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Perspective

Laser ablation in air and its application in catalytic water splitting and Li-ion battery

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SUMMARY

Pulse laser has been widely used in both fundamental science and practical technologies. In this perspective, we highlight the employment of pulse laser ablation in air (LAA) in energy-related catalytic reactions. With LAA, samples are directly ablated in ambient air, which makes this technology facile to conduct. Materials can be modified by LAA in multiple aspects, such as morphology modulation, heterojunction fabrication, or defects engineering, which are desired features for energy-related catalytic reactions. We begin this perspective with a brief introduction of this technology, including the mechanism, the experimental setup, and the characteristic of laser-ablated materials. The recent works utilizing LAA are then summarized to prove the promising prospects of LAA in the energy field. Finally, several opportunities about the future usage of LAA are proposed and discussed.

INTRODUCTION

The development of human civilization in the past centuries is based on the exploitation of fossil fuels, which has created vital crisis like global warming and environment pollution (Zhang et al., 2020; Wang et al., 2021a). And with the rapid expansion of human society, the fossil fuels are in more and more urgent shortage. Upgrading the energy section has been proposed years ago, however, with moderate progress, due to economic considerations. Thus, developing cheap, green, and renewable energy sources has been a challenging but pressing task. Meanwhile, the development of energy materials is of equal importance, which helps the harvest, conversion, and storage of clean sources. Pulse laser has been widely used in time-resolved characterization methods, such as transient absorption or fluorescence spectrum, which facilitates greatly the development of fundamental science (Tien et al., 1999; Zhao et al., 2019, 2020). Meanwhile, in the field of nanomaterial fabrication, pulse laser has been also adopted broadly in a technology termed as laser ablation (Zeng et al., 2012; Li et al., 2016). Although tremendous works of laser ablation have been reported, the adoption of this technology on energy-related applications is somehow underestimated.

Conventionally, laser ablation is conducted in liquids, which can be termed as LAL. Various nanomaterials have been prepared using the laser ablation method, such as nanowires (Xu et al., 2018; Kim et al., 2018; Teixeira-Santos et al., 2021), nanoparticles (Nguyen et al., 2021; Lim et al., 2018; Lin et al., 2020), or clusters (Xu et al., 2019; Li et al., 2019a). Usually, the solution could play an important role in LAL. For example, CdTe nanowire arrays were fabricated in liquid in a work (Chen et al., 2016), where oleic acid was adopted and played an important role. In another example (Huang et al., 2015), CdSe nanowire arrays were fabricated with the aid of oleic acid and tri-n-octylphosphine. Some energy materials have been prepared by LAL, such as niobium pentoxide for Li-ion battery (Liang et al., 2020) or silver stacking faults for hydrogen evolution (Li et al., 2019b). Also, Wang et al. reviewed the recent applications of LAL on functional micro-supercapacitors (Wang et al., 2021b). Nanomaterials that are fabricated by LAL exhibit excellent homogeneity in size, morphology, and other aspects. However, the experimental setup and product collection process can be extremely complicated. For energy materials, excellent homogeneity, which costs additional price and fabrication complexity, sometimes is not necessary. Moreover, materials with core-shell morphology and heterojunction structure cannot be produced by LAL, which could be important in energy field. Therefore, a similar but much simpler and faster approach is adopted by our group to fabricate energy materials, that is, laser ablation in air (LAA).

Distinguished from LAL, where the laser ablation is performed in selected solutions, LAA is conducted directly in ambient air condition. Nanomaterials or clusters are the final products in LAL, which are

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Table 1. Comparison of LAL and LAA

Terms	LAL	LAA
Experimental setup	Complicated	Simple
Technological route	Separate products from raw targets	Add modifications on raw materials
Solution needed	Yes	No
Surface passivation agent needed	Yes	No
Target material	Metals or alloys	Solid materials
Product homogeneity	Excellent	Moderate
Collection after ablation	Complicated	Not needed

separated from a raw target. However, in LAA, the final products are not separate parts of raw materials but more like special modifications of the raw materials. A table that summarizes the characteristics of LAL and LAA is shown in Table 1. Furthermore, versatile material can be manipulated by LAA, including metals, alloys, oxides, sulfides, and so on. Actually, there are no specific limitations on the category of solid material that is suitable for LAA treatment. As long as the optical power is appropriate and accessible, all solid materials should exhibit notable characterization changes after LAA treatment. It is amazing to find that considerable enhancement on the activity of materials can be achieved by LAA treatment for just several minutes. We believe LAA has a great potential applying in the energy-related industry. In this perspective, we will first give a straightforward introduction of the LAA technology, with emphasis on the mechanism analysis, the characteristic of laser-ablated materials, and the operation details. Then, we will summarize the most recent works applying laser ablation in energy-related catalysis. We finally put forward the perspective that laser ablation will open a new way to regulate and control catalytic materials and trigger subsequent research.

LASER ABLATION IN AIR

Mechanism

The modification of materials by LAA can be divided into two steps. First, when illuminated by pulse laser, a small part of the material near the surface absorbs the photon energy, experiencing heating effect of the pulse, and finally decomposes into free atoms or ions. The pulse heating effect on materials can be further divided into three steps: the light energy harvest, the energy transportation through electrons collisions, and the heat transfer by electron-phonon interactions. Second, after the pulse duration, the free atoms cool down with extremely rapid rate due to the large temperature gradient. Active atoms aggregate with each other and re-deposit as tremendous ultra-small spheres on the surface of the material. Meanwhile, the melting part of the material could form defects due to the loss of substance. In general, a thin layer near the surface of materials decomposed and re-deposited within a timescale of several nanoseconds.

Experimental setup

The experimental setup of a typical laser ablation in air process is schematically illustrated in Figure 1A, which is quite simple and easy to conduct. A pulse laser beam is directed to a platform carrying the desired samples. As one can see, the sample is exposed in ambient atmosphere without any complicated protection. One may concern that the sample will be oxidized during the ablation process. As discussed above, only a small part of sample experiences oxidation due to the fact that the ablation is extremely fast. In fact, in some cases, oxides can be reduced after the ablation.

Characteristic of LAA-treated materials

Samples treated by laser ablation in air exhibit versatile characterizations according to the specific materials, as illustrated in Figure 1B. For example, some samples experience morphology modification, forming most commonly a core-shell structure. The core-shell morphology could increase the specific area of a sample, facilitating the active sites loading for energy-related reactions. Some materials go through phase transition during the ablation, forming heterojunction structures. The decompose-redeposit process within nanoseconds is inherently a process of fast non-equilibrium crystallization. Thus, versatile phases could be preserved and form heterojunctions. Heterojunctions can regulate the band structure and properties of a

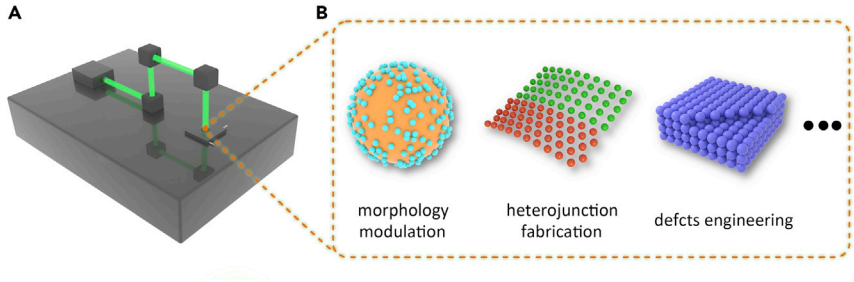

Figure 1. Influence of laser ablation on materials

(A) Experimental setup for laser ablation in air.

(B) Potential modifications of laser ablation on materials.

material, boosting light absorption and electron transportation in photocatalytic applications. Defects can also be generated in some samples. The atoms decomposed from a specific part of a material can probably aggregate and re-deposit on another part of the material, leaving the original part defects. Defects can be surprisingly favorable for energy-related catalytical applications, such as electrocatalytic hydrogen evolution, oxygen evolution, and so on.

APPLICATION OF LAA IN ENERGY-RELATED CATALYSIS

LAA could find application in many aspects, as illustrated in Figure 2. H_2 is an ideal energy source without pollution. In practice, the production of H_2 is driven by photocatalytic water splitting (Wang et al., 2021a; Hu et al., 2019), electrocatalytic water splitting (Li et al., 2019c; Wang et al., 2021c; Zhong et al., 2020, 2021a), and traditional steam reforming. Among them, photocatalytic water splitting can directly utilize solar energy, while electrocatalytic water splitting is featured with high efficiency and time continuity. Electrical water splitting can be divided into two half reactions, the hydrogen evolution reaction (HER) and the oxygen evolution reaction (OER), with the latter being the bottleneck. We found laser ablation in air exhibited itself a promising method to low the barrier of OER. Meanwhile, due to the versatile characteristics of the LAA products, photocatalytic water splitting could also be benefited. The storage of clean energy sources into electricity is another important route, where LAA could also play a role.

Application in electrocatalytic water splitting

In a previous work, we demonstrated that the morphology and conductivity of IrO_2 (a benchmark material for OER) can be improved by laser ablation in air (Zhong et al., 2019a). As illustrated in Figure 3A, under illumination by the pulse laser, IrO_2 first decomposes due to the heating effect, generating free Ir and O atoms. After the nanoseconds' duration of the pulse, hot Ir and O atoms cool down and recrystallize. Active O atoms aggregate with each other and escape as O_2. Heavier Ir atoms aggregate and deposit as ultra-small Ir spheres on the surface of IrO_2, forming a core-shell structure. scanning transmission electron microscope images shown in Figure 3B clearly demonstrate the core-shell morphology. Particularly, twin-crystal Ir spheres are observed, indicating the harsh and fast laser ablation procedure. With increased specific area originated from the core-shell structure and improved electrical

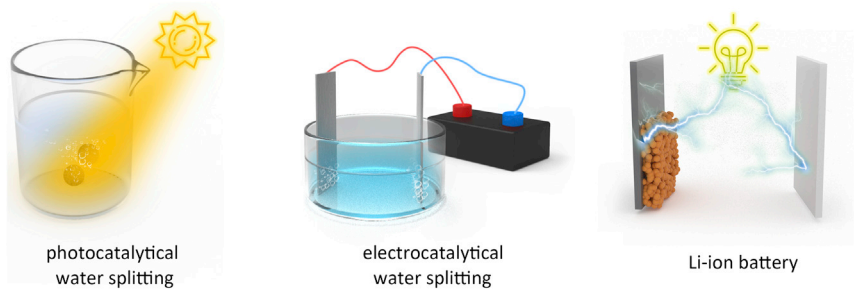

Figure 2. Potential applications of LAA

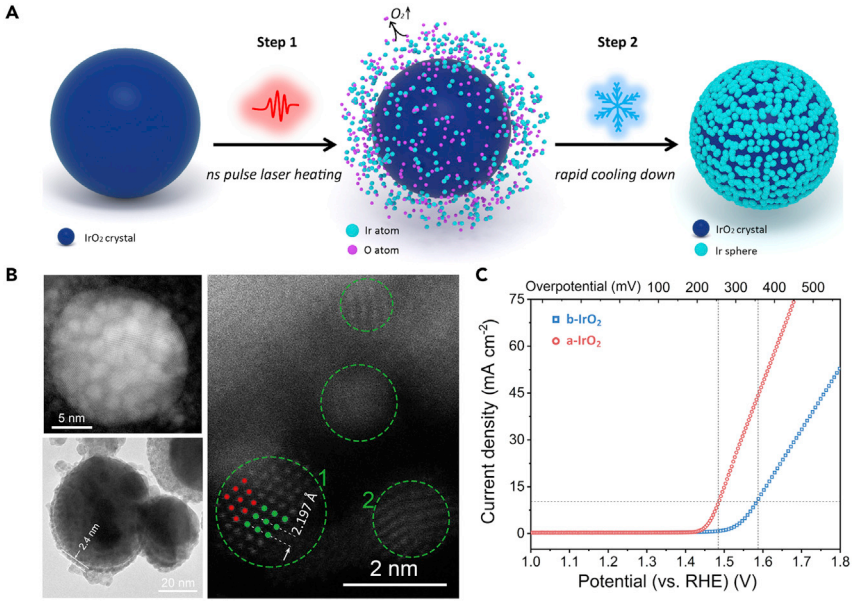

Figure 3. IrO_2@Ir electrocatalyst for OER

(A) Schematic illustration of sample fabrication.

(B) Scanning transmission electron microscope images of the Ir@IrO_2 sample.

(C) Electrochemical overpotential for OER.

Figures adopted from (Zhong et al., 2019a) with permission.

conductivity originated from Ir spheres, IrO_2@Ir performs much better than IrO_2, as depicted in Figure 3C. Specifically, the as-prepared IrO_2@Ir catalyst by laser ablation in air exhibits an overpotential of 255 mV at 10 mA cm^{-2} and a Tafel slope of 45 mV dec^{-1}.

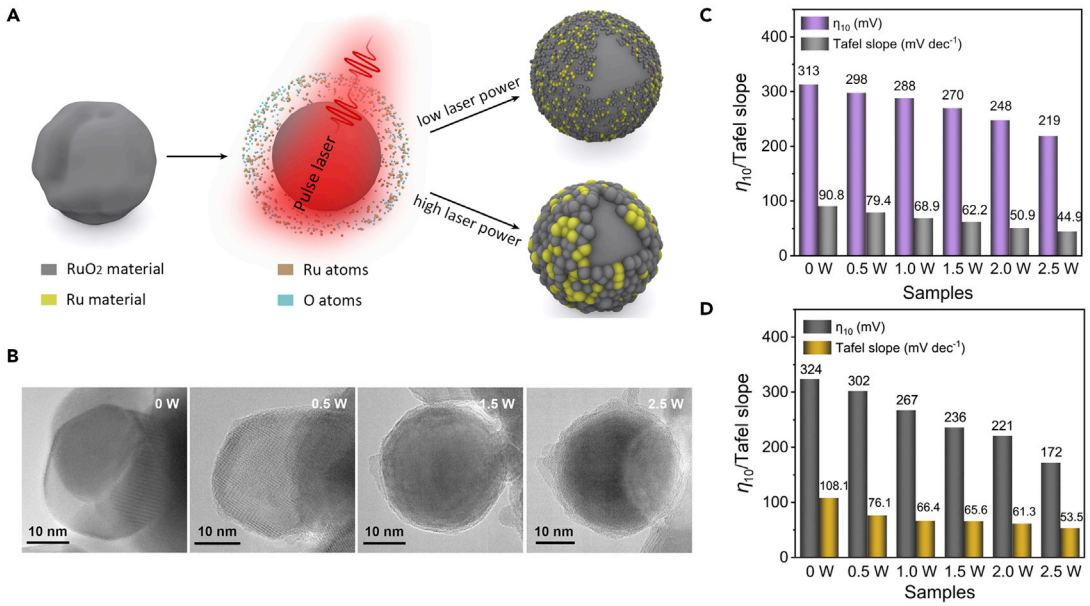

Figure 4. Lychee-shaped RuO_2@Ru/RuO_2 electrocatalyst for OER

(A) Schematic illustration of morphology modulation by laser ablation.

(B) TEM images of lychee-shaped RuO_2@Ru/RuO_2 treated using various optical powers.

(C) Electrochemical performance for OER in acidic electrolyte.

(D) Electrochemical performance for OER in alkaline electrolyte.

Figures adopted from (Wang et al., 2021d) with permission.

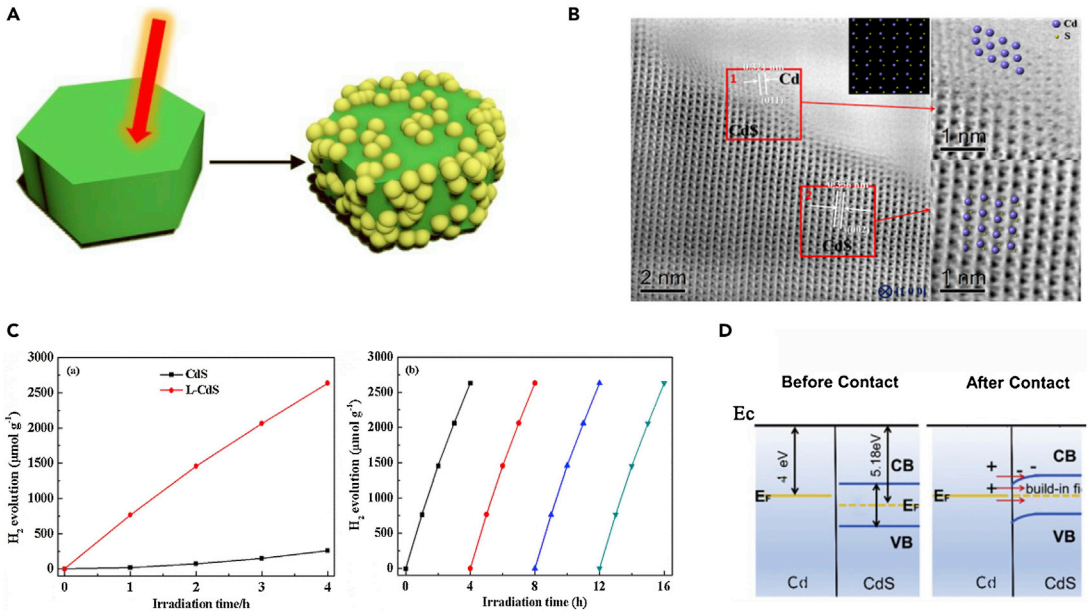

Figure 5. Cd/CdS Schottky junction for photocatalytic hydrogen production

(A) The process of laser irradiation for preparation of Cd/CdS Schottky junction.

(B) TEM image of Cd/CdS Schottky junction.

(C) H_2 evolution of CdS and L-CdS, cycling measurements of L-CdS.

(D) Band structure illustration before and after contact.

Figures adopted from (Zhong et al., 2019b) with permission.

In another work, we show the core-shell morphology can be regulated by the ablation power (Wang et al., 2021d). OER in acidic electrolyte is of particular importance since HER performs well in proton-exchange membrane systems. However, the activity of most OER electrocatalysts in acid is poor. RuO_2 is one of the best electrocatalysts that work in acid. The OER activity can be largely improved by laser ablation in air. Similar to the above work, lychee-shaped core-shell morphology is formed by laser ablation in air, as illustrated in Figure 4A. Further analysis reveals that the shell is composed of small RuO_2 or Ru spheres, and the thickness of the shell can be well regulated by the optical power of the pulse laser. It is found that the higher the ablation power is, the more Ru/RuO_2 spheres will be generated and the thicker the shell will be, as shown in Figure 4B. The electrocatalytic OER performance of the lychee-shaped RuO_2@Ru/RuO_2 is evaluated in both acid and alkaline electrolytes, as shown in Figures 4C and 4D, respectively. One can see that the OER performance is largely boosted and can be well regulated by the ablation power.

Application in photocatalytic hydrogen evolution

The separation of electron-hole pairs is crucial in photocatalysis. Fabricating heterojunctions is a promising route to hinder the recombination of electron-hole pairs. We reported that the Cd/CdS Schottky junction induced by laser irradiation has excellent photocatalytic hydrogen production performance (Zhong et al., 2019b). Figure 5A shows the laser ablation process, where Cd nanoparticles are formed *in situ* on the surface of CdS. The transmission electron microscope (TEM) image clearly shows the interface between Cd and CdS (Figure 5B). Both the (011) crystal plane of Cd and the (002) crystal plane of CdS can be observed. After forming the Schottky junction, the photocatalytic hydrogen evolution performance of the sample has been greatly improved. The hydrogen evolution rate under visible light has increased by nearly 40 times (Figure 5C). In addition, the cycle test shows that the sample has excellent stability (Figure 5C). Figure 5D shows the schematic diagram of energy band structure before and after the Schottky junction is formed. Due to the difference in work function between Cd and CdS, a built-in electric field is generated at the interface. Photogenerated carriers can be quickly separated and transferred under the action of the built-in electric field, thereby greatly optimizing the photocatalytic performance.

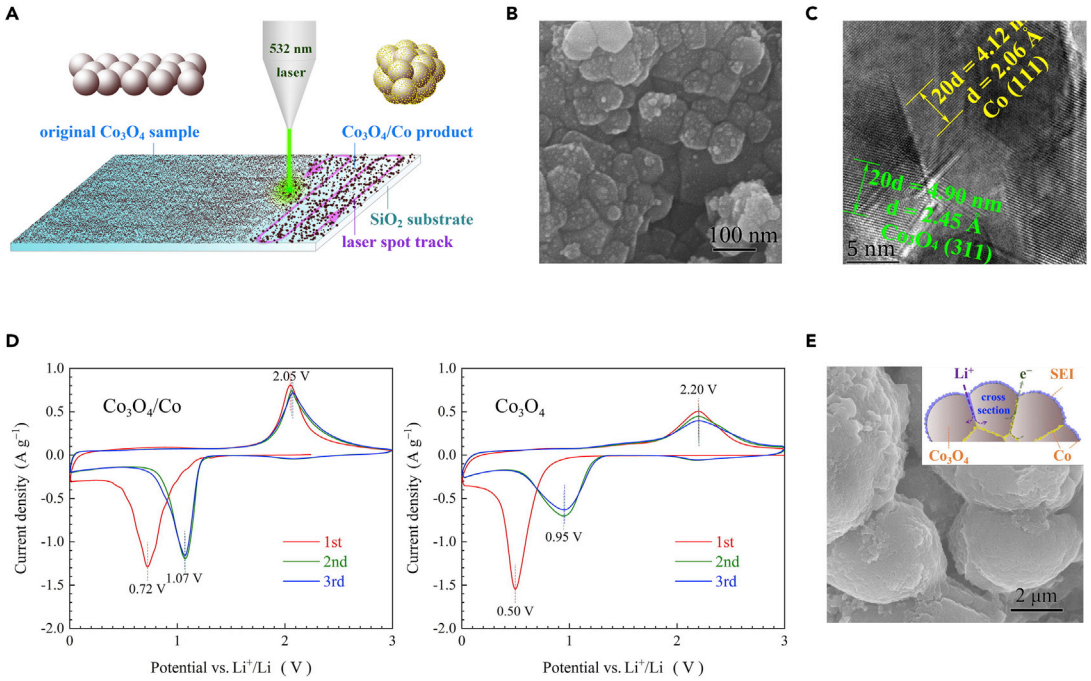

Figure 6. Application of Co_3O_4/Co in lithium-ion battery

(A) The process of laser irradiation for preparation of Co_3O_4/Co.

(B and C) SEM image and TEM image of Co_3O_4/Co.

(D) Discharge-charge curves of Co_3O_4/Co and Co_3O_4.

(E) SEM image of Co_3O_4/Co after 100 cycles. The inset shows the charge transport paths.

Figures adopted from (Zhong et al., 2021b) with permission.

Application in Li-ion battery

Laser ablation technology also has potential application in lithium-ion batteries. Zhong et al. reported the preparation of anode material Co_3O_4/Co nanocomposites by laser ablation (Zhong et al., 2021b). The preparation process is shown in Figure 6A. First, the original Co_3O_4 is evenly dispersed on the surface of silica glass. Then, it is vertically scanned and irradiated by a 532-nm pulsed laser. Then, compact Co_3O_4/Co nanocomposite was formed. As shown in Figure 6B, well-dispersed small nanoparticles are generated *in situ* on the surface of the original large particles. TEM image confirms the (111) crystal plane of generated Co (Figure 6C). The first discharge capacity and charge capacity of the Co_3O_4/Co electrode are 1115 mAh g^{-1} and 885 mAh g^{-1}, respectively (Figure 6D). Its initial coulombic efficiency is 79.4%, which is significantly better than that of the Co_3O_4 electrode (63.2%). After 100 cycles of cycles, the approximate spherical shape was maintained (Figure 6E), indicating excellent stability. In addition, the conductive network formed by Co nanoparticles on the surface of Co_3O_4 particles can improve conductivity. The small cavities provide transport channel for lithium ions and electrolyte. The above factors optimized the electrochemical activity.

In another example, micro holes on copper foil are fabricated in large scale utilizing LAA method to achieve more stable current collectors for lithium batteries (Li et al., 2020). Current collectors exhibit high importance in Li-ion batteries, which not only carry the electrode material but also collect electrical current when the battery functionalizes. Formulating porous microstructure can increase the area and thus boost the performance of collectors. Conventionally, it is extremely hard to prepare micronano holes on Cu foil current collectors, hindered by the ultrathin and soft nature of the material. Surprisingly, this problem can be well addressed by the LAA approach. As demonstrated in Figure 7, using a facile experimental setup, Li et al. were able to fabricate 250 000 micropores on a 2 cm × 2 cm Cu foil with well-defined circle shape and good hole quality. Benefitted from this microporous morphology, the collector operates with a more stable response than its unmodified counterpart at both cycle performance and rate performance.

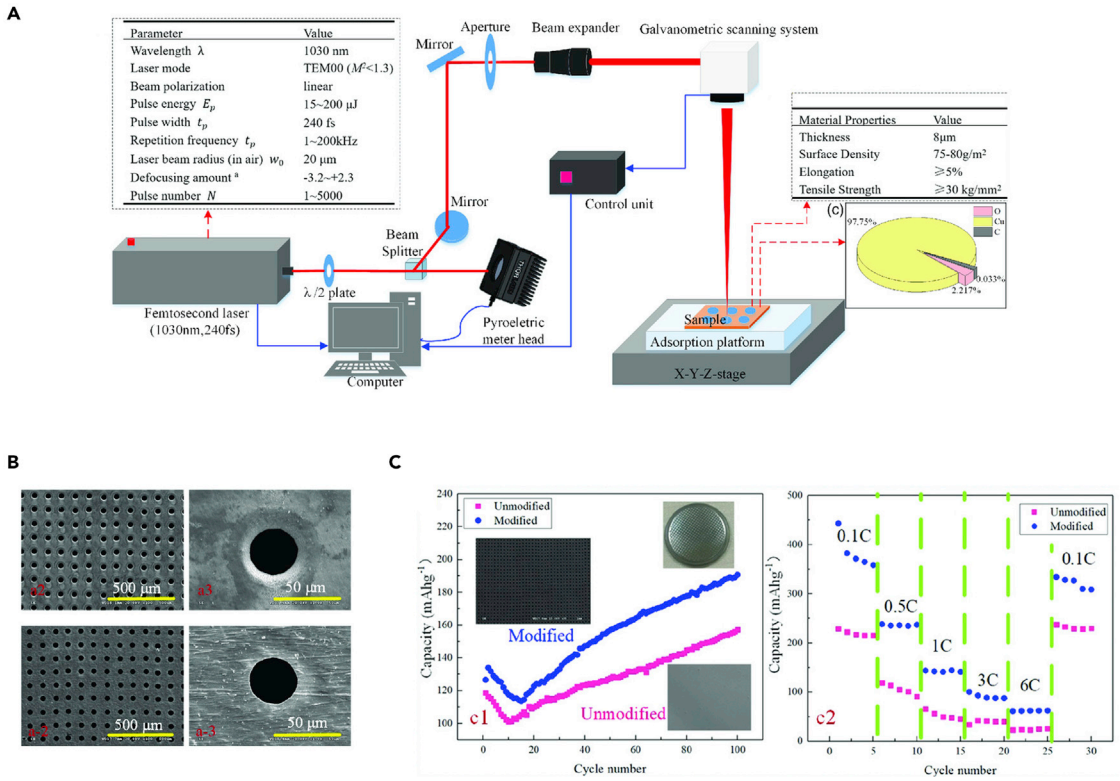

Figure 7. Large-scale microporous group on copper foil current collectors for lithium batteries

(A) Experimental setup for the fabrication of microporous group using LAL.

(B) SEM images of the fabricated microporous group.

(C) Cycle performance and the corresponding rate capability of half-cells with two types of Cu current collectors.

Figures adopted from (Li et al., 2020) with permission.

CONCLUSION AND OUTLOOK

As discussed above, the LAA technology shows promising possibility in improving the performance of electrocatalytic materials, with potential utilization in versatile energy-related applications, such as electrical water splitting, photocatalytic water splitting, and lithium-ion batteries. LAA is a fast fabrication method and facile to conduct, which has a promising prospect in energy-related applications. The development of LAA in the future is full of possibilities. We list several possible directions in the following:

(i) The precise control of LAA as a surface treatment method. LAA is a powerful modification approach, especially for energy materials. Currently, the modifications of LAA to materials cannot be precisely controlled, such as the size of generated small spheres or the style of defects. Further study on the combination of parameters of the pulse laser may lead to precise control of the modification, such as the wavelength, the optical power, the pulse duration time, and so on.

(ii) The application in the fabrication of nanoalloys. Nanoalloys have appealed much attention due to the potentials in CO_2 reduction and bifunctional water splitting (Nam et al., 2018; Kim et al., 2019; Cao et al., 2017; Zhu et al., 2019). Nanoalloys are usually prepared through decomposing corresponding metal salts. LAA may act as a promising route to fabricate nanoalloys since materials can be heated and cooled in a very short timescale.

(iii) The application in the fabrication of homogeneous heterojunctions. Recently, in contrast to heterojunctions that are constructed form two different materials, homogeneous heterojunctions, namely the phase junction, that are constructed from the same material with different phases have exhibited promising potential in hydrogen reaction (Fu et al., 2021). LAA is ideally suitable to generate different phases and thus phase junctions.

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AUTHOR CONTRIBUTIONS

L.Z. and S.S. contributed equally to this work. All the authors revised the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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