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基于激光液相烧蚀的纳米材料制备研究进展

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摘要 直径从 0.1 nm 到 100 nm 的纳米材料在微电子、光电子、生物工程、清洁能源和分子组装等领域受到了极大的关注,使纳米材料的制备得到持续探索和发展。1993 年, A. Henglein 和 A. Fojtik 利用激光液相烧蚀(laser ablation in liquids, LAL)制备了镍和金纳米粒子,引起了学者们对纳米材料制备的极大兴趣。作为一种常用的纳米材料制备方法, LAL 可以制备出尺寸小、纯度高、均匀性好的纳米颗粒,从而获得尺寸小、纯度高、分布均匀的纳米材料。本文重点介绍了毫秒激光、纳秒激光、飞秒激光作用下利用 LAL 制备纳米材料的形成机制和研究现状,总结了利用 LAL 制备纳米材料存在的困境和改进的方法。

关键词 激光液相烧蚀; 纳米材料制备; 形成机制; 毫秒脉冲激光; 纳秒脉冲激光; 飞秒脉冲激光

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Advances in the Preparation of Nanomaterials Based on Laser Ablation in Liquids

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Abstract Nanomaterials with diameters ranging from 0.1 nm to 100 nm have attracted significant attention in the fields of microelectronics, optoelectronics, bioengineering, clean energy and molecular assembly, which has led to the continuous exploration and development of nanomaterials preparation. In 1993, the preparation of Ni and Au nanoparticles using laser ablation in liquids (LAL) by A. Henglein and A. Fojtik spurred scholars to express tremendous interest in the preparation of nanomaterials. As a universal preparation method for nanomaterials, LAL can produce nanoparticles with small size, high purity and

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good uniformity, then obtaining narrow-size, high-purity and uniformly-distributed nanomaterials. In this paper, the formation mechanism and research actuality of preparing nanomaterials based on LAL under millisecond, nanosecond and femtosecond pulsed lasers were the focuses, and the existing difficulties and improved methods of preparing nanomaterials based on LAL were summarized.

Key words laser ablation in liquids; nanomaterials preparation; formation mechanism; millisecond pulsed laser; nanosecond pulsed laser; femtosecond pulsed laser

0 引言

具有表面效应、界面效应、量子约束效应、量子尺寸效应和宏观量子隧道效应的纳米材料在微电子、光电子、生物工程、清洁能源和分子组装等领域具有巨大的潜力,耐热纳米材料、生物医学纳米材料、半导体纳米薄膜、纳米陶瓷、压敏和压电纳米材料的广泛应用推动了科技的发展^[1-5]。

激光液相烧蚀(laser ablation in liquids, LAL)是一种常用的纳米材料制备方法。1993年,Henglein A和Fojtik A使用694 nm脉冲激光制备了镍和金纳米粒子胶体溶液,他们发现激光强度决定了金纳米粒子的平均尺寸^[6]。此后,LAL逐渐成为制备金属、合金、无机非金属、半导体、有机物和复合纳米晶体的手段之一。在液体环境中,脉冲激光与固体靶材之间的相互作用过程非常复杂:除了激光与靶材之间的相互作用,还存在液体与靶材之间的相互作用。激光烧蚀靶材时,液体环境会限制等离子体的形成、溅射和凝结,从而影响最终产物的形态、结构和尺寸。同时,液体中所含的活性试剂和表面活性剂也会改变最终产物的成分和表面状态^[7]。LAL有望在高温高压的极端非平衡条件下制备出传统方法无法获得的具有复杂结构和特殊性质的纳米材料,即纳米材料从单一纳米颗粒扩展到各向异性纳米结构。

本文介绍了不同脉冲激光作用下利用LAL制备纳米材料的形成机理和研究现状,指出了目前存在的关键问题和下一步的研究重点。总之,LAL为复杂结构纳米材料和复合结构纳米材料的可控制备和性能研究提供了一种新技术,对多维结构纳米材料的发展具有重要的现实意义。

1 利用LAL制备纳米材料机理

LAL的典型过程是使用短脉冲激光烧蚀液体中的固体靶材,使靶材表面发生熔化或汽化,从而产生由靶材原子、分子、自由电子、离子等形成的高温高压等离子体羽流。经过膨胀、扩散、冷却和凝结过程,等离子体羽流与液体发生化学反应。在此过程中,等离子体羽流成核并生长形成纳米粒子,纳米粒子聚集成各种纳米结构,并分散在液体中形成纳米粒子胶体^[8]。通过离心、提纯和干燥纳米粒子胶体,可获得粒径窄、纯度高、分布均匀的纳米材料。

LAL中使用的脉冲激光通常为毫秒脉冲激光、纳秒脉冲激光和飞秒脉冲激光。毫秒脉冲激光烧蚀的靶材几乎均为金属,产物为金属液滴^[9]。然而,纳秒脉冲激光和飞秒脉冲激光烧蚀的靶材是多种多样的(金属、合金、非金属等),这就导致了产物(等离子体和金属液滴)不同^[10]。因此,不同的脉冲宽度会导致不同的纳米材料形成机制,下文将逐一分析。

1.1 毫秒脉冲激光

图1为形成纳米液滴以及纳米液滴与液体介质反应。由于功率密度较低($10^6 \sim 10^7 \text{ W/cm}^2$),毫秒脉冲激光烧蚀金属靶材后金属液滴会喷射出来。高温金属液滴使液体汽化产生蒸汽,高温高压蒸汽将金属液滴

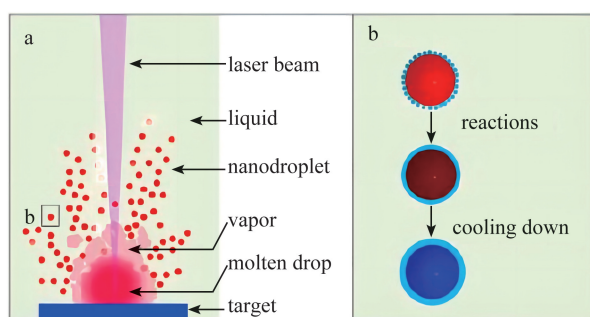


图1 形成纳米微滴以及纳米微滴与液体介质反应^[12]

(a) 形成纳米微滴;(b) 纳米微滴与液体介质反应

Fig. 1 Nanodroplets formation and reaction between nanodroplets and liquid medium^[12] (a) Nanodroplets formation; (b) Reaction between nanodroplets and liquid medium

粉碎成纳米液滴[图 1(a)], 纳米液滴冷却凝结生成纳米颗粒^[11]。如果液体本身或含有能与纳米液滴发生反应的物质, 纳米液滴会与液体发生反应生成空壳、核壳、异质结结构等纳米材料[图 1(b)]^[12-15]。表面反应的速度、程度和深度取决于激光参数和反应物的活性, 并最终决定产物的成分和形态。图 1 左侧的 b 点放大后在右侧显示为图 1(b)。

1.2 纳秒脉冲激光

纳秒脉冲激光能提供稳定性好、密度高的等离子体羽流, 长期以来已成功制备了多种纳米材料(金属、合金、非金属、复合纳米晶体)。由于靶材和液体对烧蚀过程的影响很大, 使用不同靶材和液体制备纳米材料的机理也略有不同。纳米材料的五种形成机理如下。

1.2.1 等离子体冷却-凝结-形成金属簇^[10,16]。Liu P 等人在聚乙烯吡咯烷酮(polyvinyl pyrrolidone, PVP)溶液中烧蚀铁板制备氧化亚铁(FeO)纳米粒子^[10]。图 2 为 FeO 纳米粒子的形成过程:(a) 脉冲激光作用于铁靶材后, 在固液界面迅速产生高温高压的铁等离子体;(b) 等离子体绝热膨胀, 等离子体羽流区迅速冷却形成铁簇;(c) 等离子体坍塌后, 铁簇与 PVP 溶液接触并发生反应形成 FeO 纳米粒子。

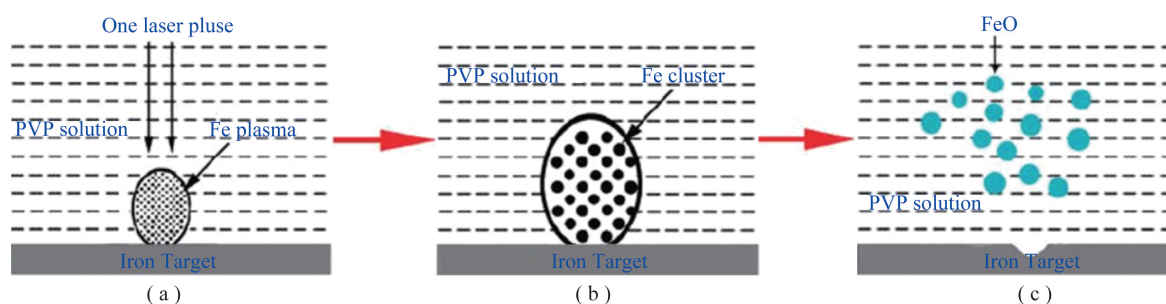


图 2 FeO 纳米颗粒形成过程^[10] (a) 生成铁等离子体;(b) 形成铁簇;(c) 形成 FeO 纳米颗粒

Fig. 2 Formation process of FeO nanoparticles^[10] (a) Production of Fe plasma; (b) Formation of Fe clusters; (c) Formation of FeO nanoparticles

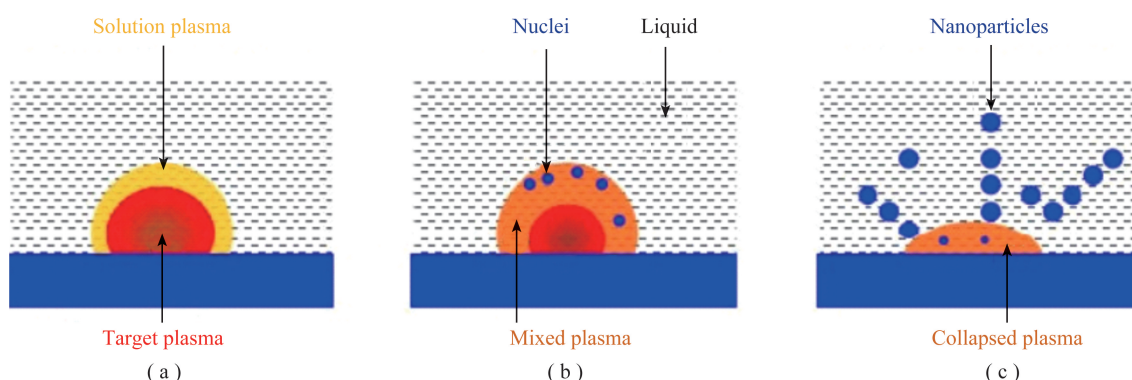


图 3 制备纳米粒子的等离子体混合机制^[12] (a) 靶材等离子体和溶液等离子体;(b) 混合等离子体和核;(c) 形成新的纳米粒子

Fig. 3 Plasma mixing mechanism of preparing nanoparticles^[12] (a) Target plasma and solution plasma; (b) Mixed plasma and nuclei; (c) Formation of new nanoparticles

1.2.2 等离子体混合-凝结-成核生长^[17-19]。Liu Q 等人在硝酸银溶液中烧蚀镍块制备银纳米颗粒和氧化银纳米粒子^[17]。图 3 为制备纳米颗粒的等离子体混合机理:(a) 靶材表面吸收激光能量产生靶材等离子体, 靶材等离子体诱导溶液等离子体产生;(b) 靶材等离子体和溶液等离子体迅速混合形成等离子体羽流区, 并在该区域形成晶核;(c) 等离子体羽流坍塌、冷却并与液体反应形成新的纳米粒子。

1.2.3 等离子体粉碎金属液滴^[20,21]。Phuoc T 等人利用镁在不同溶液(去离子水、十二烷基硫酸钠、丙酮、2-丙醇)中制备氢氧化镁、氧化镁和镁纳米粒子^[20]。图 4 为等离子体将金属液滴粉碎成纳米液滴制备纳米粒子的过程:(a) 金属靶材熔化形成金属液滴, 金属液滴将热量传递给溶液形成溶液等离子体;(b) 高温高压的溶液等离子体将金属液滴粉碎成纳米液滴;(c) 纳米液滴与液体发生反应形成新的纳米粒子。

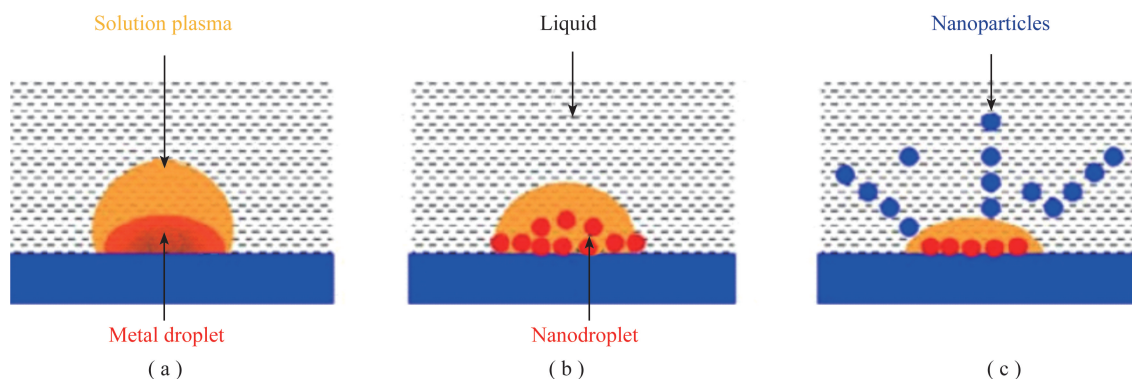


图4 等离子体将金属液滴粉碎成纳米液滴制备纳米颗粒过程^[12] (a) 金属液滴和等离子体溶液; (b) 纳米液滴; (c) 形成新的纳米颗粒

Fig. 4 Plasma crushes metal droplets into nanodroplets process of preparing nanoparticles^[12] (a) Metal droplets and solution plasma; (b) Nanodroplets; (c) Formation of new nanoparticles

1.2.4 靶材直接喷射金属液滴^[22,23]。Tsuiji T 等人在水中烧蚀 Ag、Au 和 Si 靶材,在固液界面观察到两种喷射现象^[22]。一种是由金属团簇和液滴形成的垂直射流,这种射流是纳米粒子胶体的主要成分^[23]。另一种是含有原子、分子、金属离子和水的靶材等离子体,它能持续烧蚀靶材表面并产生金属液滴。图5为靶材直接喷射金属液滴制备纳米粒子的过程:(a) 激光烧蚀靶材表面形成垂直射流和靶材等离子体;(b) 靶材等离子体形成空化泡,射流和气泡持续形成金属团簇和金属液滴;(c) 空化泡坍塌,金属团簇和液滴最终冷却结晶形成纳米颗粒。

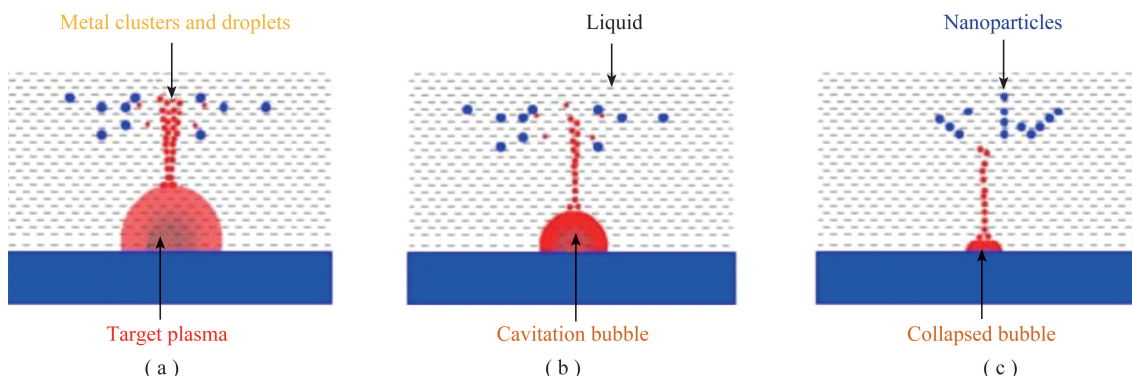


图5 激光直接喷射制备纳米颗粒的金属液滴^[12] (a) 垂直射流和靶材等离子体; (b) 空化泡; (c) 形成纳米颗粒

Fig. 5 Laser direct jets metal droplets of preparing nanoparticles^[12] (a) Straight up jet and target plasma; (b) Cavitation bubble; (c) Formation of nanoparticles

1.2.5 等离子体粉碎金属液滴和等离子体混合^[24-26]。Nichols W 等人研究了激光波长和能量对铂纳米颗粒烧蚀速率、烧蚀机制、尺寸分布和形成机制的影响^[24-26]。他们指出存在两种烧蚀机制,纳米颗粒的形成也由这两种机制决定。图6为等离子体粉碎金属液滴烧蚀机制和等离子体混合烧蚀机制:(a) 靶材表面吸收激光能量产生靶材等离子体和金属液滴,靶材等离子体和金属液滴形成溶液等离子体;(b) 靶等离子体和溶液等离子体迅速混合,形成等离子体羽流区后在该区域形成原子核,高温高压的溶液等离子体将金属液滴挤压成纳米液滴;(c) 等离子体羽流坍塌、冷却形成较小的纳米颗粒,较小的纳米液滴冷却后形成较大的纳米颗粒。

1.3 飞秒脉冲激光

Xiao J 等人基于不同激光功率密度,将飞秒激光与靶材相互作用过程分为四个阶段:相爆炸、碎裂、库仑爆炸和等离子体烧蚀^[27]。此外,Roeterdink W 等人给出了不同功率密度飞秒激光作用于硅靶材的现象^[28]。图7(a)为不同功率密度下飞秒激光诱导硅靶材的现象。图7(b~d)为飞秒激光作用于硅靶材的烧蚀过程。首先,硅靶材吸收飞秒激光能量[图7(b)]。其次,电子通过光电和热离子发射从原子中逸出[图7(c)]。再

次,靶材表面产生电场,电场导致正离子之间产生强大的排斥力。斥力大于键的结合强度,从而导致电子从靶材表面逸出,发生库仑爆炸[图 7(d)]。

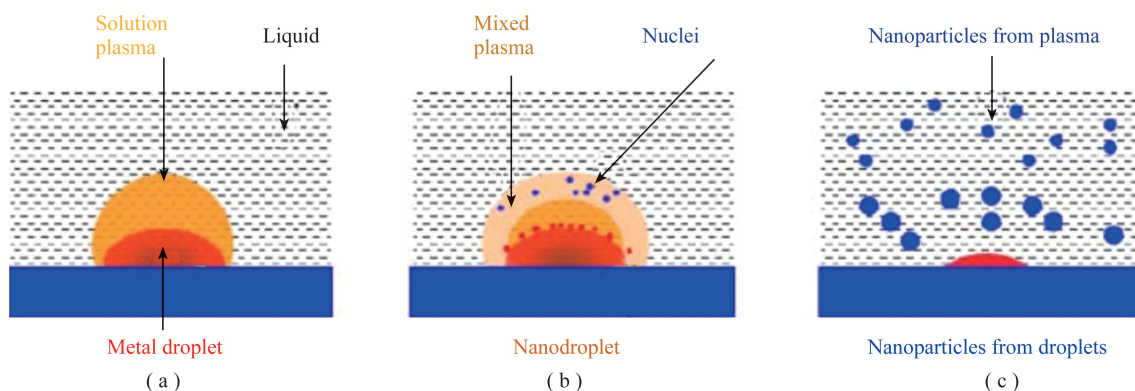


图 6 等离子体粉碎金属液滴和等离子体混合制备纳米粒子^[12] (a) 金属液滴、靶材等离子体和溶液等离子体; (b) 纳米液滴、混合等离子体和核; (c) 等离子体和纳米液滴形成纳米粒子

Fig. 6 Plasma crushes metal droplets and plasma mixing of preparing nanoparticles^[12] (a) Metal droplets, target plasma and solution plasma; (b) Nanodroplets, mixed plasma and nuclei; (c) Formation of nanoparticles from plasma and nanodroplets

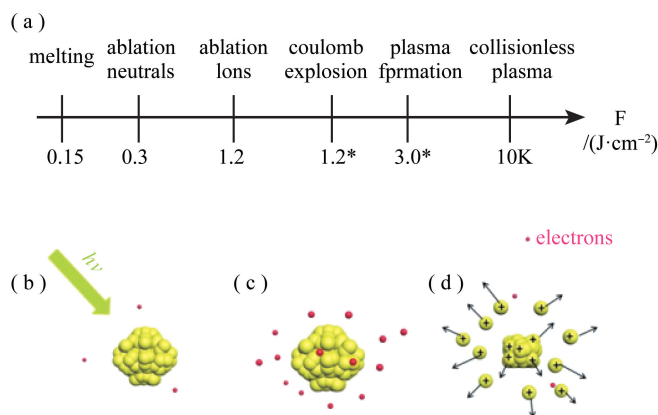


图 7 不同功率密度下飞秒激光作用于硅靶材的现象^[28] (a) 不同功率密度下飞秒激光诱导硅靶材的现象;

(b) 硅靶材吸收激光能量; (c) 电子从原子中逸出; (d) 发生库仑爆炸

Fig. 7 Phenomenon of femtosecond laser acting on silicon target at different power densities^[28] (a) Femtosecond laser-induced phenomenon on silicon target at different power densities; (b) Target absorbs laser energy; (c) Electrons escape from the atoms; (d) Coulomb explosion happens

总之,纳米材料的形成机理非常复杂。脉冲宽度、靶材和溶液都对形成机理有很大影响,我们不能简单地根据脉冲宽度来分析纳米材料的形成机理。

2 利用 LAL 制备纳米材料研究现状

根据等离子体或液滴是否与溶液发生反应,LAL 可分为非反应型 LAL 和反应型 LAL。在非反应型 LAL 中,通常选择贵金属、合金和无机非金属作为靶材,以获得高稳定性和尺寸均匀的纳米粒子。在反应型 LAL 中,选择活泼金属和化合物作为靶材,产物与溶液反应生成金属、氧化物、氢氧化物和其他化合物纳米粒子。此外,利用 LAL 制备具有纳米棒、核壳结构、复合纳米材料等特殊结构的纳米材料也取得了进展。

2.1 制备贵金属纳米粒子

1993 年,Henglein A 和 Fojtik A 首次将激光烧蚀应用于制备金属纳米颗粒(金和镍纳米粒子)^[6]。同年,Cotton T 等人利用 1 064 nm 脉冲激光在不同溶液中获得银、金和铂纳米粒子胶体,验证了上述纳米粒子具有良好的稳定性和可重复性^[29]。此后,国内外学者开始利用 LAL 制备各种贵金属纳米粒子。1996 年至 2008 年,学者们开展了银和金纳米粒子胶体制备,并将其用于表面增强拉曼散射相关研究^[30-39]。他们

还研究了实验条件(激光强度、波长、脉宽、重复频率、溶液)对粒度、稳定性、分散性和均匀性的影响^[37-53]。此外,学者们还合成了钯、铂、钨和铂/钯复合纳米粒子^[54-58]。

2.2 制备合金纳米粒子

2001年, Kim K 等人水中烧蚀金-银块状合金制备金/银合金纳米颗粒,通过改变烧蚀时间和激光能量控制纳米颗粒的尺寸分布^[59]。2002年, Yeh C 等人用纳秒激光照射金-钯和银-钯胶体混合物,制备金/钯和银/钯合金纳米颗粒^[60]。2003年, Wang G 等人利用纳秒激光在硝酸银溶液中烧蚀镍靶材,制备银/镍合金纳米棒^[61]。同年, Simard B 等人烧蚀银和金粉末混合物,制备了金/银合金纳米颗粒^[62]。2004年, Zhang W 等人利用纳秒激光烧蚀铁-镍合金,制备了铁/镍合金纳米颗粒^[63]。2005年, Kazakevich P 等人利用纳秒激光对铜-锌合金进行烧蚀,制备了铜/锌合金纳米颗粒^[64]。2006年, Koshizaki N 等人在脱气己烷和去离子水中烧蚀 $\text{Fe}_{50}\text{Pt}_{50}$ 靶材,制备铁/铂合金纳米颗粒^[65]。2008年, Zhang J 和 Lan C 用纳秒激光在乙二醇中烧蚀镍-钴合金,制备了镍/钴合金纳米颗粒^[66]。

2.3 制备无机非金属纳米颗粒

1998年和2002年, Yang G 等人用纳秒激光在水和无水丙酮中烧蚀石墨靶,得到了立方和六方结构的金刚石纳米晶体^[67,68]。2004年, Liu C 等人在去离子水和乙醇有机溶剂中制备硅纳米颗粒^[69]。同年, May P 等人用纳秒激光在水和环己烷中烧蚀石墨靶,制备了金刚石纳米晶体^[70]。2006年, Švrček V 等人利用纳秒激光在水中烧蚀结晶硅片制备硅纳米晶,但硅纳米晶的结晶性不够好^[71]。2007年, Qin W 利用毫秒激光烧蚀硅片、硅和一氧化硅粉末制备面心立方结构和金刚石结构的硅纳米晶^[72]。同年, Yang L 等人利用纳秒激光在氨溶液中烧蚀石墨靶,制备了氮化碳纳米粉末和纳米棒^[18]。2009年, Cai W 等人用纳秒激光在去离子水和乙醇中烧蚀硅板,制备了硅纳米颗粒^[73]。2010年, Semaltianos N 等人利用飞秒激光在去离子水中烧蚀硅靶,获得了硅纳米颗粒^[74]。同年, Shafeev G 等人用飞秒激光在95%乙醇中烧蚀单晶硅片,制备了球形硅纳米颗粒^[75]。2011年, Qiu J 等人利用飞秒激光在1-己烯中烧蚀大块单晶硅片,得到了硅纳米晶^[76]。2016年, Fang Y 等人用纳秒激光在乙醚中烧蚀块状磷化硼制备磷烯量子点^[77]。2019年, 王颖等人在去离子水中烧蚀碳化硅粉末,得到碳纳米颗粒^[78]。2019年, Pan L 等人利用飞秒激光在乙醇中烧蚀块状金刚石制备金刚石纳米晶^[79]。2020年, 郝娟利用飞秒激光在去离子水、乙醇等溶液中烧蚀金刚石靶材制备金刚石纳米颗粒,发现乙醇溶液中制备的纳米颗粒分散性良好、平均粒径较均匀^[80]。2023年, 陈文彬等人利用飞秒激光烧蚀甲醇溶液中的单壁碳纳米管制备了碳炔^[81]。

2.4 制备非贵金属纳米颗粒

1993年, Cotton T 等人首次利用1064 nm 脉冲激光获得了铜纳米颗粒胶体^[29]。1999年, Yeh C 等人在2-丙醇中烧蚀氧化铜粉末,制备了铜纳米粒子胶体^[82]。2000年, Tsuji T 等人用纳秒激光在十二烷基硫酸钠溶液中烧蚀铜靶制备了铜纳米颗粒,发现烧蚀效率与激光波长有关^[43]。2002年, Bozon-Verduraz F 等人利用纳秒激光在不同溶液中制备了钛和硅纳米颗粒^[49]。2007年, 徐兵用纳秒激光在不同溶液中烧蚀钛板,制备了钛纳米颗粒^[38]。2008年, Zhang J 等人用纳秒激光在乙二醇中烧蚀镍板和钴板制备镍和钴纳米颗粒^[66]。2009年, Stratakis E 等人利用飞秒和皮秒激光对铝靶进行烧蚀,制备铝纳米颗粒^[83]。2011年, Maurizio Muniz-Miranda 等人利用纳秒激光对水溶液中的铜板进行烧蚀,得到铜纳米颗粒胶体^[84]。2014年, Choi M 等人利用纳秒激光在不同溶液中烧蚀镍板,制备面心立方和六边形紧密堆积的镍纳米颗粒^[85]。2017年, 卢红利用纳秒激光在水中烧蚀铅板制备铅纳米颗粒^[86]。2020年, 黄宏坤等利用纳秒脉冲激光在去离子水溶液中烧蚀锆靶制备微米级和亚微米级锆纳米颗粒^[87]。

2.5 制备金属化合物纳米颗粒

2001年, Jiang Z 等人利用纳秒激光在2-丙醇和乙二胺的混合溶液中用硝酸银和硒粉末制备硒化银纳米颗粒^[88]。2004年, Huang C 等人用纳秒激光烧蚀十六烷基三甲基溴化铵溶液中的镓靶材,得到纺锤结构羟基氧化镓纳米颗粒,这是首次利用LAL获得各向异性的纳米结构材料^[89]。同年, Tenne R 等人利用纳秒激光在二硫化叔丁基中合成了无机富勒烯结构层状硫化铅^[90]。2005年, Cai W 等人利用纳秒激光烧蚀十二烷基硫酸钠溶液中锌板,制备出锌/氧化锌核壳结构纳米粒子,发现通过改变十二烷基硫酸钠溶液浓度可以

控制纳米粒子的组成和尺寸^[91]。2004年至2005年,Koshizaki N等人利用纳秒激光在不同溶液中烧蚀锡、钛、锌和镁板,制备出氧化锡、二氧化钛、氧化锌、氢氧化锌和氢氧化镁纳米粒子^[92-96]。2007年,Yang L等人利用纳秒激光烧蚀十二烷基硫酸钠溶液中锌粉末制备出氧化锌纳米叶状结构^[97]。同年,Fauteux C等人水和乙醇中烧蚀乙酰丙酮锌以制备氧化锌纳米结构^[98]。同年,Zhang J等人制备了单层、双层和多层结构、齿轮、盖锅、半球和碗状氧化锌纳米颗粒^[99]。同年,Zheng Z等人利用飞秒激光烧蚀去离子水中硫化镉靶材,制备出硫化镉纳米晶体^[100]。2008年,T. Phuoc等人用纳秒激光烧蚀去离子水和十二烷基硫酸钠溶液中的镁靶,制备出氢氧化镁、氧化镁和镁纳米粒子^[20]。同年,刘培生等人用纳秒激光烧蚀聚乙烯吡咯烷酮溶液中铁板和钛板,制备出氧化亚铁和金红石型二氧化钛纳米粒子^[10,101]。同年,Zheng Z等人用飞秒激光烧蚀去离子水中硫化锌-铜合金靶材,制备出铜掺杂的硫化锌量子点^[102]。2008年至2009年,Qiu J等人用飞秒激光和纳秒激光烧蚀不同溶液中的锆靶材,制备出二氧化锆纳米粒子^[103,104]。2010年,Liang C等人用纳秒激光烧蚀去离子水中的锰板,制备出单相四氧化三锰纳米粒子^[105]。同年,凌晨利用毫秒激光烧蚀不同溶液中的铅靶,获得了自组装硫化铅量子点、硫化铅中空结构、铅纳米晶、铅/硫化铅核壳结构、铅/硫化铅异质结结构和硫化铅纳米晶^[13]。2011年,Du X等人用毫秒激光烧蚀聚乙烯吡咯烷酮溶液中锌-铜复合靶,制备出铜掺杂的氧化锌量子点^[106]。同年,方合等人用纳秒激光烧蚀不同溶液中锌靶材,获得了氧化锌和锌/氧化锌核壳结构纳米粒子^[107]。同年,Wang R等人水中烧蚀二硫化钼粉末,获得了类富勒烯结构二硫化钼纳米粒子^[108]。同年,牛凯阳在不同溶液中用毫秒激光烧蚀镍、铜和铁靶材,获得氧化镍、硫化铜和铁/铁氧化物核壳结构纳米粒子^[12]。2012年,孙明艳在硫代乙酰胺溶液中用毫秒激光烧蚀锡靶,制备出锡/硫化锡核壳结构纳米粒子^[15]。同年,G. Compagnini等人用纳秒激光烧蚀二硫化钼靶材,制备出类富勒烯结构二硫化钼纳米粒子^[109]。2013年,Liang C等人利用纳秒激光烧蚀氯化镍溶液中锰靶,制备出掺锰的氢氧化镍纳米片^[110]。2014年,Durgun E等人甲醇中用纳秒激光烧蚀二硫化钼粉末,制备出二维和类富勒烯结构二硫化钼纳米结构^[111]。2015年,Liu X等用纳秒激光在不同溶液中烧蚀钛/铝合金,制备出二氧化钛/氢氧化铝和空心多孔二氧化钛纳米材料^[112]。2016年,Liu X等人用纳秒激光烧蚀硫代乙酰胺、氯化氢和十六烷基三甲基溴化铵混合溶液中锌靶材,得到硫化锌/锌核壳结构纳米复合材料和空心多孔纳米复合材料^[113]。同年,Chen M等人烧蚀硫代乙酰胺和氯化氢混合溶液中锡块,制备出三硫化二锡纳米粒子^[114]。同年,陈学敏用纳秒激光烧蚀尿素溶液中镍-铁合金靶,制备出氧化镍/层状镍铁双金属氢氧化物复合纳米材料^[115]。2017年,Cai W等人用纳秒激光烧蚀水中二硫化钼靶材,制备出球形二硫化钼纳米粒子^[116]。同年,窦康用纳秒激光烧蚀乙醇和过氧化氢混合溶液中钼靶,制备出二氧化钼纳米粒子^[117]。2019年,秦璟在异丙醇和水中用纳秒激光烧蚀硫化镉靶材,制备出硫化镉/碳纳米复合材料^[118]。2020年,郝娟利用飞秒激光在去离子水、乙醇等溶液中烧蚀氮化镓薄膜制备氮化镓纳米颗粒^[80]。2021年,陈永义在水、过氧化氢和氢氧化钠溶液中分别对铜靶和二硫化钼靶进行烧蚀,成功制备了氧化铜纳米粒子、氧化钼和四氧化三钼纳米粒子混合物(水溶液)、氧化钼纳米粒子(过氧化氢和氢氧化钠溶液)^[119]。

从以上研究现状可知:在研究初期,学者们主要利用非反应型 LAL 制备贵金属、合金和无机非金属纳米颗粒,重点研究不同实验条件(激光波长、能量、脉宽、重复频率、烧蚀时间、溶液)对纳米颗粒的粒度、形貌、均匀性和稳定性的影响。制备贵金属纳米粒子的研究相对完备。制备合金纳米粒子研究前期主要以贵金属为靶材,后期则主要以非贵金属为靶材。无机非金属纳米粒子的结构和形态主要取决于实验用溶液。合金和无机非金属纳米粒子的制备尚有一定的研究空间。非贵金属一般会与溶液发生反应,产物可能是金属、氧化物、氢氧化物和其他化合物纳米粒子。此外,目前利用 LAL 制备纳米粒子的研究重点为对纳米粒子的可控制备以及各种特殊结构和形态(球形结构、纺锤结构、核壳结构、叶状结构、中空结构、面心立方结构、金刚石结构、六边形结构、富勒烯结构、异质结结构、自组装结构、空心多孔结构、纳米棒、碳炔)纳米材料的制备。

3 总结

作为一种常用的纳米材料制备方法,LAL 具有许多传统方法无法比拟的优点,如操作简单、环保、适用性广等。磷烯、MXenes 等新兴纳米材料在切削润滑、光催化剂、能源转化、光电探测器、电化学传感、肿瘤诊

断、口腔医学、生物传感器等领域逐渐取代了传统材料,使得人们对这些纳米材料的需求日常增长^[120-130]。然而,利用 LAL 制备二维纳米材料仍存在一些亟待解决的问题:首先,缺少适合的反应溶液进行活性金属和化合物纳米粒子的制备。其次,具有特殊形态和结构的纳米粒子尚未完全达到可控制备,即未完全掌握不同实验条件(激光波长、能量、脉宽、重复频率、溶液、烧蚀时间等)对纳米粒子的粒度、形貌、结构、均匀性和稳定性影响规律。最后,各类纳米粒子的生产效率仍然偏低,无法将其大规模应用于工业生产。

综上所述,利用 LAL 制备纳米粒子拥有巨大的发展潜力和进步空间。未来可以从以下几方面进行改进:首先,研究不同溶液对不同活泼反应物生成产物和结构的影响。其次,将实验条件控制为单一变量变化研究纳米粒子的可控制备。再次,保证纳米粒子粒径和结构稳定的前提下提高纳米粒子生产效率。最后,将 LAL 与化学沉积法、光催化法等其他纳米粒子制备方法相结合,制备出各种特殊结构和形态的纳米复合材料。

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