Laser irradiation construction of nanomaterials toward electrochemical energy storage and conversion: Ongoing progresses and challenges

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Abstract
The emerging use of laser irradiation in synthesis smartly bridges “nanotechnology” and “light”, and has attracted enormous attention as an efficient synthetic methodology for versatile nanomaterials toward electrochemical energy storage and conversion devices (ESCDs). In this review, recent contributions and progress regarding the laser-induced nanomaterials for ESCDs are comprehensively summarized, with a special focus on their practical utilization in rechargeable batteries, supercapacitors and electrocatalysis. The laser-induced synthesis strategies and corresponding mechanisms involved in nano-architecture generation/regulation, including pulsed laser deposition and laser irradiation in liquid, are also discussed in detail. With the in-depth insights into the mechanisms and revolutionary advancements of laser irradiation technology, the comprehensive performances of ESCDs have been strikingly optimized. Finally, the existing challenges and future directions in this booming research field are outlined. This review will exert the significant guidance for future design and purposeful fabrication of advanced laser-induced nanomaterials with appealing properties for advanced ESCDs and beyond.

KEYWORDS
electrocatalysis, energy storage and conversion, laser-induced nanomaterials, rechargeable batteries, supercapacitors

1 | INTRODUCTION

The rapid depletion of fossil energy, along with the growing concerns for energy crisis and environmental pollution, has become a major world challenge at present.1–4 Renewable energy, including wind, solar, and biomass energies, has been extensively explored to accelerate the sustainable development of the society.5,6 Recently, the development of new energy storage and conversion devices (ESCDs) based on electrochemistry, such as rechargeable batteries, supercapacitors, and electrocatalysis, has enormous importance for addressing the above issues.7–11 Although the aforementioned devices operate with various working mechanisms, the electrochemical performance and reliability of all the ESCDs are mainly restricted by the electroactive materials involved. Hence, it is of tremendous significance to...
engineer and synthesize advanced materials rationally for the further progress of ESCDs.

In the past decades, nanotechnology and nanoscience have rapidly developed and have been widely applied in almost every energy-related field. However, the urgent requirement for different nanomaterials has stimulated the tremendous progress in synthetic methodologies. Moreover, with a rational design and synthesis protocols, nanomaterials with various structures, sizes, and compositions can be controllably fabricated, thus contributing to the composition/structure-performance relationship. However, the advancement of nanomaterial fabrication still faces many challenges. Conventionally, traditional synthesis strategies of nanomaterials are always time-/energy consuming, and subject to undesirable controllability. As a result, the facile yet cost-efficient and rapid synthesis strategies of high-performance nanomaterials for ESCDs are urgently desired.

Recently, the emerging technique of laser irradiation synthesis, which smartly integrates the nanotechnology with the “light”, has drawn wide attention from researchers worldwide as an efficient approach, and holds enormous promise for adjusting the surface and/or electronic structure of materials. Unlike the traditional thermal treatment techniques, laser irradiation synthesis is based on photothermal effect induced by a pulsed laser beam, which produces a confined soaring temperature range at a specified location with satisfactory controllability, accompanied by fast cooling after the laser irradiation. Strikingly, the thermal effect can be adjusted easily by the applied laser parameters including frequency, pulse width, and laser intensity. The main advantages of laser irradiation synthesis are the brevity of its involved time and site-specific photothermal effect, which can rapidly obtain nanomaterials with low energy consumption. The rapid heating and cooling rates (>10⁶ °C s⁻¹) can generate a particular reaction condition, which can accurately modulate specific surfaces/interfaces and microstructures of resultant materials. Furthermore, no waste of reagents should economize on costs for waste disposal, which manifests the environmental benignity intrinsic to laser irradiation synthesis. The marvelous prospects emerging from the interaction of light with materials have vitalized a large number of groups to focus their studies on laser fabrication, structural modification, performances, as well as various applications of laser-induced nanomaterials. Encouragingly, laser irradiation techniques are also be compatible with a vast range of emerging material synthesis strategies, for example, three-dimensional (3D) printing. This pioneer field for laser irradiation is just beginning.

Herein, in this review, we mainly focus on recent progress in laser-induced nanomaterials for ESCDs based on electrochemistry. After a brief presentation of research history and primary classification of laser irradiation techniques, we discuss the fundamental understanding of these approaches, that is, the reported mechanisms involved in the micro-/nanostructure generation and/or modifications. Subsequently, the structural regulation/optimization, and versatile applications of laser-induced nanomaterials for ESCDs, such as rechargeable batteries, supercapacitors, and electrocatalysis, will be comprehensively summarized. We especially focus on the unique advantages of laser irradiation, including the controllable introduction of defects and/or heteroatoms, epitaxial growth on an appropriate substrate, and elaborate fabrication of heterostructures, all of which are greatly beneficial to facilitating electrochemical performances. Finally, we briefly discuss the challenges and propose future directions of laser irradiation techniques in this emerging research field.

2 | PRINCIPAL LASER IRRADIATION METHODOLOGY

A brief presentation of the research history of various laser irradiation approaches is schematically described in Figure 1(A). In 1960, Maiman fabricated the first functional laser at Hughes Research Laboratories. After that, the laser-material interaction was first utilized to fabricate semiconductors and dielectric thin films using a ruby laser in 1965, which is termed pulsed laser deposition (PLD). It was more than 30 years ago that laser irradiation in liquids (LIL) was first implemented to manufacture and process laser-induced nanomaterials. In 1987, Patil et al. pioneered laser ablation in liquids (LAL). The first studies on laser fragmentation in liquids (LFL) and laser melting in liquids (LML) were reported around 1998 and 1999 by Kamat’s et al. and El-Sayed’s et al., respectively. Since then, lasers have been extensively employed in the fields of science and technology, among which laser irradiation technology to make functional materials is unquestionably one of the vital applications. The advantages, disadvantages, and mechanisms of this unique technology are elaborately discussed in Sections 2.1 and 2.2. The increased attention is emphasized by the growth in the volume of published research in the past two decades collected from the Web of Science with the keywords “pulsed laser deposition” and “laser irradiation in liquid”, as profiled in Figure 1
(B). It is especially noteworthy that the number of publications sharply increased after 2010, highlighting that the emerging field is attracting more and more research attention as a rising star.

To date, a great number of materials have been utilized to generate lasers, enabling highly adjustable laser wavelengths ranging from ultraviolet (e.g., XeCl, 308 nm; KrF, 248 nm; ArF, 193 nm; F₂, 157 nm) to infrared (e.g., CO₂ laser, 10.6 mm) (Figure 2(A)). The produced laser light is characterized by outstanding directionality, coherence, and monochromaticity, enabling selective processes and a localized soaring temperature range within a localized interaction volume. Figure 2(B) plots the diverse types of laser irradiation effects and their applications. As a whole, the field of laser irradiation is interdisciplinary, including physics, chemistry, fluid mechanics, molecular dynamics, spectroscopy, and related subdisciplines (Figure 2(C)). Accordingly, irradiation of the target material via laser-induced techniques is a very complicated process. It is based primarily on the absorption of laser energy in the target material, leading to diverse effects including heating, ablation, plasma formation, vaporization, as well as melting. The extent of the above effects is mainly dependent on the features of the incident laser (laser wavelength, pulse width, laser intensity, frequency, and interaction time) and the optical and thermophysical performances of the target material (refractive index). The penetration depth that the laser pulse permeates into the surface of the target is typically in the range of 10 nm. At sufficiently high laser energy, the target material is commonly transformed to plasma, which is characterized by high temperature, high pressure, and high density. Afterward, the enormous pressure diversity between the laser-induced plasma and environmental atmosphere gives rise to a fast expansion of the plasma species, and subsequently, it cools quickly. Under appropriate condensation conditions, the plasma species can nucleate and grow into desired configurations on an appropriate substrate or in a liquid. Accordingly, laser irradiation, as a booming technology, has achieved significant progress in the wide applications involving fabrication, modification, and processing of nanomaterials.

On account of the wide range of laser sources and greatly adjustable operations, many synthesis and processing approaches of laser-induced nanomaterials have been put forward, such as the PLD, LAL, LFL, and LML. The methodologies above only represent a fraction of the newly developed laser-induced techniques in

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**Figure 1** (A) The development chronology of various laser irradiation approaches. (B) Statistical data of the volume of publications themed on pulsed laser deposition and laser irradiation in liquid reported from the year 2000 to the present (December 2020)
recent years. Laser-induced strategies are a flourishing field that deserves worldwide attention. Especially, the inherent characteristics, that is, tuning structures and accurate performances of materials at lower thermal budgets, substantially open up a selective route to expanding novel nanomaterials for ESCDs.

2.1 Pulsed laser deposition

Laser-material interactions can be employed in different deposition conditions (substrate temperature and environmental atmosphere) to construct various nanomaterials. The PLD, as a physical vapor deposition technique, is deemed to be a scalable and universal avenue for thin-film growth. In general, a PLD system is primarily composed of a laser source, optical path system, deposition chamber, pumping system, and gas path system, as illustrated in Figure 3(A). The laser-induced nanomaterials are always deposited directly onto the substrate. Because the laser source is placed outside the deposition chamber, the deposition process is implemented in an ultrahigh vacuum or the presence of a background atmosphere.

In essence, the PLD technique is conceptually simple. Even so, the PLD process itself hides a vast range of complexity and is insufficiently understood. The mechanism of the PLD technique is simply described as follows. In the process of PLD, the surface of the solid target material is typically impinged by a focused laser beam with high kinetic energy (up to a few kiloelectron volts) at an angle of 45° during a short time in the vacuum chamber. The solid target is generally rotated to avoid repeated irradiation of the same spot on the target. The material is ablated/vaporized from the solid target by the interaction between the laser pulse and target to form a plasma species containing ions, atoms, molecules, electrons, clusters, and molten globules. The ejected plasma species can rapidly expand and their distribution is usually symmetric around the target surface. Afterward, the ejected flux strikes and deposits as a thin film directly on the substrate. The above process stages are always superimposed cyclically and interrelated. Up to now, the PLD approach has been successfully employed to deposit a variety of thin films including carbon-based materials, oxides, nitrides, and amorphous, single-crystal or polycrystalline thin films with diverse configurations.

Compared with other traditional techniques, the PLD approach is versatile, rapid, and cost-effective, enabling the fabrication of thin films with good adhesion to the substrates and higher crystalline quality at comparatively low temperatures. The PLD also makes it possible to introduce any dopants at any concentrations. The thickness and morphology of the generated thin films are closely regulated by modulating the primary deposition parameters including the fluence, laser wavelength, pulse width, spot size, repetition rate, background gas composition, target-to-substrate distance, target composition, substrate temperature, and pressure of the background gas. The main advantages of PLD are summarized in Figure 3(B). Encouragingly, composite thin films with more complicated compositions can be fabricated easily by depositing several different targets to accomplish co-irradiation. Nevertheless, the maximum target-to-substrate distance and the angular location of the substrate may be fixed owing to the geometrical...
confinements of the deposition chamber. Remarkably, a high-quality thin film with the desired thickness, architecture and composition can be obtained by rationally adjusting the aforementioned interdependent deposition parameters.44–47

2.2 | Laser irradiation in liquids

In contrast to PLD, in which thin films are formed and fixed on an appropriate substrate in the ultrahigh vacuum or the presence of a background gas,36,44,48 LIL (Figure 3 (C)) accelerates the formation of nanomaterials and only requires simple precursors and processing equipment (just a solid plate, and water or solution). The liquid environment not only offers some highly controllable parameters for laser irradiation synthesis but dramatically influences morphologies and microstructures of the achieved laser-induced nanomaterials. Interesting morphologies and size modifications of nanoparticles (NPs) fabricated using laser processing have been obtained.49–51 LIL also has the advantages of the reduced formation of side products, and absence of catalysts, as shown in Figure 3(B).

The LIL is generally classified into three primary categories, that is, the LAL, LFL, and LML. Since the first report by Patil et al. in 1987 on the LAL31 and pioneering synthesis of nano/subnano colloidal metal particles using the LAL by Henglein in 1993,52 the LAL has been established as an extensible yet universal approach for simple fabrication of nanomaterials.53,54 As derivative laser irradiation techniques in liquid, both the LFL and LML allow elaborate size regulation of the as-obtained nanomaterials over a wide range from micrometers to nanometers. In this section, we will introduce the three laser-induced methods in liquids and mainly focus on the formation principles of nanomaterials with several representative examples and relevant discussions about the chemical reactions involved.
2.2.1 Laser ablation in liquids

In general, the material ablation by a laser pulse is one of the primary steps in laser manufacturing and commonly used for cutting, welding, patterning, and drilling the surface of target materials. Micro- and nanomachining using lasers is a universal strategy for material processing and is a booming topic. Unlike laser machining, where the workpiece surface is the desired product, LAL is utilized to prepare NPs. When a laser beam strikes the bulk target material (solid foils) immersed in liquid, plasma species and cavitation bubbles are formed, in which the ablated product condenses. The desired NPs are dispersed in the liquid medium after the bubbles burst (Figure 4(A)). Normally, a portion of the ablated product is unavoidably oxidized or reduced at the surface of the resultant NPs, for instance, those reduced by oxygen vacancies or the oxidized noble metal NPs. However, the major compositions of the nanomaterials after LAL are universally identical with the bulk target materials. As a result, LAL is usually deemed to be a physiochemical synthesis approach with initial processes controlled by the laser-plasma and cavitation bubble mechanism.

The aforementioned plasma species and cavitation bubble events during LAL can be divided into three phases: (i) the plasma phase, (ii) cavitation bubble phase, and (iii) phase in which desired NPs are dispersed in the liquid medium after bubble collapse, as schematically shown in Figure 4(B). At sufficiently high laser pulse intensity, plasma is produced on the surface of the target material, which consists of highly ionized and neutral species. The initial density of plasma is approximately \(10^{20} \text{ cm}^{-3}\), derived from the target, which depends on the pulse laser energy. The duration of the plasma in a wide range from tens of nanosecond (ns) to a few microsecond (\(\mu\)s), for each laser pulse adds to the liquid environment and laser parameters (e.g., laser intensity, pulse width, or repetition rate). The pressure and temperature of plasma species can reach hundreds of pascals and thousands of kelvins, respectively. During plasma attenuation, the high energy of plasma is shifted to the surrounding liquid, generating a vapor layer with approximate volume as the plasma. In 2015, Tamura’s group surveyed the plasma-cavitation bubbles relationship and discovered that both of them coexist in the inchoate phase. The ejected plasma species can interact with the liquid medium and trigger chemical reactions in the bubbles. Meanwhile, the rapid plasma quenching generates, and the vapor layer can grow into a cavitation bubble. Subsequently, the cavitation bubble experiences a sequence of expansion and shrinkage. At the end of the cavitation bubble lifetime, the bubbles

![Figure 4](https://example.com/figure4.png)

**Figure 4** (A) Scheme of laser ablation in liquids (LAL). Reproduced with permission from Reference 22 Copyright 2017, American Chemical Society. (B) Main stages occurring during the relaxation of the system for each laser pulse. Reproduced with permission from Reference 67 Copyright 2014, Royal Society of Chemistry. (C) Schematic illustration of the formation of FeO nanoparticles (NPs) by laser ablation in liquids (LAL). Step I: Production of iron plasma above the iron target quickly after one laser pulse. Step II: Ultrasonic and adiabatic expansion of the plasma and formation of iron clusters. Step III: Formation of FeO NPs. Reproduced with permission from Reference 60 Copyright 2008, American Chemical Society
remain attached to the target materials surface or entirely disappear.\textsuperscript{22}

It is of great importance to regulate the size of the LAL-fabricated product via an in-depth comprehending of where and how NPs grow. The mass abundance of primary NPs (<10 nm) alters depending on the bubbles’ shape. Several major NPs can be probed in nearby cavitation bubbles.\textsuperscript{69} Previous studies have shown that the excited species from the laser-irradiated substrate are in direct contact with the vapor, which may give rise to chemical reactions, and NPs have been confined in cavitation bubbles. More importantly, liquid can be transformed into a plasma-heated supercritical phase during LAL. The supercritical liquid and cold bubble vapor could offer appropriate conditions for the crystallization of NPs. In the case of a bulk iron target (Figure 4(C)), iron plasma species are produced at the interface between solid and liquid after one laser pulse. Subsequently, ultrasonic and adiabatic expansion of plasma results in rapid cooling of the plume area. Accordingly, the thus-obtained iron clusters can interact with the surrounding liquid, leading to the formation of FeO NPs.\textsuperscript{60}

2.2.2 | Laser fragmentation in liquids

If a powder suspension is used as the ablating target, instead of a general bulk target material (i.e., solid foils), lots of intriguing phenomena would appear. The suspended NPs will experience the laser fragmentation (i.e., LFL) or melting (i.e., LML) processes in liquids, generating smaller particles with identical or diverse shapes and chemical compositions. The laser fragmentation of large particles in liquids is produced by the laser energy absorbed by the involved particles themselves (Figure 5(A)).\textsuperscript{22} To implement the LFL, solid-state lasers are usually combined with focusing optics to produce a laser focus in the diameter range of several tens of micrometers to hundreds of micrometers. The difference in the focus position can result in an evident change in laser fluence. Accordingly, LFL efficiency is highly sensitive to focus change. Overall, the efficiency of LFL strikingly increases with the laser fluence, and the maximum value is limited by the liquid’s optical breakdown.\textsuperscript{70} Thus, the strong focusing is necessary unless high-pulse-energy lasers that do not require focusing are practicable. Side products derived from the LML are often detected in the laser-induced products during LFL with short focus distances. Although side products can be averted by using high-pulse energy lasers, it may still be restricted still by a low repetition rate, resulting in a lower yield. Size selectivity can be accomplished using LFL\textsuperscript{71}; however, the physisorption of laser-induced nanomaterials on the initial solid suspended particles may reduce the reproducibility and efficiency because of the inherent disadvantage of consecutive solid suspended particles-product mixing.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure5.png}
\caption{(A) Scheme of laser fragmentation in liquids (LFL). Reproduced with permission from Reference 22 Copyright 2017, American Chemical Society. (B) Schematic illustration of the size-reduction and fragmentation mechanisms of plasmonic nanoparticles (NPs) in a liquid. Reproduced with permission from Reference 71 Copyright 2015, American Chemical Society. (C) Schematic diagram depicting changes at the surface of Si nanocrystals during the laser fragmentation process. Reproduced with permission from Reference 86 Copyright 2011, American Chemical Society. (D) TEM image and corresponding SAED patterns of primal ZnO hollow nanospheres and the formed ZnO quantum dots after irradiation. Reproduced with permission from Reference 88 Copyright 2011, American Chemical Society.}
\end{figure}
Based on theoretical calculations and experimental investigations, mechanisms of photothermal evaporation and Coulomb explosion are proposed, as depicted in Figure 5(B). Koda's group first reported the photothermal vaporization mechanism for LFL. The finding implies the size reduction of NPs in an aqueous solution by laser processing of a pulsed Nd:YAG laser at 532 nm. The size reduction via surface evaporation begins when the temperature of particles surpasses the boiling point of the bulk precursor. Next, the cooling of vaporized atoms in the liquid results in their condensation into even smaller particles. Amendola et al. demonstrated that the average size of produced Au NPs can be further reduced with an increase in the pulsed nanosecond laser fluence from 88 to 442 mJ cm\(^{-2}\) during LFL. The Coulomb explosion mechanism for LFL (Figure 5(B)) reveals that electrons are ejected from the bulk precursor to produce ionized NPs. Based on the metal-cluster fission and the liquid-drop model, a multiply charged cluster is unstable only if the fractured Coulombic force is greater than the cohesive force. Thus, as-obtained ionized NPs are rapidly fragmented on account of internal charge repulsion.

The particle size of the LFL-induced products attributes to properties of the suspended particulates and laser parameters, including incident laser fluence, pulse widths, as well as laser irradiation time. Kabashin et al. revealed that the particle size of the crystalline nano-Si could be governed by regulating the concentration of initial solid suspended particles. Note that in the contribution, a coalescence-based formation mechanism via LFL underlines the significance of suspended solid concentration. In addition, the size of silicon NPs, as shown by Intartaglia et al., is remarkably decreased to 3 nm in the case of LFL at 355 nm (high absorbance), which is smaller than that (40 nm) at 1064 nm. The LFL method, as a top-down process, primarily involves physical changes. However, it is occasionally accompanied by phase transformation or chemical reduction/oxidation reactions. For instance, the reductive LFL of CuO and Cu\(_2\)N in acetone leads to the formation of metallic copper or Cu\(_2\)O NPs. Typically, the reductive ablation process is ascribed to the solvents (diethyl ketone or acetone) on account of diverse high-temperature cooling durations. Intriguingly, as elucidated by our previous work, the fragmentation of suspended ZnCo\(_2\)O\(_4\) nanosheets (NSs) via nanosecond-pulsed laser irradiation does not influence the composition of ZnCo\(_2\)O\(_4\), consequently producing ZnCo\(_2\)O\(_4\) NPs.

The surface chemistry of LFL-produced products also generally changes. Svrček et al. designed the surface of electrochemically etched silicon nanocrystals (NCs) via ns-LFL and reported several modification stages influencing the surface chemistry of these silicon NCs, including the cleavage of Si-dimers by water vapor and condensation of hydroxyl groups or OH ions with H atoms (Figure 5(C)). In this case, once the fragmentation process begins, the replacement of Si-H bonds with Si-OH bonds is facilitated. Moreover, Si NC aggregates are fragmented. Consequently, Si NCs formerly unexposed to the water are subjected to cleavage of Si-dimers by water molecules. The above processes can continue for about 15 min in water during LFL. When the LFL continues for more than 15 min, OH surface coverage is complete. The adjacent OH terminations will continue to condense and accelerate surface oxidation, resulting in strained bonds and structural defects. After 45 min processing in water, the core of Si NCs relatively decreases in size. Interestingly, fabrication of oxygen vacancies during LFL can also be deemed as a local chemical reduction process. The defects can be incorporated into the generated zinc oxide particles during LFL, as depicted by Lau and Barcikowski and Zeng et al. (Figure 5(D)). Encouragingly, it can promote the charge transport of bulk materials by introducing rich vacancies and defects into LFL-induced nanomaterials. For example, electrochemically etched Si NCs via LFL in aqueous solutions can invert the hydrophobic surface into a hydrophilic one, improving the external quantum efficiency. Recently, abundant oxygen vacancies were fabricated on the surface of transition metal oxides (TMOs), which dramatically alters the electric conductivity and electrochemical activity of TMOs, and consequently accelerating the kinetics of the involved redox reactions. In view of lots of theoretical and experimental studies on LFL, it can be expected that the derived model will be extended to other nanomaterials in the future.

### 2.2.3 Laser melting in liquids

Moderate isochoric photothermal NP processing by LIL media has been well elucidated and is currently progressed for synthesizing submicrometer spheres using LML (Figure 6(A)). One particularly notes that LML is a relatively simple and scalable technique augmenting the particle size in a controllable approach. More importantly, no chemical reagents are required in the transformation process of particle size. El-Sayed et al. accomplished the pioneering achievement (Figure 1(A)) in the LML field, and then, the Koshizaki and coworkers extended the applications to metals, oxides, carbides, and even semiconductors.

In common, LML is categorized into two types, that is, fusion-LML increasing the crystal size of particles and
reshaping-LML (an isochoric process). With fusion-LML, the ZnO NPs (<50 nm) are transferred into size-tailored spheres of about 340 nm in size with a small size dispersity, as shown by field emission scanning electron microscopy images (Figure 6(B,C)). The average size of single-crystalline ZnO submicrometer spheres (Figure 6(D,E)) can be precisely tailored by regulating the input laser fluence. Okamoto et al. expanded the application of LML to fabricate single-crystal microspheres with high sphericity derived from several anisotropic materials. As a whole, LML can not only modulate the size of the resulting submicrometer-size spheres but also regulate their surface structure and composition. Furthermore, LML can be utilized to melt NPs or nanorods (NRs) into nanowebs or nanochains.

As for the isochoric reshaping, solid suspended particles and as-obtained laser-induced products via LML possess the same volumes. Reshaping through LML can transform not only bulk materials (Figure 6(F)) but also nanoctahedra, nanocubes, NRs, and so on. At present, agglomerated NPs are always used to generate submicrometer spheres via LML, which includes fusion and reshaping of suspended NPs. Cao group reported that LML-induced morphology evolution and phase transformation from Fe$_3$O$_4$ cube particles to submicrometer spheres can be finely modulated by laser fluence and irradiation time (Figure 6(G)). It is due to the high temperature of particle surface and high cooling rate involved in LML that reshaping and phase transformation take place. The LML process may be accompanied by the chemical reduction/oxidation processes.

3 | STRUCTURAL REGULATION AND APPLICATIONS OF LASER-INDUCED NANOMATERIALS

Electrochemical energy storage and conversion always involve electrochemical reactions driven by electrons from electrode materials, including carbon-based materials, transition metal compounds (TMCs, oxides/sulfides/nitrides/carbides), and metal NCs. Laser irradiation, as a simple, scalable, yet universal approach, has been extensively...
nanomaterials have been utilized as electrode materials with multifunctions. Specific structures and performances of laser-induced nanomaterials can be accurately regulated by smart techniques. In this section, the recent progress in laser-induced nanomaterials for ESCDs (rechargeable batteries, supercapacitors, and electrocatalysis) will be comprehensively summarized along with their highlighted advantages, which will offer several directive perspectives in rational design and fabrication of advanced laser-induced nanomaterials for high-performance ESCDs.

3.1 | Rechargeable batteries

The ongoing progress in rechargeable batteries is highly crucial for ESCDs benefiting from their high energy density, long cycling life, sustainability, and environmental friendliness. Therefore, they have witnessed increasing concerns.10 Rechargeable batteries, which apply with diverse pairing materials, can perform reversible redox reactions at specific voltages to generate high energy density. Among rechargeable batteries, lithium-ion batteries (LIBs), sodium-ion batteries (SIBs), zinc-ion batteries (ZIBs), and metal-air batteries (MABs) have attracted increased attention from researchers worldwide.9,15,16,107–109 However, some key issues are restraining their progress, including low capacities, inferior cycle life, and sluggish reaction kinetics. In addition, intrinsic reaction mechanisms unavoidably lead to large volume variation and sluggish electrochemical kinetics, which are extremely detrimental to the improvement in electrochemical properties. Accordingly, purposeful regulation in electrode materials to alleviate volumetric change and boost electrochemical reactions becomes one of the predominant topics in the rechargeable batteries field.110–112

The merits of laser-induced nanomaterials in precise yet simple structural control provide a reliable and prominent platform to investigate the underlying relationship between structure and performance. Table 1 summarizes typical the laser-induced nanomaterials for rechargeable batteries.

3.1.1 | Lithium-ion batteries

The PLD technique with highly flexibility has been extensively utilized to fabricate electrodes for LIBs.9,15,16,107–109 In the typical PLD process, target materials are induced into crystalline/amorphous NPs. Indeed, nanomaterials derived from PLD are composed of ultrafine particles, whose highly active sites are exposed, and meanwhile, the ions’ diffusion distance is radically shortened as well. The PLD approach exhibits extensive applicability, and many PLD-derived nanomaterials have been utilized as electrode materials with superior performance for LIBs, including CoO-Co (Figure 7(A–C)), SnO2, graphite, Fe3O4, LiNi0.5Mn1.5O4, VO2, GeS, and GeS2.113,115,117,118,120–125 The PLD-derived CoO-Co electrode, where the nano Co dispersed well in the CoO matrix, exhibits good structure integrity (Figure 7(A, B)).115 The reaction kinetics of the electrode is strikingly enhanced, thanks to the improved electronic conductivity from the nonelectroactive metallic Co. Moreover, PLD can also fabricate freestanding and multicomponent electrodes.113,126,127 A freestanding amorphous GaN@Cu electrode was engineered via a low-temperature PLD technique, which demonstrates outstanding rate capability and superlong cycle life (509 mAh g−1 at 10 A g−1 after 3000 cycles) because of its high electronic conductivity, synergistic contributions from diverse components, and structural integrity with cycling (Figure 7(D–H)).126 Even so, the primary deficiency of the method lies in its low efficiency, high cost, and high demand for a complicated vacuum and laser system.

Except for general laser irradiation synthesis, an epitaxial growth on an appropriate substrate has also been investigated for LIBs. An anodic aluminum oxide-assisted PLD was developed to fabricate epitaxial array of Fe3O4 nanodots (NDS) on Cu substrate as a binder-free anode for LIBs.118 The high-quality deposition of well-ordered Fe3O4 NDS array on Cu foil with tight contact (Figure 7(I–K)), thus facilitating rapid electronic transport. Owing to the unique structural merits, unlike the laser-induced Fe3O4 film, the Fe3O4 array anode reveals excellent capacity and long cycling life (350 mAh g−1 at 400th cycle) even at a super-high rate of 5C (Figure 7(L)). The superior electrochemical lithium storage performance can be ascribed to the synergistic contributions from sluggish internal passivation, released internal stress, and outstanding structural stability all rendered by the NDS’ array structure of Fe3O4.

Furthermore, PLD has been applied to fabricate solid-state film electrolytes with high ionic conductivity for solid-state batteries.114,128,129 A cubic Li-garnet film electrolyte established using a multilayer processing strategy, which combines the Li6.25Al0.25La3Zr2O12 with Li3N layer via the PLD, displays a high ionic conductivity of $2.9 \pm 0.05 \times 10^{-5}$ S cm−1 (at 23°C).114 Appealingly, the electrolyte prepared at a treatment temperature as low as 400°C can still maintain stable ionic conductivities, enormously enhancing the commercial application of solid-state electrolyte. This approach will enable future solid-state batteries with more space for electrodes via the overall cell design and optimization.

Besides, LIL, as a booming and high-efficiency synthesis strategy, has been applied to produce laser-induced electrode materials for LIBs. More practically, LIL does not require additional additives and specific solvents, enabling the synthesis of surfactant-free products in
### Table 1: Summary of different laser-induced nanomaterials for rechargeable batteries

| Application | Laser source | Laser conditions | Medium | Laser-induced nanomaterial | Performance | Ref. |
|-------------|--------------|------------------|--------|----------------------------|-------------|------|
| Anode for LIBs | KrF excimer laser | 248 nm, 300 mJ, 5 Hz | 10⁻⁵ Pa | Amorphous SnO₂/TiO₂ films | 175 µA h cm⁻², 13.8 µA cm⁻², 200 cycles | 113 |
| Anode for LIBs | KrF excimer laser | 248 nm, 400 mJ pulse⁻¹, 6 Hz | 0.2 Pa | CoO-Co | 830 mA h g⁻¹, 500 mA g⁻¹, 200 cycles | 115 |
| Electodes for LIBs | KrF excimer laser | 248 nm, 500 Hz | — | NiS-VGCF (VGCF: vapor grown carbon fiber) | 300 mA h g⁻¹, 3.8 mA cm⁻² | 116 |
| Anode for LIBs | Pascal Mobile Combi-Laser MBE | 300 mJ pulse⁻¹, 10 Hz | 4 × 10⁻⁶ Torr | Epitaxial array of Fe₂O₄ nanodots | 550 mA h g⁻¹, 5C, 100 cycles | 118 |
| Cathodes for LIBs | KrF excimer laser | 248 nm, 180 mJ, 10 Hz | 350 mTorr | Li₁₂Mn₀.₅₄Ni₀.₁₃Co₀.₁₃O₂ | 70 µA h cm⁻², 2 µm | 119 |
| Anode for LIBs | KrF excimer laser | 248 nm, 300 mJ, 5 Hz | 6 × 10⁻⁵ Pa | SnO₂ nanoflower | 827.7 mA h g⁻¹, 5C, 100 cycles | 120 |
| Cathodes for LIBs | KrF excimer laser | 2 J cm⁻², 10 Hz | 26 Pa | LiNi₀.₅Mn₁.₅O₄ | 116.8 mA h g⁻¹, 0.5 C, 100 cycles | 121 |
| Anode for LIBs | ArF excimer laser | 248 nm, 10 Hz | 10⁻² Pa | Amorphous GaN@Cu | 509 mA h g⁻¹, 10 A g⁻¹, 3000 cycles | 126 |
| Anode for LIBs | Nd:YAG laser | 355 nm, 2 J cm⁻², 10 Hz | 2 × 10⁻³ Pa | NiO-NiSe | 495 mA h g⁻¹, 20 mA g⁻¹, 50 cycles | 127 |
| Solid electrolyte for LIBs | KrF excimer laser | 248 nm, 5/10 Hz | 0.04 hPa | Li-garnet solid-state battery films | Ionic conductivity: 2.9 ± 0.05 × 10⁻⁵ S cm⁻¹ | 114 |
| All solid-state electrolytes for LIBs | Nd:YAG laser | 266 nm, 10 Hz | 9 × 10⁻⁶ Torr | Garnet Li₆.₄La₃Zr₁.₄Ta₀.₆O₁₂ films | — | 128 |
| Anode for LIBs | KrF excimer laser | 400 mJ pulse⁻¹ cm⁻², 10 Hz | Water | Nano-ZnCo₂O₄/porous rGO | 746 mA h g⁻¹, 1.0 A g⁻¹, 250 cycles | 85 |
| Anode for LIBs | Nd-YAG fiber laser | 532 nm | Water | SnO₂ nanotubes | 1400 mA h g⁻¹, 0.1 C | 130 |
| Anode for LIBs | Nd:YAG Q-switched pulsed laser | 1064 nm, 650 mJ, 10 Hz | Hexane, 2-propanol, methanol | Black phosphorus | 404.4 mA h g⁻¹, 1.0 A g⁻¹, 200 cycles | 131 |
| Anode for SIBs | KrF excimer laser | 248 nm, 300 mJ, 10 Hz | — | γ-Fe₂O₃ thin film | 335.2 mA h g⁻¹, 475 mA g⁻¹ | 107 |
| Anode for SIBs | Q-Switched Nd-YAG laser | 355 nm, 2.5 J cm⁻², 10 Hz | Ar | SbSn-P | 252 mA h g⁻¹, 250 mA g⁻¹, 100 cycles | 109 |
| Anode for SIBs | KrF excimer laser | 248 nm, 200 mJ, 5 Hz | P₂₀₂ ~ 6, 13 and 30 Pa | Amorphous VO₂ | 136 mA h g⁻¹, 44.1 mA g⁻¹, 100 cycles | 124 |
| Anode for SIBs | CO₂ laser system | 10.6 µm, 86 W | 10⁻⁵ mbar | Hard carbon | 262 mA h g⁻¹ | 133 |
| Anode for SIBs | KrF excimer laser | 248 nm, 150 mJ pulse⁻¹, 5 Hz | 2 × 10⁻² mbar | Polycrystalline Na₂Ti₆O₁₃ | 42 mA h g⁻¹, C/20 | 134 |
| Cathode for SIBs | KrF excimer laser | 248 nm, 2 J cm⁻², 10 Hz | P₂₀₂, 65 Pa | P2-type Na₀.₆₇Ni₀.₂₅Mn₀.₇₅O₂ | 166.3 mA h g⁻¹, 13 mA g⁻¹, 130 cycles | 136 |

(Continues)
environmentally friendly solvents. The key characteristics of laser-induced electrode materials from LIL are featured with small sizes and large surface areas, which are beneficial in mitigating volume change and promoting kinetic performance. This technique puts forward simple requirements on the targets if only they can be excited by a laser. Many laser inactive/active materials can be used as targets to synthesize electrode materials. Therefore, the applicability of LIL is nearly infinite. A vast range of electrode materials, not only nanodimensional materials but also their hybrids have been fabricated via the LIL. Recently, it has been established as a novel avenue by creating intrinsic oxygen defects to modify the electrochemical properties of electrodes for LIBs. The oxygen defects can significantly facilitate the quick ion diffusion and charge transfer during continuous charge/discharge processes, as well as afford more electrochemically active sites, thereby promoting the performance of batteries. Yuan and coauthors prepared the nano-ZnCo$_2$O$_4$/porous reduced graphene oxide (L-ZCO@rGO) hybrids with exceptional capacitive lithium storage by the LFL process (400 mJ pulse$^{-1}$ cm$^{-2}$, 30 Hz). During LFL, the ZnCo$_2$O$_4$ NSs can be crushed into NPs by the fragmentation effect. Simultaneously, resultant L-ZCO NPs can be impinged by laser-induced protons from water, introducing rich oxygen defects on the surface of L-ZCO NPs (Figure 8(A, B)). Here, the one-step laser irradiation, as “one stone”, kills three birds in half an hour, that is, simultaneously introducing rich oxygen vacancies into the obtained ZCO NPs and creating mesopores in rGO NSs. The thickness, lateral size, and architectures of L-BP can be modified just by altering laser duration, laser energy, and solvent type (Figure 8(H–K)). The laser-induced exfoliation holds enormous promise in controlled and ultrafast exfoliation of other two-dimensional materials for advanced ESCDs.

Apart from the fine synthesis of anode/cathode materials and solid-state electrolytes for LIBs, laser irradiation has been verified as a powerful technique to modulate the as-fabricated electrode for further promoted electrochemical performance. A pulsed laser is applied to scan the electrode surface to fabricate patterned microchannels, enabling further mitigation of volume variation. Typically, the laser porosificated Si anode, as reported by Sämann et al., delivers a remaining capacity of >130 mAh g$^{-1}$ after 3000 cycles, 6.5 times higher than plain Si without laser treatment. It should be contributed by the laser porosification-generated pores, which mitigates the volume change induced mechanical stress and reduces cracks in the active layer over consecutive charge–discharge cycles.

### 3.1.2 Other batteries

In addition to the aforementioned common LIBs, other batteries, such as SIBs and ZIBs, are demonstrating more competitive merits as promising rechargeable batteries. Several PLD-fabricated nanomaterials have been utilized as
electrode materials with excellent performance for SIBs, such as $\gamma$-Fe$_2$O$_3$, SbSn-P, VO$_x$, hard carbon, SnO$_2$, and Na$_2$Ti$_6$O$_{13}$.

Typically, Zhou and coworkers reported the as-deposited SbSn-P thin films well-anchored on stainless steel (wavelength: 355 nm, repetition rate: 10 Hz, and laser energy: 2.5 J cm$^{-2}$), and direct application in SIBs as self-supported electrodes in the absence of any binders and conductive carbon.

The introduction of phosphorus into Sn–Sb alloy played a pivotal role in forming a stabilized solid electrolyte interphase film, reducing the charge transfer resistance and facilitating Na$^+$ diffusion in the electrode, thereby improving the electrochemical performance. Of particular note, the phase separation after the initial cycle may be beneficial for stabilizing the reversible alloying–dealloying reactions regarding Sn, Sb, and P during the subsequent cycles. Another example is PLD-derived P2-type Na$_{0.67}$Ni$_{0.25}$Mn$_{0.75}$O$_2$ thin films, which are fabricated by adjusting substrate temperature and postdeposition annealing and applied as a satisfactory cathode material for SIBs. Owing to the optimization in crystalline structure and enhanced stability of P2-O2 phase transition platform at about 4.2/4.1 V, the film-type P2-Na$_{0.67}$Ni$_{0.25}$Mn$_{0.75}$O$_2$ electrode displays high capacities, weak polarization, and outstanding cycling stability within a wide voltage window (1.5–4.3 V). Recently, Wang and coworkers constructed uniform and size-tailored BiVO$_4$ in acetone via LFL.

**FIGURE 7**  (A) Scanning electron microscopy (SEM), (B) TEM images, and (C) electrochemical performance of the pulsed laser deposition (PLD)-derived CoO-Co nanocomposite. Reproduced with permission from Reference 115 Copyright 2017, Elsevier. TEM and EDS mapping images of the discharged (D,E) and charged (F,G) amorphous GaN@Cu. (H) Rate capability and long-time performance of amorphous GaN@Cu. Reproduced with permission from Reference 126 Copyright 2019, Wiley-VCH. (J,L) Atomically resolved high angle annular dark field-scanning transmission electron microscopy (HAADF-STEM) images at Cu-Fe$_3$O$_4$ interface. (K) X-ray photoelectron spectrometer (XPS) of Fe 2p from Fe$_3$O$_4$ array. (L) Cyclic performances of Fe$_3$O$_4$ array and film at 5C. Reproduced with permission from Reference 118 Copyright 2020, Elsevier.
elaborate regulation of irradiation parameters (e.g., laser fluence and irradiation time) results in BiVO₄ with rich oxygen vacancies. Interestingly, the original yellow color of BiVO₄ itself turns to be black during LFL, which is ascribed to the contribution of oxygen defects. As anode materials for SIBs, the laser-generated BiVO₄@rGO results in a high reversible capacity (565 mA h g⁻¹ at 0.1 A g⁻¹), long-term cycling stability (470 mA h g⁻¹ after 200 cycles), and superior rate capability (297 mA h g⁻¹ at 2.0 A g⁻¹), benefiting from the nanosize-dependent fast kinetic characteristics and rich oxygen vacancies of laser-induced BiVO₄.

Appealing results are also found in simple, fast, and high precision laser engraving quasi-solid-state aqueous ZIBs. During the laser engraving process, a pulsed laser is applied to directly carve the freestanding films. Compared with traditional synthesis strategies, the laser engraving directly engrafted free-standing VO₂ (B)-multiwalled carbon nanotubes cathode and zinc nanoflakes anode films into finger-like electrodes. The assembled ZIBs exhibit superior flexibility, high-temperature stability, and electrochemical performance. The specific capacity, energy density, and power density are estimated to be as high as 314.7 μAh cm⁻², 188.8 μWh cm⁻², and 0.61 mW cm⁻², respectively. In particular, the specific capacity has no distinct recession with the temperature rising to 100°C and bending angle increasing to 150°.

### 3.2 Supercapacitors

Supercapacitors with high power densities, as an efficient energy storage system, have attracted considerable
interest worldwide for their potential application in ESCDs. Supercapacitors are commonly designed into a symmetrical configuration with two identical carbonaceous electrodes of high porosity and large surface area in aqueous/nonaqueous electrolytes, which is well known in carbon-based electrical doublelayer capacitors. If fabricated with TMCs or conductive polymers (CPs), which accomplish charge storage via quick reversible faradaic redox reactions occurring near the electrode surface, the supercapacitors can be called pseudocapacitors. Owing to their unique merits including simple and safe architecture, rapid charge/discharge response, high reliability, high power density, and long-term life span, supercapacitors have great feasibility to complement rechargeable batteries in certain fields. Nevertheless, the intrinsic low energy density primarily prevents their practical applications. Consequently, reasonable design and modulation of electrode materials to augment the charge storage capability turn out to be the primary topics for supercapacitors.

Thus far, diverse supercapacitive materials, such as carbon materials (CMs), TMCs, and CPs have been extensively surveyed. Although several articles have extensively reviewed the advances in electrode materials for supercapacitors, most of them are synthesized using conventional synthetic methods which are commonly time- and/or energy-consuming. As a simple yet high-efficiency materials-processing strategy, laser irradiation has been extensively utilized to regulate specific structures of electrode materials for advanced supercapacitors. A variety of laser-induced nanomaterials involved supercapacitors are summarized in Table 2.

Since the pioneering report by Kaner and coworkers in 2012 on the process of GO reduction by laser-scribing graphene membrane, the laser-induced graphene (LIG) has been applied in supercapacitors as a flexible self-supported film electrode. The optimal LIG film with a highly porous network is mechanically robust, exhibiting

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**Table 2** Summary of different laser-induced nanomaterials for supercapacitors

| Laser source               | Laser conditions                                      | Medium | Laser-induced nanomaterial          | Performance                   | Ref. |
|----------------------------|-------------------------------------------------------|--------|-------------------------------------|-------------------------------|-----|
| CO₂ laser system           | 10.6 μm, 14 μs, 4.8 W                                 | —      | LIG                                | >9 mF cm⁻², 0.02 mA cm⁻²      | 148 |
| Hybrid optical laser       | 1055–1075 nm, 1 W, 25 kHz                             | —      | Graphene nanoribbon fibers         | 234.8 F cm⁻³, 2 mV s⁻¹        | 151 |
| Femtosecond fiber laser    | 1030 nm, 400 fs, 120 kHz                              | —      | rGO/Au                             | 10.2 F cm⁻³, 100 V s⁻¹        | 152 |
| CO₂ laser cutter system    | 10.6 μm, 14 μs                                       | —      | LIG                                | 120 F g⁻¹, 20 mV s⁻¹          | 21  |
| CO₂ laser cutter system    | 10.6 μm, 14 μs                                       | —      | B-doped LIG                        | 16.5 mF cm⁻², 0.05 mA cm⁻²     | 153 |
| Nd:YAG laser               | 266 nm, 200 mJ cm⁻², 10 Hz                            | Ar or N₂, 20 Pa | N-doped rGO-NiO | 114 F cm⁻³, 10 mV s⁻¹      | 158 |
| CO₂ laser system           | 10.6 μm                                              | —      | Hollow Co₃O₄/LIG                    | 60.0 mC cm⁻²                   | 159 |
| CO₂ laser system           | 10.6 μm                                              | —      | LIG/ferrrocene                      | 178 F g⁻¹, 1 A g⁻¹             | 160 |
| CO₂ laser system           | 10.6 μm                                              | —      | LIG/Fe₂O₄                          | 719.28 mF cm⁻², 2 mV s⁻¹       | 161 |
| KrF excimer laser          | 248 nm, 300 mJ pulse⁻¹ cm⁻², 10 Hz                   | Water  | Oxygen-deficient TiO₂/LIG          | 14.1 Wh kg⁻¹, 8.5 kW kg⁻¹      | 26  |
| KrF excimer laser          | 248 nm, 400 mJ pulse⁻¹ cm⁻², 10 Hz                   | Water  | Oxygen-vacancy abundant Co₃O₄/LIG  | 978.1 F g⁻¹, 1 A g⁻¹           | 89  |
| KrF excimer laser          | 248 nm, 460 mJ pulse⁻¹ cm⁻², 10 Hz                   | Water  | Porous hollow MoO₃₃–doped MoS₂     | 73.4 μWh cm⁻², 240 μW cm⁻²     | 162 |
| CO₂ pulsed laser           | 10.6 μm                                              | —      | MoS₂-LIG                           | —                              | 163 |
| Nd:YAG laser               | 1064 nm, 8 ns, 10 Hz                                  | 10⁻⁴ Pa | Uniform MoS₂ with sulfur vacancy on carbon nanotube | 512 F g⁻¹, 1 A g⁻¹           | 164 |

Abbreviations: LIG, laser-induced graphene; YAG, yttrium aluminum garnet.
a large specific surface area (1520 m² g⁻¹) and a record conductivity (1738 S m⁻¹). The LIG assembled supercapacitors with gel electrolyte, give a power density of ~20 W cm⁻³, which is 20-fold higher than the activated carbon-based supercapacitors. Additionally, Tour’s group innovatively transformed commercial polyimide (PI) films into the vertically stacked porous graphene by facile laser induction.¹⁴⁸ When integrated into series and parallel connections, the PI-derived graphene-based device can render a working voltage window twofold higher than the single devices, exhibiting the ability of laser irradiation technology to construct integrated graphene-based supercapacitors. Furthermore, the in situ produced and/or modulated electrodes on current collectors by a laser-induced method, including carbon spheres-derived 3D graphene, laser-scribed activated carbon, and graphene nanoribbon fibers, also provide outstanding supercapacitive properties with large specific capacitance, long cycle life, and high rate capability.¹⁴⁹–¹⁵¹ The application of femtosecond (FS) lasers can afford exceptional device properties by using multiphoton polymerization, reduction, melting, and etching. Hu et al. explored a simple FS laser in situ reduction approach to render an integrated electrode (rGO/Au FS) by patterning both the rGO electrode and Au current collector (Figure 9(A,B)).¹⁵² In this way, the rGO electrode with superb conductivity of

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**FIGURE 9** (A) Scanning electron microscopy (SEM) and (B) HRTEM images of nanoporous structures of rGO/Au femtosecond (FS) electrodes. (C) Cyclic voltammetry (CV) curves and (D) stack capacitance Cₛ as a function of the scan rate of the rGO CW, rGO FS, rGO/Au CW, and rGO/Au FS at 100 V s⁻¹. Reproduced with permission from Reference 152 Copyright 2020, Royal Society of Chemistry. (E) Schematic of a B-laser-induced graphene (LIG)-microsupercapacitor (MSC) device. (F) Galvanostatic charge–discharge curves of LIG-MSC and B-LIG-MSC with different H₃BO₃ loadings. (G) Areal capacitance, (H) capacitance retention at different bending radii, and (I) CV curves at different bending cycles of 5B-LIG-MSC. Reproduced with permission from Reference 153 Copyright 2015, American Chemical Society.
charge/discharge processes. To date, numerous pseudocapacitive materials, including Fe3O4 NPs, nanostructured NiO, and hollow Co3O4 nanoboxes, have been incorporated into LIPCs to effectively regulate their pseudocapacitive behaviors.157-162 Kaner et al. presented a high-performance hollow Co3O4/LIG hybrid by encapsulating the hollow Co3O4 nanoboxes uniformly with a 3D macroporous LIG via a laser-scribing technique.159 As a consequence, the hollow Co3O4/LIG electrode exhibited a remarkable volumetric capacity of 60.0 C cm⁻³ (corresponding to 542.3 C g⁻¹) and excellent cycling stability. Another example is the hybrid with the Fe3O4 NPs uniformly anchored on LIG (LIG/Fe3O4) (Figure 10(A,B)).161 The resulting flexible in-plane hybrid MSCs using LIG/Fe3O4 and high capacitive LIG as the anode and cathode, respectively, show superior electrochemical behaviors in terms of areal capacitance (719.28 mF cm⁻²), energy density (60.20 μWh cm⁻³), prominent cycling stability, and excellent mechanical flexibility (Figure 10(C,D)). The superior electrochemical behaviors are primarily ascribed to the synergistic effects: LIG/Fe3O4 with hierarchical porous structure and dandelion-like Fe3O4 NPs for quick transport of ions and charge, the outstanding conductive network of LIG/Fe3O4 effectively accelerates charge transfer, the excellent wettability and prominent capillary behaviors in the water of LIG/Fe3O4 leading to good ion transfer reaction kinetics, and a reversible H⁺ ion (de)intercalation reaction with Fe3O4 NPs, thereby improving energy density.

Similar to doping treatments of CMs, defects can be introduced into lattices of TMCs, such as doping with heteroatoms and anion vacancies.28,163,164 The anion vacancies, which can reduce the valence state of metal ions and serve as electron reservoir during the charging/discharging process, are positively charged, facilitating the augment in electron density of donors. In this regard, Cao and coworkers reported oxygen-vacancy-abundant ultrathin Co3O4 NPs/graphene (UCNG) via LFL (Figure 10(E)).49 As confirmed by electron paramagnetic resonance absorption spectroscopy and density functional theory calculations (Figure 10(F)), the UCNG hybrid possessed more Co²⁺ in Co3O4 NPs, further demonstrating the existence of oxygen vacancies, which should be ascribed to the reducing action by using electrons produced in the reaction system. Remarkably, the UCNG hybrid maintains 99.3% of the initial capacitance after continuous 20 000 cycles and 93.7% of capacitance retention with the current density increasing from 1 to 10 A g⁻¹ (Figure 10(G)), featuring its outstanding cycling stability and rate capability for supercapacitors.

3.3 Electrocatatysis

Electrocatalysis refers to the heterogeneous catalysis process that can boost electrochemical reactions on the
surface of electrocatalysts. In general, the energy-related application in electrocatalysis is the water cycle which involves a sequence of hydrogen- and oxygen-related electrochemical processes, including water splitting and fuel cells (FCs). In regard to water splitting, the involved hydrogen evolution reaction (HER) and oxygen evolution reaction (OER) generate H₂ and O₂, respectively. For hydrogen-oxygen FCs, power can be produced through oxygen reduction reaction (ORR) and hydrogen oxidation reaction. However, the innate sluggish kinetics seriously restricts the properties of corresponding energy devices on account of multiple-electron transfer processes in electrocatalytic reactions. As a consequence, appropriate electrocatalysts that can radically accelerate electrocatalytic reactions have been intensively explored. Recently, the laser irradiation technique has been confirmed as a powerful tool to modulate the electrocatalytic materials for enhanced catalytic performance, including structural defects, heterostructures, and novel electrode design (Table 3).

The PLD approach has been widely utilized to fabricate high-performance nanomaterials for electrocatalysis, facilitating improved electrocatalytic performance. As a typical example, Tour et al. irradiated pinewood with high lignin content into hierarchical porous LIG with excellent electrical conductivity (10 Ω per square) using CO₂ laser scribing (Figure 11(A)). The depth of pine-derived LIG (P-LIG) with a few-layered structure
was about 800 μm embedded in wood (Figure 11(B,C)). The fabricated P-LIG-Co-P by depositing active Co-P onto the P-LIG manifests an HER current density of 62 mA cm⁻² at a low overpotential of 200 mV along with a Tafel slope of 35 mV dec⁻¹ (Figure 11(D,E)). Recently, it has aroused focused attention to increase the electrocatalytic performance of TMOs by introducing structural defects, such as vacancy manipulation and elemental doping. The cation/anion vacancies via the PLD technique are a primary factor that regulates electrocatalytic performances of laser-induced nanomaterials. For instance, Chen’s group fabricated rich oxygen vacancies NiO thin film with well-controlled thickness and surface structure through the laser-mediated process, coupled with subsequent Ar plasma treatments.  

### TABLE 3 Summary of different laser-induced nanomaterials for electrocatalysis

| Application | Laser source | Laser conditions | Medium | Laser-induced nanomaterial | Performance | Ref. |
|-------------|--------------|------------------|--------|---------------------------|-------------|------|
| OER         | KrF excimer laser | 248 nm, 2 J cm⁻², 10 Hz | Oxygen, 25 mTorr | TiN₂O₅ thin films | 290 mV, 10 mA cm⁻²; 57 mV dec⁻¹ | 171 |
| HER and OER | CO₂ laser system | 10.6 μm, 14 μs (75 W), 394 pulses cm⁻¹ | Ar/H₂ | P-LIG-Co-P, P-LIG-NiFe | P-LIG-Co-P, HER, 200 mV, 62 mA cm⁻²; 35 mV dec⁻¹; P-LIG-NiFe, OER, onset overpotential 270 mV, 78 mV dec⁻¹ | 172 |
| OER         | — | 300 mJ pulse⁻¹, 5 Hz | Oxygen, 1 Pa | Oxygen vacancies rich NiO | 9.76 mA cm⁻² at 1.7 V | 173 |
| HER         | KrF excimer laser | 248 nm, 3.1 J cm⁻² pulse⁻¹, 20 Hz | Ar | Oxygen-incorporated amorphous MoSₓ | 170 mV, −100 mA cm⁻²; 35.1 mV dec⁻¹ | 174 |
| OER         | KrF excimer laser | 248 nm, 2.2 J cm⁻² pulse⁻¹, 10 Hz | Oxygen, 300 mTorr | (001) LaNiO₃ on SrTiO₃ substrates | >150 μA cm⁻² at 1.63 V | 177 |
| OER         | KrF excimer laser | 248 nm, 6 J cm⁻², 20 Hz | Oxygen, 100 m Torr | (100) IrO₂ on a (100) SrTiO₃ substrate | 81 μA cm⁻² at 1.53 V, 63 mV dec⁻¹ | 178 |
| Electrocatalyst for HER | Nd:YAG laser | 1064 nm, 95 μJ, 100 kHz | Acetone | NiFeₓCy encapsulated in ultrathin graphene | 48 mV dec⁻¹ | 179 |
| Electrocatalyst for OER | Nd:YAG laser | 1064 nm, 6 × 10⁶ W cm⁻² | Water | Co₃O₄ NPs | 294 mV, 10 mA cm⁻²; 74 mV dec⁻¹ | 180 |
| OER         | Nd:YAG laser | 355 nm, 8 ns, 210 mJ pulse⁻¹, 10 Hz | Water | Ti³⁺ and La³⁺ co-doped NiFe LDH | 260 mV, 10 mA cm⁻² | 181 |
| ORR and OER | Nd:YAG focused beam laser | 1064 nm, 700 mJ, 15 kHz | Water | Rich vacancies Co₃O₄ ultrasmall NPs | ORR: half-wave potential of 0.878 V, 47 mV dec⁻¹; OER: 271 mV, 10 mA cm⁻², 42 mV dec⁻¹ | 182 |
| OER         | Nd:YAG laser | 532 nm, 10 ps, 500 μJ | Ethanol | Rich defect CoO NPs | 369 mV, 10 mA cm⁻²; 46 mV dec⁻¹ | 183 |
| ORR and OER | Nd:YAG laser | 1064 nm, 7 ns, 270 mJ, 15 Hz | Ethanol | NiCo₂O₄/pyridinic-N-doped mesoporous graphene | ORR: half-wave potential of 65 mV; OER: 40 mV, 10 mA cm⁻² | 184 |
| OER         | Nd:YAG laser | 1064 nm, 7 ns, 200 mJ, 15 Hz | Water | NiO/NiFe LDH | 205 mV, 30 mA cm⁻²; 30 mV dec⁻¹ | 185 |

Abbreviations: HER, hydrogen evolution reaction; LDH, layered double hydroxide; LIG, laser-induced graphene; NPs, nanoparticles; OER, oxygen evolution reaction; ORR, oxygen reduction reaction; YAG, yttrium aluminum garnet.
Oxygen vacancies can be incorporated into the NiO thin films by inert Ar plasma treatment. The utilization of Ar plasma treatment can avert complications resulting from the anion doping. When evaluated as an electrocatalyst for OER, the current density at 1.7 V (vs. RHE) for the NiO thin film subjected to 50 W Ar plasma treatments was 9.76 mA cm\(^{-2}\), exhibiting 5.8-fold improvement compared with that for the pristine NiO film. In addition, Fonzo et al. developed a high nonequilibrium synthesis technique using PLD, that is, the disorder engineering approach, to produce a high-performance oxygen-incorporated amorphous molybdenum sulfide with a metastable structure as an HER electrocatalyst.\(^{174}\) The disorder engineering method can produce highly defective clusters with a vast range of active sites, and self-assemble them in arrays of quasi-one-dimensional hierarchical nanostructures, which guarantees an increased surface area and fast electronic/ionic transport. The optimized hierarchical oxygen-incorporated amorphous MoS\(_x\) electrocatalyst delivered extremely small overpotentials (170 mV at \(-100\) mA cm\(^{-2}\)), superb kinetics...
Apart from these common laser-induced syntheses of electrocatalysts, atomic epitaxial growth on an appropriate substrate has also been designed and developed.\(^{175,176}\) Chung's group reported the influence of both tensile and compressive strains on the generation behaviors of Ruddlesden–Popper (RP) two-dimensional planar faults in (001) epitaxial LaNiO\(_3\) thin films by using two distinct substrates of LaAlO\(_3\) and SrTiO\(_3\), as illustrated in Figure 11(F,G).\(^{177}\) The number of RP faults for SrTiO\(_3\) substrate is about a two times higher than that for the case using the LaAlO\(_3\), indicating that the RP-fault formation can be improved for the LaNiO\(_3\) film under a tensile strain field. Meanwhile, even fewer misfit dislocations are observed in the film on SrTiO\(_3\) than those in the film on LaAlO\(_3\), thus confirming that the effective lattice constant of a LaNiO\(_3\) film is enlarged with the generation of RP faults. In particular, as symmetry-breaking lattice defects, the RP faults in LaNiO\(_3\) are particularly active sites for OER. The film on SrTiO\(_3\) gives a current value approximately threefold higher (>150 \(\mu\)A cm\(^{-2}\) at 1.63 V) than that for the fault-free (001) LaNiO\(_3\) film. Besides, the PLD technique was also used for heteroepitaxial growth of (100) IrO\(_2\) on a (100) SrTiO\(_3\) substrate (wavelength: 248 nm; pulse width: 17 ns; repetition rate: 20 Hz; and laser fluence: 6 J cm\(^{-2}\)).\(^{178}\) The heteroepitaxial growth resulted in an orthorhombic distortion of the tetragonal structure of IrO\(_2\) due to the symmetry mismatch. Accordingly, the [IrO\(_6\)] geometry was distorted, thus contributing to an improved electrocatalytic activity of the (100) epitaxial IrO\(_2\) film.

As a booming laser-induced nanomaterial synthesis technique, LIL has been applied to generate electrocatalysts for enhancing electrocatalytic performances.\(^{179–181}\) However, anion vacancies of irradiated material through LIL are primarily generated on the surface. To introduce interior anion vacancies facilitating fast electron transport, Du et al. and Tüysüz et al. proposed the combination of LIL
with chemical oxidation/reduction reactions. The pulsed laser (wavelength: 1064 nm; frequency: 15 Hz; energy: 700 mJ) resulted in vaporization of the Co target, producing Co vapor. The thus-obtained Co vapor further reacted with dissolved oxygen in the water to generate ultrathin (about 2.1 nm) laser-induced Co$_3$O$_4$ NPs possessing rich surface and interior oxygen vacancies (Figure 12(A)). The synergistic contributions from ultrasmall size, exterior/interior oxygen vacancies guaranteed abundant active catalytic sites, fast electronic transport, and intrinsic activities, favoring superb electrocatalytic performances for both ORR and OER. As for the ORR activity (Figure 12(B)), laser-induced Co$_3$O$_4$ with rich oxygen vacancies possessed a half-wave potential of 0.878 V, even outperforming the commercial Pt/C catalyst (20 wt% Pt). For OER (Figure 12(C)), it afforded an overpotential of 271 mV at 10 mA cm$^{-2}$ along with a Tafel slope of 42 mV dec$^{-1}$.

Except for structural defects, the synergistic contribution from different electrocatalysts via LIL, such as NiCo$_2$O$_4$/graphene, NiO/NiFe layered double hydroxide, and LIG-Co-P, contributes to an enhanced electrocatalytic properties. Recently, Yang et al. synthesized the NiCo$_2$O$_4$/N-doped mesoporous graphene NSs hybrids (wavelength: 1064 nm; pulse width: 7 ns; frequency: 15 Hz; energy: 270 mJ), in which the relative concentrations of pyridinic and pyrrolic nitrogen dopants were precisely regulated (Figure 12(D)). More significantly, the pyridinic-N-Co bonds at the NiCo$_2$O$_4$/mesoporous graphene interface primarily facilitated synergistic catalytic effect, thereby improving both the ORR and OER activities of hybrids. The NiCo$_2$O$_4$/N-doped mesoporous graphene offered a half-wave potential of 65 mV for the ORR process, as well as an overpotential of 40 mV at 10 mA cm$^{-2}$ for the OER (Figure 12(E,F)), outperforming the benchmark OER catalyst RuO$_2$.

4 | CONCLUSION AND OUTLOOK

To build a global energy interconnection and meet power demand worldwide, effective use of renewable energy, which demands high-performance ESCDs, is hugely stimulated. It plays a significant role in the realization of carbon-c crest and carbon neutralization in the shortest time. The ever-growing interest in novel energy storage materials and laser irradiation techniques has witnessed the increasing concerns recently for laser-involved synthesis, structures, and surface/interface regulation of nanomaterials toward ESCDs. This review mainly focused on the recent research progress in rational design and controllable fabrication of laser-induced nanomaterials and their promising applications in rechargeable batteries, supercapacitors, and electrocatalysis. Up to now, a mass of highly active materials have been elaborately tuned at atomic-, nano-, and/or macroscales into the desired structures through diverse laser irradiation approaches, including PLD, LAL, LFL, and LML, achieving striking enhancements in electrochemical properties for advanced ESCDs. In spite of these achievements in LIBs and SIBs, the laser irradiation synthesized nanomaterials have few applications for other rechargeable batteries, such as potassium-ion batteries, aluminum-ion batteries, lithium-sulfur batteries, MABs, and so on. Further exploration of advanced laser-induced nanomaterials with superior electrochemical behaviors for other rechargeable batteries is still urgently needed.

Although prominent advances in laser irradiation techniques, investigations in the flourishing topic remain in a preliminary stage and several huge challenges need to be resolved. It is worth noting that a majority of researchers in the past few years are inclined to put more endeavors into fabricating laser-induced electrode materials with remarkable merits, including abundant structural defects, porous structure, superior electronic conductivity, and so on, to construct advanced ESCDs. Nevertheless, the precise mechanisms are still under debate, even though the photothermal evaporation effect, Coulomb explosion effect, and/or their synergistic contribution are put forward to elucidate the particular material processing. As a consequence, the huge difference in the formation mechanisms has greatly hindered more comprehensive structure regulation of laser-induced nanomaterials in a reasonable way. For this, a thorough and in-depth understanding of the involved mechanisms by using several advanced in situ techniques (in situ synchrotron radiation X-ray diffraction, X-ray absorption fine structure, and in situ XPS, etc.) is even more urgent and meaningful. Currently, the contributions in dynamically forming processes of materials and corresponding theoretical simulations, which offer an insightful point of view for laser irradiation techniques, are not always enough. Besides, the cogent cognition in the underlying relationships between structures and electrochemical performance should have further light shed on them.

Typically, laser irradiation, as a fast synthesis avenue, is utilized to prepare electroactive materials with relatively simple experimental parameters. However, it suffers from low production yield and relatively high cost-efficiency. Thus, laser irradiation synthesis of electrode materials is mainly applicable to laboratory synthesis rather than large-scale industrial applications currently. To meet industrial demands and open the avenue to further commercialization, the electrospark granule deposition (EGD) technology (Figure 1(A)) as a pulsed deposition process would be an appropriate choice. The highlighting characteristics of the EGD approach lie in its simplicity, high adhesion to an
appropriate substrate, epitaxial growth on the substrate, and low cost of the involved equipment. More competitively, EGD configuration is more productive and allows automatic deposition, thus greatly facilitating the mass and automated production of active materials. We strongly believe that more high-performance materials can be rationally designed through EGD, and even other new industrial technologies in the near future toward more practical ESCDs.

Despite the research history of the laser-induced nanomaterials for ESCDs based on electrochemistry being very short, a rapid and sustained development has occurred in recent years. Any further breakthrough in laser technology will stimulate the rapid development of multifunctional nanodimensional materials toward next-generation ESCDs. Although the aforementioned challenges still exist, we firmly believe that the increasing numbers of studies and rapid evolution of this field are greatly encouraging and expected to surmount current disadvantages. Overall, fruitful advancements have been obtained in the applications of laser-induced nanomaterials for ESCDs, and they hold great promise in other booming energy-related fields as well. We really expect that this review will afford in-depth understanding and inspiration on the future precise design of advanced laser-induced nanomaterials with substantially improved performance for energy storage and conversion, and beyond.

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CONFLICT OF INTEREST
The authors declare no conflict of interest.

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