoseconds and thus much shorter than the time required by the excited electrons to release energy. As a result, a heat diffusion zone is not formed. Meanwhile, the electric field generated because of the ultrahigh peak power of the femtosecond laser pulse is comparable to the coulomb-bound field within the atom. Hence, the excitation of the electrons cannot be explained by conventional linear resonance absorption, and the third-order nonlinear-effect-based interactions between the laser and the material are dominant. Thus, the ablation finishes without the re-cooling of the material, which results in a smooth surface and high-precision ablation. Femtosecond laser fabrication has three important advantages, namely, no heat diffusion, sub-micrometer scale resolution, and direct 3D fabrication, compared with conventional laser fabrication. Furthermore, the multiphoton absorption and ionization threshold only depends on the atomic characteristics of the material being processed. This condition makes femtosecond laser fabrication suitable for use with nearly any material. Moreover, femtosecond laser processing allows maskless subtractive and additive material manufacturing of complex 3D micro/nanostructures with high resolution, which enables it to have broader application prospects in many research and application fields, such as precision manufacturing, surface modification, and biomedical science and engineering.

B. FEMTOSECOND LASER FABRICATION OF MICRO/NANOSTRUCTURES

Different micro/nanostructures may provide divergent functionalities and may be used as valuable surface structures for research and applications. For example, random micro/nano hierarchical structures can be used to modify surface properties, e.g., controlling surface hydrophobicity, and specially designed micro/nano structures can provide unique optical and surface-friction functionalities. Technologies have been developed to fabricate micro/nanostructures on the desired surfaces for mimicking the abovementioned functionalities. Femtosecond laser ablation could contribute to fabricating micro/nanostructures of high dimensional resolution and spatial precision [20]–[22]. The technology has been proven effective for enhancing the machining precision and the realization of large-area ablation without requiring multiple fabrication steps necessary for micro-/nano-lithographic technologies. These advantages may allow the fabrication of functional micro/nanostructures with less processing flow while providing myriads of applications. For example, surface structures formed using femtosecond laser ablation could be used for bubble manipulation [23]. A femtosecond laser was also used to fabricate slippery periodic microgrooves that allow for the anisotropic sliding of bubbles underwater [24]. The method has also been used to fabricate lotus-leaf-like surfaces for aeronautical applications [25] and double hierarchical surfaces that exhibit high fog-collection efficiency [26]. Specific and uniform micro/nanostructures can also be fabricated. For instance, the laser induced periodic surface structure (LIPSS) technique has been used in the fields of tribology, wettability analysis, and mechanics because it allows one to vary the polarization of the laser and its scan speed, pulse energy, and pulse number. This technology would allow for the fabrication of LIPSS suitable for use in various applications [27]–[29]. In addition, micro/nanostructures are used widely for numerous purposes in the field of optics. Nanostructures manufactured using femtosecond lasers could allow for polarization-controlled image switching [30]. Microlasers can be created by performing femtosecond laser ablation on halide-based perovskites to improve the Q-factor [31]. In summary, micro/nanostructures can be fabricated using femtosecond lasers on the surfaces of various materials, and such structures have much potential for use in a wide range of applications employed across multiple scientific fields.

C. MANUFACTURING AND APPLICATIONS OF SENSORS

Improvements in the performances of sensors are tied to the development of new high-performance materials and innovative structures. A trend toward miniaturization, digitization, systemization, networking, and multifunctionality exists. However, progress in these areas depends on the discovery of novel materials and improvements in ability to fabricate structures with greater accuracy and ease. Femtosecond lasers can be used to produce new high-performance sensors because they allow for constructing functional micro/nanostructures commodiously on various materials with high precision, as shown in Table 1. In 2014, researchers fabricated ozone sensors on flexible substrates using two techniques, namely, photolithography and laser ablation [32], as shown in Fig. 2(a), (c). In 2016, Krasnok's group used a femtosecond laser to reshape a metal to produce an optical device [33]. Femtosecond lasers can be used to fabricate field-effect transistors based on reduced graphene oxide (rGO) [34], as shown in Fig. 2(b), (f), because of their photoreduction characteristics. New pressure sensors can be processed using a flip chip fabricated with a femtosecond laser, as shown in Fig. 2(c), (g) [35]. Researchers also used femtosecond laser direct writing to synthesize conductive rGO/polydimethylsiloxane composite films, which were subsequently used to produce a voice recognition sensor with self-cleaning and anti-interference abilities [36], as shown in Fig. 2(d), (h). Moreover, femtosecond lasers have been used to fabricate high-precision structuring of the polyimide layer for curved aircraft structures [37]. A fiber Bragg grating structure could be processed in the core of the fiber and were able to embed Fourier optics and non-diffractive beam devices on the end face of the fiber [38]. In addition, the Bragg grating was inscribed with a near-infrared femtosecond laser through point irradiation to modulate the refractive index of the material [39].

Sensors based on novel materials and structures are being developed for use in various fields and applications, such as industrial automation, personalized medicine, signal measurements, beam steering, optical switching, and harmonic generation. The characteristics of femtosecond lasers will ensure that they play a significant role in the development of these sensors.

TABLE 1. **Sensors Processed by Femtosecond Laser**

Material	Application	The performance parameters	Reference
Zno、Ti and Pt films	Ozone gas sensors	Range of detection from 5 ppb to 300 ppb R/R0 = 26 ± 2.3； work temperature below 200℃	Ref.32
Au	Selective reshaping of the metal components	The height of the Si nanocones was ≈200 nm; the period was ≈600 nm; the total size of each array was 17 μm × 17 μm	Ref.33
GO	Oxide FET	Ion/Ioff ratio of 2.04; hole mobility of 0.27 cm^2/Vs;	Ref.34
GO	Self-cleaning acoustic sensor	Contact angle = 162° and sliding angle = 1° ultrahigh gauge factor (GF) of 8699; ultralow detection limit of ε = 0.000064%; high SNRs (40-50 dB)	Ref.36
Glass	Pressure reference cavity	A sensitivity of 10 mV/V/bar; stable operation up to 7 bar absolute pressure	Ref.35
Silica and polymer optical fibres	Fibre Bragg grating	Chirp >25nm/cm an amplitude modulation with period of 662 μm	Ref.38
Epoxy-based negative photoresist material EpoCore	Fibre sensor	Sensitivity for the state variables temperature, humidity, and strain to be 45 pm/K, 19 pm/%, and 0.26 pm/μ	Ref.39

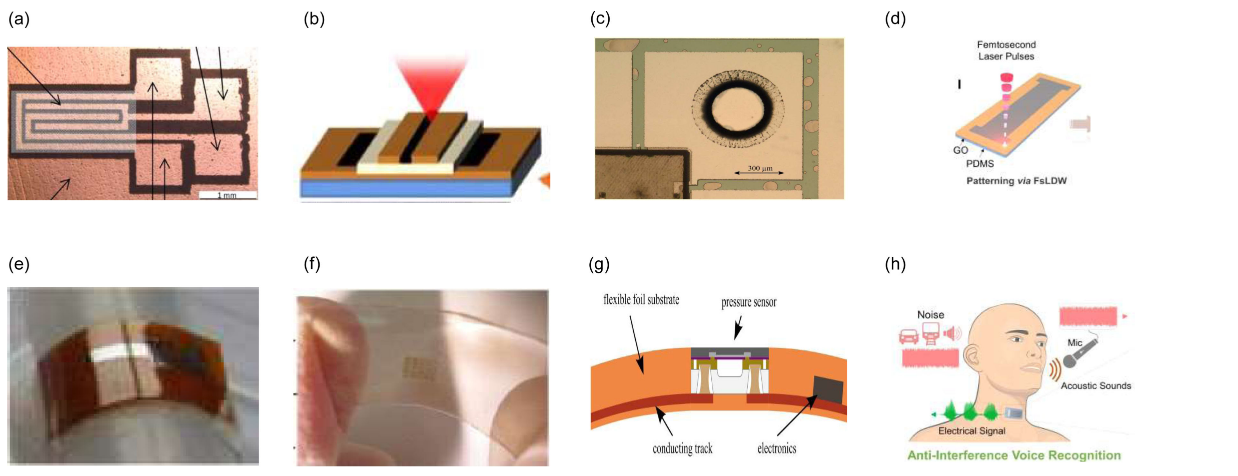

FIGURE 2. **(a) Photograph of an ozone sensor based on laser ablation technology (with permission from © Elsevier). (b) Process for manufacturing full-carbon FET based on a photoreduction mechanism (with permission from © IEEE). (c) Structure of a new type of pressure sensor made using a flip chip fabricated with a femtosecond laser (with permission from © IEEE). (d) Manufacturing principle of graphene-based self-cleaning voice sensor (with permission from © American Chemistry Society). (e) Photograph of the ozone sensor depicted in (a). (f) Photograph of FET depicted in (b). (g) Photograph of sensor depicted in (c). (h) Application of voice sensor shown in (d).**

TABLE 2. Optical Devices Processed by Femtosecond Laser

Material	Application	The performance parameters	Reference
Glass film	Halide-perovskite microlasers	At room temperature in a broad spectral range (550-800 nm) with Q-factors up to 5500;	Ref.31
Coupled spherical silver nanoparticles	Plasmon resonances	Electric field enhancement increase by a factor ~3 (for the symmetric mode) and ~10 (for the antisymmetric mode)	Ref.41
Single-mode optical fiber: SMF-28e	Fluidic cavity with inlets/outlets	Refractive index (RI) sensitivity was estimated to be 1135.7 nm/RIU in the tested RI range	Ref.42
Photoresist	Compound lens in a supporting shell	A large field of view (FOV) of 80° resolutions of up to 500 lp/mm	Ref.43
LiNbO3	An integrated optical coupler	28.5 μm gap corresponds to a ratio of 40/60 of power splittings; the left mode has ~40% and the right mode ~60%;	Ref.44
An 80 nm a-Si:H film	Magnetic and electric Mie-type resonances	The crystallite size is larger than ~20 nm the resulting array of nanoparticles has a period of about 0.9 μm ;	Ref.45
Flat fibre (hollow silica)	Different key optical structures	The depth of ring resonator is 9um; the width of line is 10um;	Ref.46
Multi-core fiber	Multi-core fiber Bragg grating (MC-FBG)	Four 3rd order FBGs with Bragg resonances λB = 1541.01 ± 0.02, 1547.82 ± 0.02, 1532.66 ± 0.02, and 1537.42 ± 0.02 nm and extinction ratios of 13.97 ± 0.4, 16.02 ± 0.4, 10.08 ± 0.4 and 13.40 ± 0.4 dB	Ref.47
50-nm thick silver films	Down-scaling of sensing elements for vis-IR surface-enhanced spectroscopies	The evaluated total enhancement factors (~300–400 times) are comparable to the previously reported values ~102–103	Ref.48

D. MANUFACTURING AND APPLICATIONS OF OPTICAL DEVICES

Optical devices have several advantages over electronic devices, such as no interference from electromagnetic radiation and wider applicability. In general, optical devices require extreme precision because the incident light can only interact stably with a substrate when the surface structures of the substrate are of a specific size. For instance, resonant metallic nanoparticles can induce plasmon resonances to localize and enhance the optical field and are used in several practical applications, including electromagnetic field enhancement and sensing [40], optical switching [41], and microlaser emissions [31]. However, conventional manufacturing methods have difficulty ensuring that the devices will be of the desired size and fabricated at the desired position. Thus, new high-precision methods are required. Femtosecond laser processing induces a smaller heat-affected zone and thus improves the precision of the fabrication, and femtosecond lasers can be used to fabricate complex structures with high precision at any location on the sample. These characteristics have been applied to develop new optical devices, as shown in Table 2. For example, an all-in-fiber optofluidic sensor has been fabricated [42], as shown in Fig. 3(a)-(c), using femtosecond-laser-assisted chemical etching; meanwhile, femtosecond two-photon direct laser writing (DLW) has been used to manufacture multi-lens objectives at the top of optical fiber [43]. Furthermore, evanescent wave couplers have been fabricated using femtosecond laser micromachining [44], and optical nanoresonators could be developed using femtosecond laser ablation [45]. A photonic integration platform could be built using femtosecond laser technology [46], and fiber Bragg gratings could be raised in a multicore fiber using the point-by-point inscription technique [47]. In addition, mask-less-patterned thin plasmonic films were realized using multiplexed femtosecond

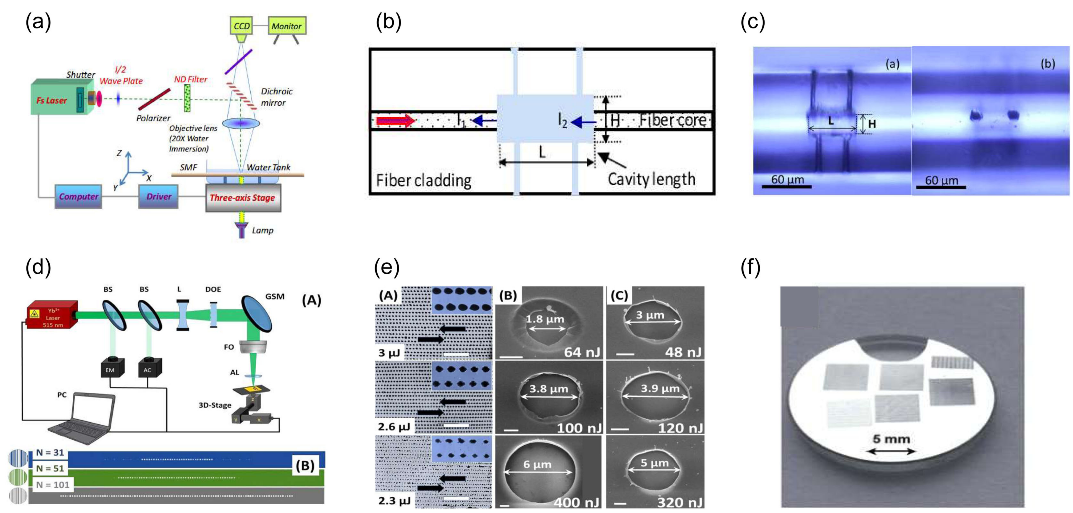

FIGURE 3. **(a) Femtosecond-laser-assisted chemical etching system. (b) Schematic diagram of horizontal and vertical microchannel structures fabricated in optical fibers. (c) Optical images of horizontal and vertical microchannel structures fabricated in optical fibers (with permission from © The Optical Society). (d) Schematic diagram of sub-megahertz femtosecond laser system with high multiplexing and sharp focus. (e) Microwell array. (f) Infrared sensor with integrated microwell grating (with permission from © Elsevier).**

TABLE 3. **Microfluidic Chips Processed by Femtosecond Laser**

Material	Application	The performance parameters	Reference
Single mode optical fiber (SMF)	Form refractive index structures such as optical waveguides and gratings	A linear strain response of 2.7 ± 0.5 pm $\mu\varepsilon^{-1}$ measure the temperature-dependent refractive index of water over a 30 ° C to 92 ° C temperature range	Ref.54
Silicon wafer	Creating buried microfluidics by opening the bottom of a parylene-coated channel		Ref.55
Thick silica glass	3D microchannels	High aspect ratio was ~30;	Ref.56
Glass	Multi-cellular culture systems	The detection limit can reach 10^{-9} M	Ref.57
Fused silica	Fabrication of complex three-dimensional microcoils	30 µm in diameter, 13 mm in length, aspect ratio more than 400:1	Ref.58

laser pulses [48], as shown in Fig. 3(d)–(f). In conclusion, femtosecond laser technology can be used to manufacture optical devices with different functions because of its universality to materials and its processing precision that can reach the wavelength level.

E. MANUFACTURING AND APPLICATIONS OF MICROFLUIDIC CHIPS

Microfluidic platforms are being increasingly used in chemical analysis [49], chemical synthesis [50], biological cell analysis [51], biological transformations [52], and optical detection [53]. However, fabricating high-performance, microscale, and multifunctional microfluidic chips remains a challenge. Femtosecond lasers can be used to overcome this challenge because of their ability to fabricate 3D structures on different kinds of materials, as shown in Table 3. In 2014, Haque *et al.* used femtosecond laser writing and femtosecond laser irradiation followed by chemical etching to write 3D optical circuits and microfluidic systems in single-mode optical fiber and coreless fiber to fabricate multifunctional microfluidic chips [54], as shown in Fig. 4(b), (e). Researchers have also proposed an advanced method for manufacturing microfluidic structures comprising of channels and inputs/outputs buried within a silicon substrate based on single-level lithography. Thus, they created an observation window within an opaque silicon substrate for observing the flow state within the channels [55], as shown in Fig. 4(c), (f). Cheng's group used simultaneous spatiotemporal focusing to

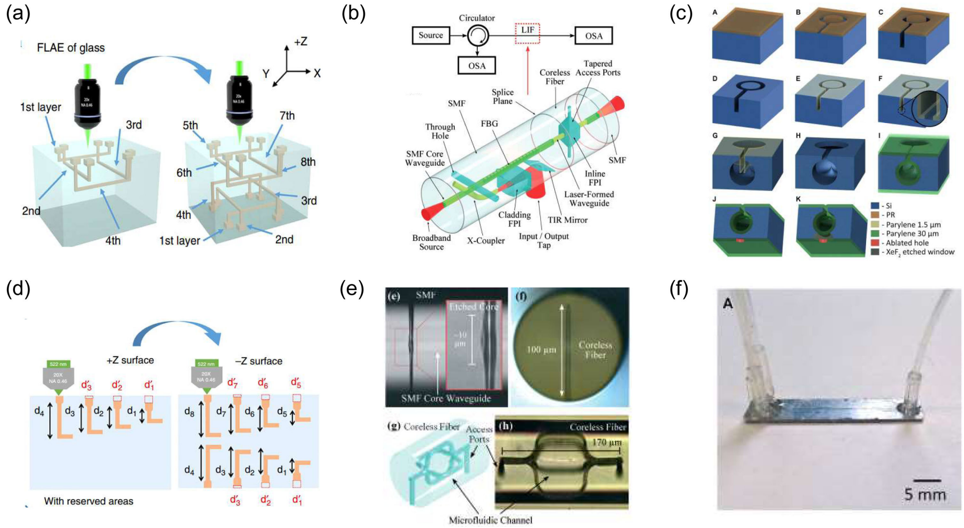

FIGURE 4. (a) Microfluidic sensor fabricated using femtosecond-laser-based direct writing technology (with permission from © Royal Society of Chemistry). (b) Schematic of the process of femtosecond laser writing and FLICE in SMF and coreless fiber to write a 3D optical circuit and microfluidic system (with permission from © Royal Society of Chemistry). (c) Structure of microfluidic chip manufactured using single-level lithography (with permission from © Springer-Verlag GmbH Germany). (d) Images of self-driven microfluidic chip depicted in (a). (e) Images of microfluidic chip depicted in (b). (f) Optical image of microfluidic chip depicted in (c) during operation.

fabricate 3D microchannels on thick quartz glass, which is a method that avoids the problem of nonlinear self-focusing encountered in the case of conventional focusing methods. This technology is expected to be applicable in the case of other transparent materials as well, such as polymers and crystals [56]. With respect to the manufacturing of detection chips, femtosecond lasers can be used to readily fabricate periodic surface structures to produce self-driven microfluidic surface-enhanced Raman spectroscopy detection chips [57], as shown in Fig. 4(a), (d). In terms of microcoils, Chen's team proposed the use of femtosecond laser to fabricate complex three-dimensional microcoils in fused silica [58] and achieved a high-level integration [59]. Femtosecond-laser based micro/nanomanufacturing technology is capable of high-precision 3D processing and can be used for flexible positioning in various types of nonplanar microfluidic chips and thus the integration of a large number of functional components. This technology will aid the development of future microfluidic chips that exhibit even better performance and a greater number of functionalities.

F. MANUFACTURING AND APPLICATIONS OF FLEXIBLE ELECTRONIC DEVICES

Femtosecond lasers can directly create 3D micro/nanostructures with very high precision. They can also process materials with high speed and efficiency, which are difficult to achieve with conventional micro/nanofabrication technologies such as chemical or plasma etching. The surface functionalization of materials through the fabrication of micro/nanostructures can yield devices that exhibit unique properties, such as the ability to enhance electrical and light signals. Such devices have wide applicability in wearable electronics [60], infrared sensors [61], and portable devices. Fabrication techniques based on femtosecond lasers have been studied by researchers to address the various challenges encountered in the manufacturing of high-performance, versatile, and durable devices, as shown in Table 4. Flexible photodetectors consisting of micro/nanostructures were fabricated by femtosecond laser irradiation on silicon-based materials in an SF6 atmosphere [62]. The devices exhibited high responsiveness over a wide range of wavelengths owing to their high light absorption efficiency, which was attributable to the etched micro/nanostructures, as shown in Fig. 5(b), (i). Furthermore, graphene oxide (GO) can be reduced using a femtosecond laser for use as conductive electrodes. Thin films of GO produced in this manner can be used as sensing elements. In addition, a noncontact electronic skin fabricated using a femtosecond laser showed high sensitivity and fast response and recovery rates [63], [64], as shown in Fig. 5(a), (d), (h). Finally, soft substrates have been adopted to fabricate all-graphene-based flexible sensing devices using femtosecond laser direct writing for improving the adhesion between graphene and underlying substrates.

For example, polyimide sheets were irradiated with a femtosecond laser beam, and a carbonized structure was formed [65]. The fabricated structure was then functionalized with Au nanoparticles through surface plating and low-temperature sintering, as shown in Fig. 5(g). Subsequently, hydrated GO and a chloroauric acid nanocomposite were reduced by femtosecond laser direct writing to form in-plane micro-supercapacitors and three-layer supercapacitors with capacitances as high as 37.5 mF/cm^2 [66]. Moreover, highly conductive graphene can be formed from natural

TABLE 4. Flexible Electronic Devices Processed by Femtosecond Laser

Material	Application	The performance parameters	Reference
Sulfur-Hyperdoped Ultrathin Silicon	Free-Standing Flexible Photodetectors	The responsivity remains above 10 A/W from 440 to 1120 nm the spectral range is broadened to 1200 nm the rise and fall times are 68 and 172 μs, respectively;	Ref.62
GO	Noncontact Electronic Skin	The measured impedance operating at 1 kHz shows nonobvious fluctuation (less than 0.1%) at each RH level for 30 days high-spatial-resolution sensing capability: can response to sharp pencile tip	Ref.63
GO	Strain sensor	Linear current-voltage characteristics upon applying different strains in the range of 0-14% the GF was calculated to be 58.2 within 14%.;	Ref.64
PI	High performance hybrid supercapacitors	Pecific capacitance is about 20 mF/cm^2 at 10mV/s ;	Ref.65
GO	In-plane micro-supercapacitors (MSCs)	Large specific capacitances of 0.77 mF cm^{-2} (17.2 F/cm^3 for volumetric capacitance) at 1 V/s, and 0.46 mF/cm^2 (10.2 F/cm^3) at 100 V/s;	Ref.66
Natural woods and leaves	Green graphene electronic components of electrical interconnects, flexible temperature sensors, and energy-storing pseudocapacitors	The lowest sheet resistance of LIG is around 10 Ω/sq ; the response and recovery times were 7.0 and 6.2 s, respectively;	Ref.67
Silver (Ag) nanoparticle-based ink, PVP, PI	Interlayer interconnection between multi-layer circuits	The pattern with a conductivity of 6.21 × 105 S/cm;an rms roughness of 7.09 ± 0.5 nm on PI without damage;the conductivity of bulk silver of 6.3 × 105 S/cm;the pattern on the PVP layer had slightly lower conductivity of 4.25 × 105 S/cm.	Ref.68
Bacterial Cellulose-Based Paper	High-resolution patterns of the conductive poly(p-phenylene vinylene)	Transferring PPV with a resolution on the order of 10 μm; The transferred patterns display a width of 12 μm, height of approximately 630 nm, and surface roughness (Ra) of 51 nm.	Ref.69

wood and leaves through one-step irradiation with femtosecond laser pulses under ambient conditions [67], as shown in Fig. 5(k). This technology should aid the development of green electronic components and devices, such as electrical interconnects, flexible sensors, and energy-harvesting devices. In another study, a silver nanoparticle-based ink was printed on a flexible substrate through a solution-processable patterning method to create multilayered electrical circuits using laser sintering combined with laser ablation [68], as shown in Fig. 5(c), (e),(j). Cellulose-based flexible electronics are being explored to improve the biocompatibility of electronic devices. With such devices in mind, the phenomenon of forward transfer induced by a femtosecond laser can be used to create patterns of conductive poly(p-phenylenevinylene) on a substrate of bacterial cellulose [69]. The thus-fabricated patterns exhibited excellent electrical properties, and the resolution was as high as 10 μm without material degradation, as shown in Fig. 5(f). Thus, the outstanding characteristics of femtosecond laser processing allow for the manufacturing of novel, versatile, and flexible electronic devices.

G. PRINCIPLE AND APPLICATIONS OF SURFACE MODIFICATION

In recent years, the modification and functionalization of material surfaces is an effective approach for endowing materials with specific functionalities and thus improving their performance and that of the devices based on them. Conventional surface modification techniques include chemical etching, surface deposition, optical lithography, and hot embossing. However, these procedures are usually complex and lack versatility and durability. They also cannot be used with biocompatible materials, such as titanium and quartz glass. By contrast, femtosecond lasers can be used for the micro/nanoprocessing [76] of functional surfaces, as shown in Fig. 6(a) and Table 5. The interfacial wettability can be controlled by fabricating micro/nanostructures with specific

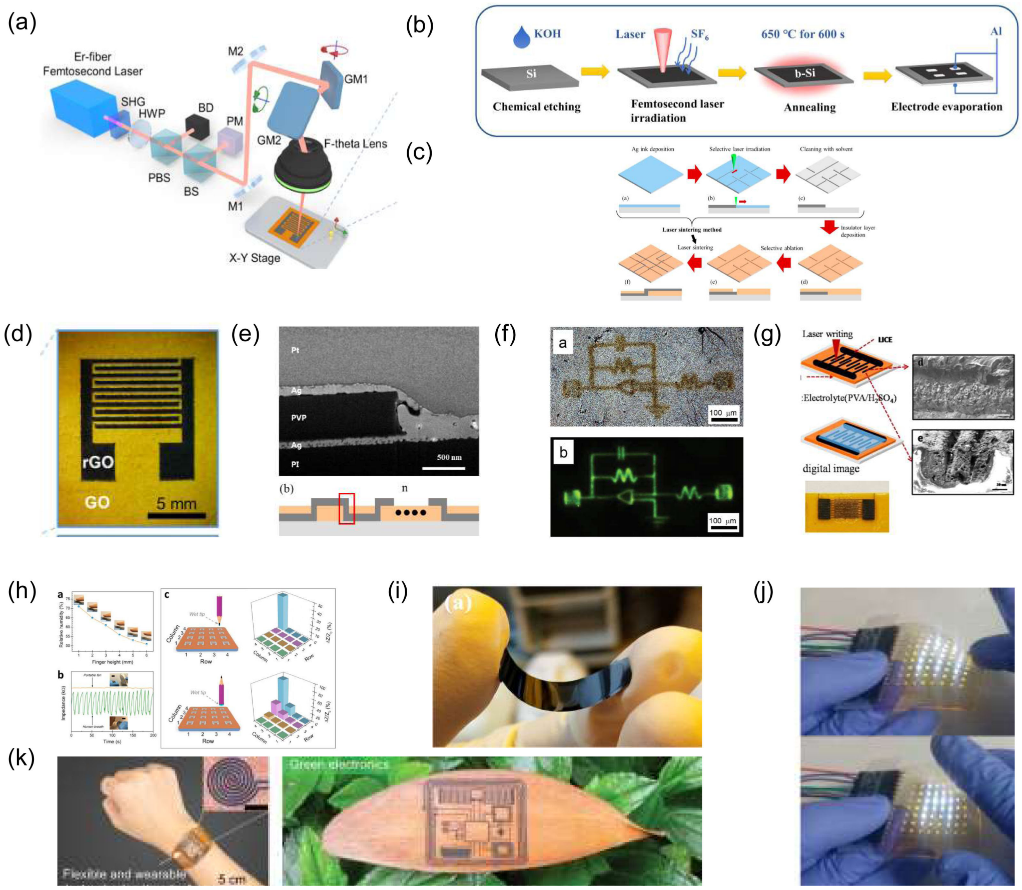

FIGURE 5. **(a) Schematic of femtosecond laser system (with permission from © American Society of Chemistry). (b) Femtosecond laser irradiation of silicon-based materials in SF_6 atmosphere (with permission from © American Society of Chemistry). (c) Fabrication of multilayered electrical circuits (with permission from © MDPI). (d) Interdigitated electrodes. (e) Structure of multilayered electrical circuits. (f) Conductive patterns on bacterial cellulose substrate (with permission from © Wiley-VCH Verlag). (g) Laser-induced carbonization structures (with permission from © Laser Institute of American). (h) Testing spatial resolution of electronic skin. (i), (j) Characterizing curing characteristics. (k) Testing conductive properties (with permission from © Wiley-VCH Verlag).**

shapes. Femtosecond laser processing is not only mask free but also highly suitable for rapid and large-scale surface functionalization. Studies have shown that micro/nanostructured surfaces formed using femtosecond laser processing can be used for the separation of oil and water [70], directional transmission [71], and fog collection [76], as shown in Fig. 6(e), (h), and (i), respectively. For example, rough microstructures were created on a polytetrafluoroethylene sheet using a femtosecond laser. The surface exhibited superhydrophobicity and the ability to prevent water adhesion [72]. Compared with micro/nanostructured surfaces formed through chemical functionalization, those formed using femtosecond laser processing were more stable and for longer periods, even after being subjected to strong acids and high temperatures. In nature, the shark skin and lotus leaves can present superhydrophobicity in air. Inspired by shark skin and lotus leaves, a hierarchical rough structure was formed at the micro/nanoscale on silicon and polydimethylsiloxane surfaces, which resulted in superaerophobicity under wet conditions and superhydrophobicity in air [73], as shown in Fig. 6(b). Using femtosecond laser ablation, porous microstructures could be fabricated on the surfaces of polymers, such as polyamides, polyethylene, and polycarbonates. By combining this technique with chemical treatment, the surface-displayed improved slipping characteristics [74], as shown in Fig. 6(g). In addition, micropit arrays were fabricated on glass surfaces using femtosecond laser direct writing to increase the local roughness and endow superhydrophobicity [75], as shown in Fig. 6(c),(d). A femtosecond laser, which was based on biomimetic designs inspired by the characteristics of insects, such as the beetle's elytra, was used to create a hierarchical structure on a substrate, which resulted in a contact angle of less than 10° [76]. A Teflon-like polymer was patterned on the surface to increase the contact angle to as much as 150°. The wetting surface simultaneously exhibited superhydrophobicity and superhydrophilicity and could be used for fog collection with high efficiency. Instead of coating antibacterial materials, controlling the surface wettability by using a femtosecond laser to fabricate micro/nanostructures is an alternative method for preventing bacterial adhesion and the formation of biofilms [77] and for collecting bubbles [78], as shown in Fig. 6(f).

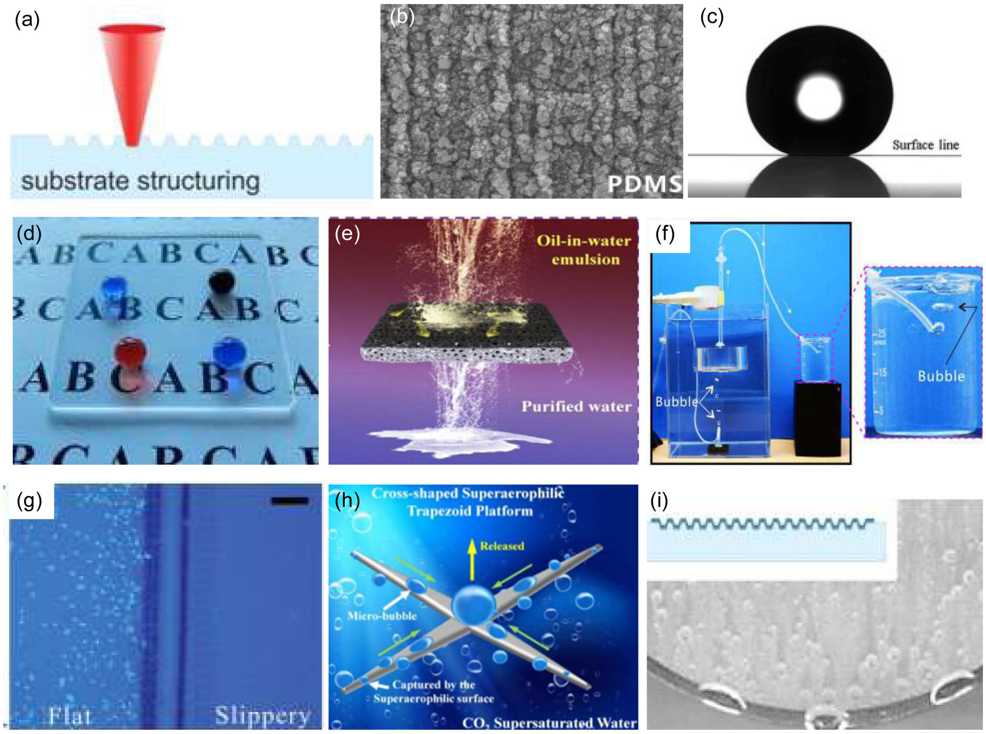

FIGURE 6. **Femtosecond-laser-induced surface modification and functionalization. (a) Schematic of femtosecond laser manufacturing process (with permission from © American Society of Chemistry). (b) SEM images of surface structures (with permission from © American Society of Chemistry). (c) Schematic showing wettability measurements (with permission from © Royal Society of Chemistry). (d) Surface modification of transparent materials. (e) Separating oil–water mixtures (with permission from © Royal Society of Chemistry). (f) Collection of bubbles (with permission from © Royal Society of Chemistry). (g) Change in biocompatibility (with permission from © Wiley-VCH Verlag). (h) Directional transport of air bubbles (with permission from © Elsevier). (i) Fog collection ability.**

Asymmetrical structures with hydrophobic and superhydrophobic properties have been fabricated for the transportation of gas bubbles along a desired direction [79]. Surfaces that exhibit biocompatibility and antibacterial properties can be achieved through the one-step fabrication of nanofeatures [80]. Interestingly, periodic surface structures formed on flexible materials using a laser beam result in color changes when the materials are stretched or bent by a mechanical force [81].

III. FEMTOSECOND LASER TPP

A. PRINCIPLE OF TPP

TPP is another main method for the fabrication of micro/nanostructures by femtosecond lasers [82]. It has two primary features. The first is that TPP only occurs at the focal point of the laser [83]. Thus, it is suitable for fabricating 3D micro/nanostructures. The second is that the volume of the reaction point is the cube of the laser wavelength, which ensures high precision. TPP consists of the following steps. First, the focusing of the femtosecond laser beam onto the transparent photosensitive polymer material triggers the photopolymerization reaction of the monomers and oligomers owing to two-photon absorption [84], which causes the polymer material to solidify. 3D micro/nanostructures are then formed after the removal of the unsolidified material using a specific solvent. For TPP of transparent and flexible materials, single-photon absorption between the femtosecond laser and the material is prevented by the ultrashort laser pulse. This way ensures that the material is not damaged or modified before the femtosecond laser propagates to the focal point. TPP using a femtosecond laser is a maskless and simple process.

TPP can be used to fabricate nearly any 3D micromechanism by selecting the appropriate 3D transfer platform and optical shutter. Furthermore, the resolution can be reduced to the sub-diffraction limit in the transverse and the axial directions by using a high-numerical-aperture objective lens (NA = 1.35–1.40) and the spatiotemporal focusing technique. Unique structures can be fabricated quickly by using special optical devices [85], [86]. And, structures could be controlled to move or assembled under special circumstances [87]. At present, 20 nm-wide suspension structures can be fabricated by TPP using a femtosecond laser with a wavelength of 800 nm. These advantages make femtosecond laser TPP highly suited for use with novel materials, magnetically driven robots, soft robots, and in other fields.

B. DEVELOPMENT IN RESOLUTION AND BIOCOMPATIBILITY OF TWO/MULTIPHOTON POLYMERIZATION

1) INCREASING RESOLUTION

Given the rapid pace of developments in materials science, new materials are continually being discovered for use in various applications, such as biomedical engineering, exoskeleton

TABLE 5. Surfaces Modification Processed by Femtosecond Laser

Material	Center wavelength	Pulse duration	Repetition	Laser power	Scanning speed	Spacing	Surface performance	Application	Reference
Ti foam	1030 nm	250 fs	75 kHz	8 W	2 m/s	10 μm	Superhydrophilic in air superoleophobic in water	Separation of oil and water	Ref.70
PTFE	1030 nm	250 fs	75 kHz	10 W	3 m/s	10 μm	Superhydrophobicity in air superaerophilicity in water	Unidirectional transportation of bubble	Ref.71
PTFE	800 nm	50 fs	1 kHz	20 mW	5 mm/s	5 μm	Superhydrophobicity	Separation of oil and water	Ref.72
PDMS	800 nm	50 fs	1 kHz	15 mW	2 mm/s	2 μm	Superhydrophobic in air superaerophilic in water	Control of bubbles' behavior	Ref.73
PET	800 nm	50 fs	1 kHz	30 mW	4 mm/s	4 μm	Superoleophilic and hydrophilicity is amplified	Liquid repellence and anti-cell proliferation	Ref.74
Silica glass	1030 nm	800 fs	200 kHz	4 W		30 μm	Superhydrophobicity and transparency	Large area self-cleaning glass	Ref.75
Borosilicate glass	520 nm	380 fs	200 kHz	6.2 J/cm²		16 μm	Superhydrophobic	Fog-collection	Ref.76
Titanium alloy	1030 nm	500 fs	1 kHz	0.3 J/cm²	5 mm/s	100 μm	Increase hydrophilicity	Prevent the colonization of titanium surfaces by bacteria and subsequent formation of biofilm	Ref.77
PDMS	800 nm	50 fs	1 kHz	50 mW	8 mm/s	8 μm	Superhydrophobic in air superaerophilicity in water	Air bubbles selectively passing	Ref.78
PTFE	1030 nm	250 fs	75 kHz	5 W	0.6 m/s	10 μm	Superhydrophobicity in air superaerophilicity in water	Directional transportation and collection of gas bubbles	Ref.79
Titanium	1030 nm	300 fs	1 kHz	0.49 J/cm²	300 mm/s		Superhydrophobic	Repelling bacterial attachment, limiting biofilm formation and simultaneously enhancing the biocompatibility	Ref.80

manufacturing, and soft robotics. However, the resolution of conventional manufacturing methods cannot be reduced to below the micron level. Therefore, further reducing the processing resolution is one of the main areas of focus in this field. The femtosecond laser interacts with materials in micro/nanoscale, and this phenomenon considerably improves its processing resolution. Researchers have proposed a range of solutions, such as using improved materials and manufacturing methods, by fully utilizing the abovementioned feature. Zheng *et al.* designed and synthesized a new water-soluble two-photon initiator to achieve TPP in aqueous-phase photoresist systems with a resolution of 180 nm [88], as shown in Fig. 7(b). Tudor *et al.* used poly(ionic liquid)s to improve the z-direction resolution of the fabricated microstructures [89]. Shukla *et al.* proposed a new method of two-photon writing in gold precursor-doped photoresist for preparing subwavelength-resolution metallic nanostructures in the polymer matrix of a photoresist film [90] to improve the processing resolution of metallic structures. Furthermore, Sun *et al.* used a "dual-3D fabrication approach" to realize the microscale production of smart structures and fabricated smart microflowers, microvales, and microclaws [91], as shown in Fig. 7(a). New methods for improving the processing resolution have primarily focused on the development of novel smart materials and improvements in the processing methods. However, various issues, such as poor biocompatibility and low efficiency, remain. Therefore, considering how to improve the resolution of the structures fabricated and how to increase the biocompatibility of the materials used is important.

2) IMPROVING BIOCOMPATIBILITY

The biocompatibility needs to be improved while ensuring controllable 3D fabrication. However, most currently available devices are made of commercial polymers, which have poor biocompatibility. Therefore, the controllable, high-precision fabrication of micro/nanostructures using biocompatible materials is a major goal in this research area. A few such 3D complex structures have been fabricated using the

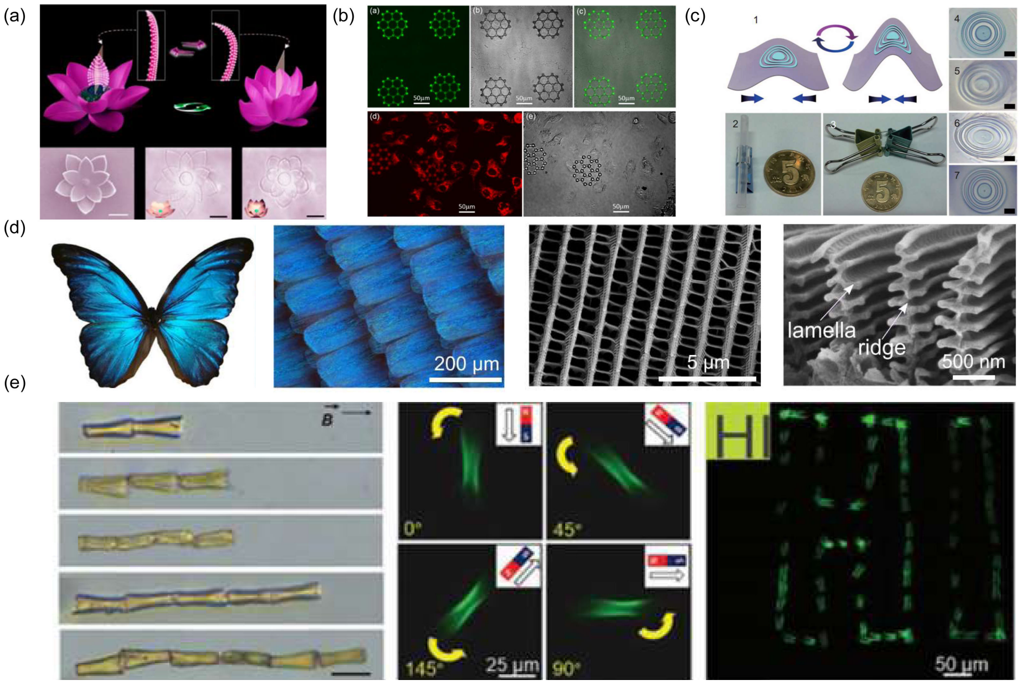

FIGURE 7. (a) Use of femtosecond laser processing for fabricating 3D devices. Flexible control of internal 3D material property gradient (light-based cross-link density) to achieve 3D construction of intelligent materials (with permission from © American Society of Chemistry). (b) Carbazole derivative (BMVPC) was encapsulated within cucurbituril 7 (CB7) as host, and water-soluble two-photon initiator based on host-guest chemistry was designed and synthesized. PEGDA hydrogel scaffold, cocultured with HeLa cells and scaffold microstructure, showed good biocompatibility (with permission from © American Society of Chemistry). (c) Novel soft biodegradable protein micro-KPL was prepared on PDMS substrate using femtosecond laser processing system (with permission from © CIOMP). (d) Color-inducing structure and composition of butterfly wings. Wings consist of lamellar nanostructures and stratum corneum (with permission from © Nature Publishing Group). (e) Propelling and steering of magnetic tubular micromotor with external magnetic field and targeted delivery of microcargoes (with permission from © Wiley-VCH Verlag).

femtosecond laser technique. Weiss *et al.* combined TPP with flexible manufacturing to create a 3D cell culture scaffold for the tissue engineering of artificial cartilage [92]. Sun *et al.* fabricated tunable protein harmonic diffractive microlenses and a microscale kinoform phase-type lens (micro-KPL) derived from protein-based soft micro-optics [93], [94], as shown in Fig. 7(c). Zyla *et al.* successfully created a layered nanostructure that mimics the shape of the wings of a butterfly [95], as shown in Fig. 7(d), to fabricate biomimetic structures. Sun *et al.* fabricated a joint-like cantilever structure by TPP using PEG-DA with good biocompatibility and high stimulus-response [96]. Plamadeala *et al.* used TPP to produce bioinspired microstructures called microneedles, which have great potential for use in medical applications [97]. These structures, which exhibit excellent biocompatibility and fast response, highlight the potential of femtosecond lasers in the field of biomedical engineering and soft robotics.

C. APPLICATIONS OF TPP IN MAGNETICALLY DRIVEN MICROROBOTS

Magnetically driven microrobots have great potential for use in biomedical engineering [98]–[100] and several other fields [101]–[103]. The key reason is that they can be controlled without physical contact and allow targeted delivery of cells in-vivo [104], as shown in Fig. 8(a). However, the mixing of magnetic nanoparticles in the material used to fabricate these robots can result in various problems, such as unevenness of shape, high surface roughness, and low mechanical strength. Femtosecond laser TPP has the advantages of high precision and stable processing, which can improve the surface smoothness and increase the mechanical strength of the fabricated structures, as shown in Table 6. These advantages can in turn significantly improve the strength and functionality of the magnetically driven microrobots [105], as shown in Fig. 7(e). At present, two methods have been used to improve the performance of magnetron robots: changing their microstructure and using new materials. For instance, femtosecond lasers can be used to fabricate a predesigned microstructure with high precision. This approach improves the motion performance of the devices and thus their ability to operate on objects and biological samples [106]. In this manner, the load capacity [107] and the targeted transport ability of magnetically driven robots can be improved [104]. With respect to the use of new materials, surface-modified Fe3O4 nanoparticles were dispersed in a gel-type photoresist, and 3D hydrogel micronails were prepared using a femtosecond laser [108]. These structures showed obvious and reversible external-magnetic-field-driven bending characteristics. Microswimmers that allow for the release of doxorubicin have also been fabricated through two-photon-based 3D printing on a natural polymer derivative of chitosan in the form of a magnetic polymer nanocomposite [109]. The microswimmers also showed good biocompatibility. Femtosecond laser technology has the inherent characteristics of high precision and flexibility, which can help

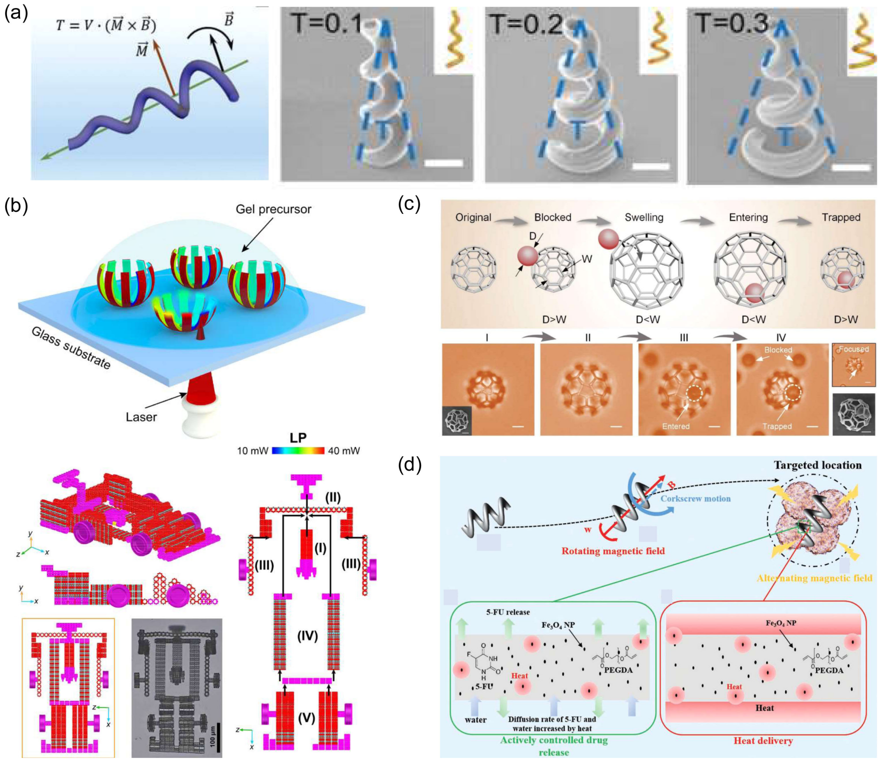

FIGURE 8. **(a) Diagram of magnetic spiral structure. SEM images of 45°-tilted microhelices with different taper angles. Transportation of single neural stem cell (NSC) (with permission from © Wiley-VCH Verlag). (b) Using TPP, complex structure was designed, analyzed, and simulated, and design, fabrication, and control of "Transformers" at nanometer scale (transforms from racing car shape into humanoid robot structure) was realized (with permission from © American Association of the Advancement of Science). (c) Microcage structure was fabricated and used for capturing and releasing particles based on changes in pore diameter in response to pH. Magnetic layer was coated on microcage to realize transport of particles based on magnetic field (with permission from © Wiley-VCH Verlag). (d) Magnetically driven biodegradable hyperthermia microrobot constructed using two-photon polymerization technology. Robot can be used to actively control drug release and hyperthermia therapy (with permission from © Wiley-VCH Verlag).**

reduce surface roughness and allow precise control over the micro/nanostructure shape during the fabrication of magnetic robots. Structures produced by mixing novel materials have great potential for use in targeted transportation, biomedicine, and other fields because of their superior operability and biocompatibility.

D. APPLICATIONS OF TPP IN SOFT ROBOTICS

As mentioned earlier, high-resolution, fast-response micro/nanostructures can be fabricated using DLW (direct laser write). This fabrication technique has led to rapid developments in the field of soft robotics, as shown in Table 7. There are still a few major problems to be solved in order to achieve wider applications for this technology, such as ensuring that the robotic devices adapt to complex biological microenvironments, constructing multi-kinetic surfaces, and achieving the rapid manufacturing of specific structures. Various solutions have been proposed to solve these problems. In 2015, Wiersma *et al.* reported the fabrication of microscopic walkers that are driven by light and are based on a liquid crystal elastomer artificial muscle [110]. These microscopic walkers can walk, rotate, and jump when they interact with the environment. In 2018, Sun *et al.* proposed a microrobot with a burr-like porous spherical structure driven by a magnetic field gradient. These microrobots were fabricated by TPP for targeted drug delivery to cells in the body [111]. In 2019, Duan's group used the laser direct writing technique to fabricate reconfigurable 4D-printed micromechanical structures through printing on heterogeneous stimulus-responsive hydrogels [112]. They also proposed a design strategy for smart microjoints (SMJs) and used these SMJs to fabricate microcrawlers that exhibited fast crawling and multimode movement [113]. On the basis of these devices, they proposed reconfigurable microsized building blocks that can respond to external stimuli [114], as shown in Fig. 8(b). In the same year, Choi *et al.* produced a spiral-shaped magnetically driven biodegradable micro/nanorobot that can be used for actively controlled drug release and thermotherapy [115], as shown in Fig. 8(d). In 2020, Duan *et al.* fabricated a hydrogel-based light-driven microactuator by TPP. The device exhibited a fast light response [116]. In addition, Chu *et al.* fabricated bioinspired microstructures shaped similar to leaves and flowers based on a pH-responsive hydrogel. They also added magnetic particles to produce a microcage for capturing and moving microparticles [117], as shown in Fig. 8(c). The DLW technique can solve many of the problems associated with material biocompatibility and the fabrication of multifunctional structures in the field of soft robots. Thus, this method has great potential in the miniaturization of soft robots suitable for use in biomedical engineering.

TABLE 6. Magnetically Driven Microrobots Processed by Femtosecond Laser

Material	Size	Moving mode	Application	Reference
SZ2080, Ni/Ti bilayer	Heights (45–75 μm), diameters (6–18 μm), pitch numbers (2–4), taper angles (0.1–0.6 rad), and pitch periods (10–30 μm)	Corkscrew locomotion	Transportation of nanoparticles (SiO2 and Ag) and neural stem cells (NSCs)	Ref.104
Fe_3O_4, SZ2080	Height of 75 μm, minimum central diameter of 11 μm, and wall thickness of 1.1 μm.	Rotate and move by magnetic torque and force	Targeted delivery of Hela cells and doxorubicin drug particles	Ref.105
Fe_3O_4, su-8	Outer diameter of approximately 9 μm and a length of 60 μm.	Wobblefree corkscrew propulsion	Enabling increased load capacity for biomedical applications.	Ref.107
Fe_3O_4,PEGDA	10μm	External magnetic field driven with a wide θ of 52.4°	First report of a magnetically actuated 3D hydrogel microstructure fabricated by TPP in which the dimensions reached ~10 μm.	Ref.108
Acetic acid, ChMA, LAP, PEG/ amine, SPIONs	6 μm diameter and 20 μm length, double helices	Actuated and controlled in an aqueous environment with an average speed of 3.34 ± 0.71 μm/s under a 10 mT and 4.5 Hz rotating magnetic field	The combination of light triggered drug delivery with magnetically powered microswimmer mobility.	Ref.109

TABLE 7. Soft Robots Processed by Femtosecond Laser

Material	Size	Drive mode	Application	Reference
Liquid crystalline elastomers (LCEs) , azo dye	60 × 30 × 10 μm	Liquid crystalline monomer composition and actuating response of a liquid crystalline elastomer (LCE) microstructure	Micro swimmers , micro jumpers , origami photonic devices	Ref.110
Heterogeneous stimulus-responsive hydrogels	Line widths of ~250 nm	pH-responsive properties	Smart and multifunctional micromachine candidates for various engineering applications	Ref.112
PEGDA,PETA,MNPs, 5-FU	6-120 μm	Controlled by a rotating magnetic feld (RMF)	Actively control drug release and thermotherapy	Ref.115
pH-responsive hydrogel	<100μm	pH-responsive properties	Micromanipulation, single-cell analysis, and drug delivery	Ref.117

IV. SUMMARY AND FUTURE PROSPECTS

In this review, we have summarized the recent developments of femtosecond laser processing in creating micro/nanostructures and their applications. Femtosecond lasers induce a limited heat-affected zone and thus exhibit ultrahigh precision due to their high pulse energy, high frequency, and ultrashort pulse duration. Currently, the smallest features that can be fabricated using femtosecond lasers are approximately 9 nm [118]. The wavelength of femtosecond lasers can range from visible to infrared light. Thus, they can be used to process various materials, such as metals, semiconductors, dielectrics, and polymers. In addition, femtosecond lasers are suitable for fabricating large-area, albeit they may take a long time, designed micro/nanostructures on various substrates. These micro/nanostructures play an important role in the development of novel optical devices, sensors, flexible electronic devices, and surface modification. With the help of femtosecond lasers, the current large size, slow response, single function, low portability, poor biological compatibility, and other defects will be overcome by processing functional structures on specific new multifunctional, biocompatible materials. Therefore, femtosecond lasers can accelerate developments in fabricating new functional structures on materials that are widely utilized in sensors and other devices. The principle of two-photo absorption has extended the applicability of femtosecond lasers. More smart materials can be polymerized with TPP technology with the development of this technique. For example, TPP technology can be used to realize cross-scale arbitrary structures with stimulus response functions and deformation capabilities on various materials. Thus, enabling the fabrication of micro/nanorobots that exhibit responsiveness to different stimuli, including chemical, optical, magnetic, and heat stimuli. Various multifunctional soft actuators and robots can be fabricated in the micro/nanoscale, which have great potential for applications in biomedicine, such as delivering drugs in vitro, releasing drugs controllably, and manipulating of single cells. However, most currently available femtosecond laser processing systems use a single beam for scanning or use optical devices such as a spatial light modulator for patterning. These devices are expensive and slow and thus unsuitable for large-area fabrication. Furthermore, the distance between the sample and the laser focal point during fabrication needs to be more precisely controlled. This requirement increases the complexity of the systems. Therefore, the most important advantages of femtosecond lasers processing, namely, high resolution, high precision, independence from environmental conditions, and applicability to a range of materials, would be enhanced if the processing speed can be increased.

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