

3D 打印微波吸收材料研究进展

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摘要:近年来,随着3D打印技术逐渐成熟化与商业化,这种新兴制造技术开始应用于吸波材料的设计与制备中。本工作从3D打印频率选择表面类和超材料类吸波材料、3D打印蜂窝类吸波材料、3D打印陶瓷类吸波材料和3D打印其他吸波材料等几个方面综述了3D打印技术在微波吸收材料制备方面的研究进展,对3D打印技术在微波吸收材料制造中存在的打印材料局限性、材料力学性能缺乏、微观结构的测试分析等问题进行了阐述,同时对3D打印技术在微波吸收材料制造领域未来的发展趋势,如小型化、多功能、智能化也进行了展望。

关键词:3D打印;微波吸收;超材料;吸波蜂窝;陶瓷基吸波材料

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微波吸收材料是一种可以将表面接收到的电磁波吸收或者大幅度衰减的材料,主要应用于飞行器电磁隐身^[1-2]、电磁兼容^[3]、传感器装置^[4-5]等。按照吸波原理划分,微波吸收材料可分为电磁损耗吸收材料、结构谐振吸收材料和表面电容电阻吸收材料。电磁损耗吸收材料是通过材料中的电损耗吸收剂或磁损耗吸收剂实现对入射波的能量吸收和转化^[6-8],主要的吸收剂有羰基铁粉、铁氧体和碳系吸收剂(碳纤维、碳纳米管或石墨烯等);结构谐振吸收材料是通过材料特殊的谐振结构设计,使入射波入射到波长1/4的间隔结构后与反射波相互干涉而抵消,典型的谐振型结构有Salisbury screen^[9]和Jauman absorbers^[10];表面电容电阻材料的吸收机理是通过高阻抗表面(high impedance surface)实现对电磁波的谐振吸收,如频率选择表面(frequency selective surface, FSS)^[11-12]和最近热门的超材料吸波体^[13-15]。目前,学者们采用多种多样的制备工艺得到多种微波吸收材料,例如吸波涂料涂覆成型工艺,蜂窝和泡沫浸渍吸波胶体成型工艺,结构型吸波材料的复合材料成型工艺以及电阻型吸波材料的表面贴片或电镀金属导电物成型工艺。3D打印技术又称增材制造技术,与上述吸波材料传统制备工艺相比,3D打印技术具有制造成本低、易成型、可打印不规则形状从而增加设计优

化的自由度等优点^[16-20],其主要工作原理是以数字模型为基础,按照一定分层厚度和预定堆积轨迹,将金属或非金属材料逐层叠加制造出特定模型或者结构,主要分类如表1所示^[21]。

考虑到吸波材料的结构组成特点,可以用于吸波材料制备的3D打印制造技术主要有熔融沉积成型法(fused deposition modeling, FDM),选择性激光烧结法(selective laser sintering, SLS)和光固化成型法(stereo lithography apparatus, SLA),打印材料为透波的介电材料(树脂基或陶瓷基的粉末或丝材)和吸收剂(磁损耗型、电损耗型或电/磁损耗复合型),材料主要为球形粉体混合物或特殊制备的打印丝材^[26]。本研究主要从3D打印FSS类和超材料类吸波材料,3D打印蜂窝类吸波材料,3D打印陶瓷类吸波材料和其他3D打印吸波材料等几个方面综述3D打印技术在微波吸收材料制备的研究进展,并对打印技术在材料制造中存在的问题进行阐述。

1 3D 打印 FSS 类和超材料类吸波材料

FSS或超材料吸波体是通过高阻抗表面来实现对电磁波的谐振吸收。主要形式为:在介质层表面形成规则几何排列周期结构的金属导电谐振面,底部则采用金属板对透过的电磁波进行全反射,金属板与表面金属耦合形成磁谐振器^[27]。根据强谐振理论,这种结构仅能实现窄带吸波,而且平面结构对于入射波的极化方式和角度敏感^[28],无法实现

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表 1 增材制造分类^[21]
Table 1 Sort of additive manufacturing^[21]

No	Manufacturing material	Material form	Source	Technic
1	Metallic materials	Powder/particle materials	Laser additive manufacturing	Single step additive manufacturing
2	Inorganic Non-metallic Materials	Filament materials	Electron beam additive manufacturing	Multi-step additive manufacturing
3	Polymer Materials	Strip/sheet materials	Arc and wire additive manufacturing	Composite additive manufacturing
4	Biological material	Liquid material	Stereo lithography apparatus	—
5	—	—	Hot melt additive manufacturing	—

多极化方式和大角度斜入射吸收。为了解决上述问题, 研究者们采取不同方式, 如多模式谐振的独特单元结构设计^[29-30], 多重谐振结构超材料/磁性吸波材料复合吸波体^[14], 拓扑结构单元嵌套或阵列排布等方式进行复合^[31-32], 引进电子元器件^[33-34]以及周期结构三维设计等^[35-38]。3D 打印技术由于其制造技术的便捷性和可设计性, 可将上述解决方案引用到三维结构 FSS 或超材料吸波体的制备中, 从而克服传统设计造成的窄带问题以及平面结构带来的人射波极化方式和角度敏感问题。

熊益军等^[39]利用 3D 打印技术制备了一种三层宽频超材料吸波体, 打印方法为 SLS, 材料结构的表层和中间层为单元尺寸不同的周期阵列结构, 底层为吸波平板结构, 材料厚度 4.7 mm, 周期性结

构材料为羰基铁粉和尼龙混合粉体, 综合考虑材料吸波性能和制备过程中熔融固化等制造工艺因素, 选取尼龙体积分数为 30%, 激光功率为 45 W, 扫描间距为 0.3 mm。吸波体反射率测试表明, 在电磁波垂直入射下, 4~18 GHz 频率范围内, 反射率 ≤ -10 dB, 吸收峰分别出现在 5.3 GHz 和 14.1 GHz。Zhou 等^[40]也利用 SLS 打印方法制备超材料周期性结构吸波体, 如图 1 所示。打印材料同样为羰基铁粉和尼龙的混合粉体, 混合物中羰基铁粉体积分数为 70%。为减少成型缺陷, 采用两种颗粒粒径大小相近的粉体, 其中尼龙粉体颗粒为 20~150 μm , 羰基铁粉球磨后粒径约为 120 μm 。制造工艺上, 最优打印参数为预约温度 130 $^{\circ}\text{C}$, 扫描间距 0.13 mm, 激光功率 40 W, 频率范围 8~18 GHz, 反射率 ≤ -10 dB。

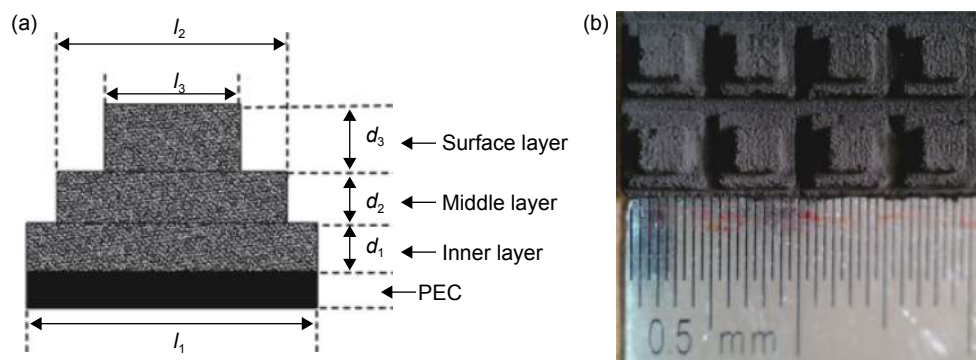


图 1 周期性结构打印^[40] (a) 周期结构单元尺寸; (b) 打印材料样品图

Fig. 1 Periodic structure printed^[40] (a) geometry of periodic structural unit; (b) sample of printed material

Kronberger 等^[41]研究 3D 打印超材料中打印材料的介电性能。打印材料为芬兰 ColorFabb 公司所生产的不同金属铜填充的 ABS 或 PLA 导电丝束, 该复合丝束材料具备较高的导电性(表 2), 同时因渗滤效应^[42]该丝束也具备一定磁性, 有利于材料吸波性能的提升。

Lleshi 等^[43]利用 FDM 打印技术一步法制备出

表 2 导电丝材的介电性能@10 GHz^[41]

Table 2 Material parameters @10 GHz of the filaments^[41]

Material	Density/ ($\text{g}\cdot\text{cm}^{-3}$)	ϵ_r	$\tan\delta_\epsilon$	μ_r	$\tan\delta_\mu$
BrassFill	2.4	8.15	0.015	1.25	0.2
BronzeFill	2.95	8.0	0.01	1.41	0.03
CopperFill	3.0	8.3	0.08	1.35	0.025

金属电介质材质的锥体多层结构超材料 (tapered hyperbolic metamaterials) 吸波体, 该结构可拓宽吸收频带, 解决仿真模拟电路谐振吸波中的窄带问题, 同时大角度斜入射吸波性能也有所提高。该吸波体的打印材料分别为导电金属丝材 (棕色) 和电介质丝材, 如图 2 所示。因为打印技术和材料的限制, 打印图案并不是很细致精准, 然而这个外形不完美的缺陷却能增加电磁波在结构中的散射, 从而提高吸波效能, 最终材料在 9~18 GHz 频率范围内具有 95% 的吸收能效, 在 7.6~17.4 GHz 频率范围内具有 90% 的吸收能效。

Lim 等^[44] 利用 3D 打印技术研制一种类阶梯状的十字周期微型化超材料结构。如图 3 所示, 首先打印 PLA 材质的周期结构, 然后使用刷子将导电银浆刷于 PLA 模板上制成吸波超材料。研究发现, 通过增加阶梯结构的层数, 可使吸收频带向低频偏移。在 5.18 GHz 处材料的吸波效率高达 99.8%, 同时材料对于入射波的极化方式并不敏感。

表 3 总结了近年来利用 3D 打印技术制备 FSS 类和超材料类吸波材料的性能。从表 3 中可

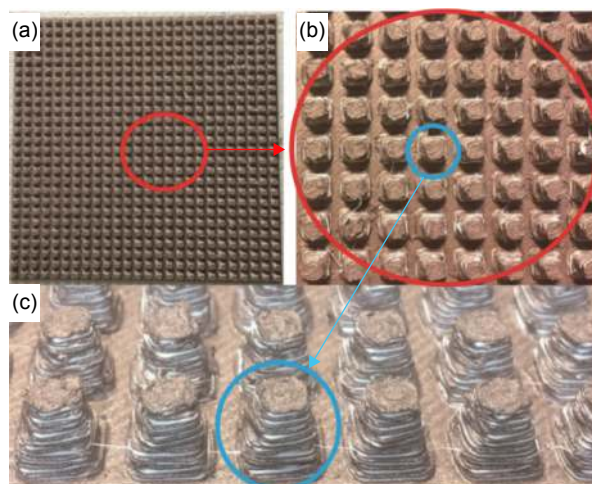


图 2 FDM 打印技术 3D 打印吸波体结构^[43] (a)、(b) 结构整体俯视图; (c) 结构单元侧视图

Fig. 2 FDM printing techniques 3D-printed absorber structure^[43] (a), (b) top view of whole structure; (c) side view of structural cells

以看出, 超材料吸波领域 3D 打印技术主要集中在 SLS 法、FDM 法, 介电打印材料主要为成熟的商业

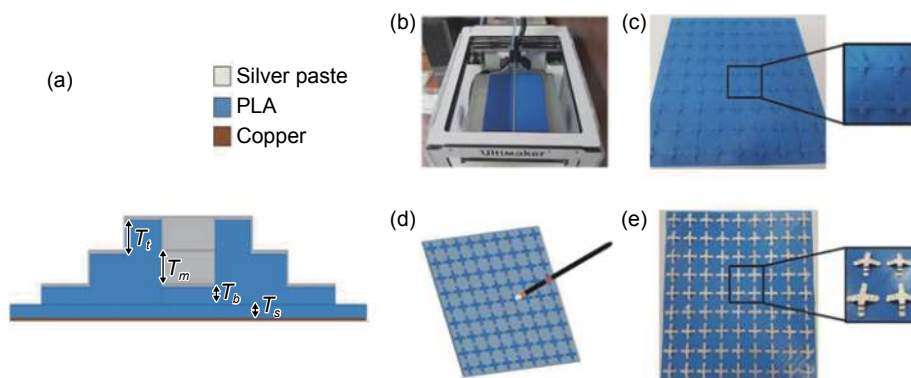


图 3 3D 打印十字阶梯型超材料吸波体^[44] (a) 结构单元侧视图; (b), (c), (d), (e) 3D 打印吸波材料的制备过程

Fig. 3 3D printed stair-like Jerusalem cross metamaterial absorber^[44] (a) side view of unit; (b), (c), (d), (e) fabrication process of 3D printed absorber sample

表 3 3D 打印 FSS 类和超材料类吸波材料

Table 3 Sorts of FSS absorber and metamaterial fabricated by 3D-printing

Measured absorption band/GHz	Bandwidth/GHz	Thickness/mm	Commet	Ref.
8.0-18.0	0.7	3.0	SLS with Nynol+CIP, copper plate	[40]
8.0-12.0	0.6	2.13	3D with BronzeFill filaments+ Nynol or ABS	[41]
7.6-17.4	0.78	7.8	Single step FDM, metal-dielectric multilayer	[43]
4.6-6.0	1.00	2.9	FDM with silver paste-loaded PLA, copper plate	[44]
3.5-24.0	1.49	16.0	FDM with PLA, copper plate	[45]
4.0-12.0	1.00	10.0	FDM with carbon-loaded ABS, copper plate	[46]
8.0-18.0	0.77	40.0	FDM with PLA+CIP, all dielectric	[47]

热塑性打印塑料尼龙、ABS、PLA, 电阻层主要为金属导电材料, 也可以是碳基铁粉/碳系吸收剂与树脂基的混合物。

2 3D 打印蜂窝类吸波材料

与块体吸波材料相比, 蜂窝类吸波材料在多极化方式入射和大角度斜入射的吸波效能更好, 同时兼具优异的力学性能, 作为结构功能一体化材料应用在电磁吸波领域^[48-50]。蜂窝类吸波材料多以芳纶类或玻纤类六角或四角蜂窝作为基体, 在其上浸渍含电损耗型或磁损耗型吸收剂的树脂预浸料成型而成, 或在蜂窝中直接填充具有电磁损耗功能的

电阻层或泡沫体。传统工艺对蜂窝的图形化结构进行电磁性能设计困难, 近年来, 一些研究者们利用 3D 打印技术对蜂窝结构电性能进行设计, 以期获得材料吸波性能的进一步突破。

Jiang 等^[45] 使用 3D 打印技术和丝网印刷技术制备轻质蜂窝结构, 如图 4 所示。在打印的 PLA 蜂窝壁上增加碳基电阻层, 形成周期性三维蜂窝类超材料吸波体, 该结构体不仅保留了蜂窝结构对电磁波吸收效能, 且因为加入了电阻层, 进而形成类超材料吸波体。研究表明, 当材料厚度为 15.1 mm, 密度为 0.255 g/cm^3 时, 材料在 3.53~24 GHz 频段范围内吸收率低于 -10 dB, 同时提高了水平极化方式时 $0\sim 70^\circ$ 大角度的吸波性能。

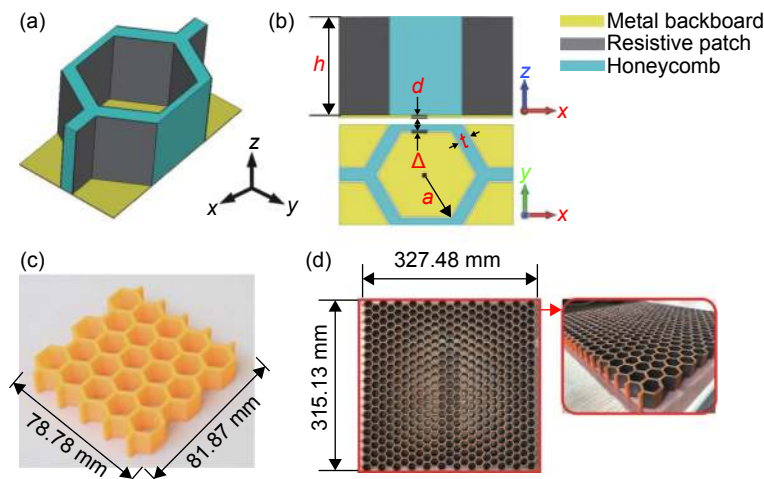


图 4 三维蜂窝结构^[45] (a)单元示意图; (b) x - z 和 x - y 平面单元视图; (c)用于压缩试验的蜂窝样品; (d)三维蜂窝结构样品
Fig. 4 Three-dimensional honeycomb structure^[45] (a)unit cell diagram; (b)views of unit cell in plane x - z and x - y ; (c)honeycomb sample for compressive test; (d)sample of the three-dimensional honeycomb structure

Vincent 等^[51] 通过 FDM 方法打印一种吸波蜂窝结构, 并进一步开发出具有复杂拓扑结构的蜂窝吸波体。打印材料为 PLA/炭黑的丝材, 材料经过高温喷嘴熔融, 在 Z 轴移动的衬板上沉积形成最终打印图案。使用同样方法可做出具有拓扑图型的多层蜂窝结构。拓扑型多层蜂窝结构在 12.5~18 GHz 的反射率高于六角蜂窝型, 见图 5。该方法可直接一步法实现吸波蜂窝制备过程, 工艺简单。

3 3D 打印陶瓷类吸波材料

陶瓷基耐高温吸波材料是为满足电磁吸波体在高温环境下的应用而发展起来的新型结构吸波一体化材料, 主要优势为耐高温、耐腐蚀、抗氧化, 解决常温吸波材料无法满足的工程应用^[52-54]。目前研究较多的陶瓷类吸波材料有掺杂改性 SiC 陶

瓷类^[55-57]、掺杂改性钡铁氧体陶瓷 ($\text{BaFe}_{12}\text{O}_{19}$, BFO) 类^[58-59]、聚合物先驱体裂解转化陶瓷类型^[60-61]。从结构类型上看, 多孔陶瓷因存在结构上的空气孔洞, 与致密陶瓷材料相比, 增加了电磁波入射阻抗匹配, 同时在材料内部多次折射, 从而提高电磁波的吸收效率^[62]。在 3D 打印制造领域中, 这一类陶瓷基吸波材料也受到了研究者的关注。Mei 等^[63] 首先利用 SLA 光固化成型法 3D 打印技术制备不同倾斜角度的 Al_2O_3 陶瓷多孔蜂窝结构, 然后利用化学气相渗透法 (CVI) 在 Al_2O_3 陶瓷结构上生长微米级晶须结构 SiC_w 。通过调节多孔蜂窝结构角度和纤维含量, 最终获得宏观结构和微观材料设计的协同优化, 结果表明: 当孔洞倾斜角度为 30° , 材料厚度 3.5 mm 时, 材料的最大反射率可达到 -63.65 dB, 有效吸收带宽覆盖整个 X 波段 (8.2~12.4 GHz) (图 6)。

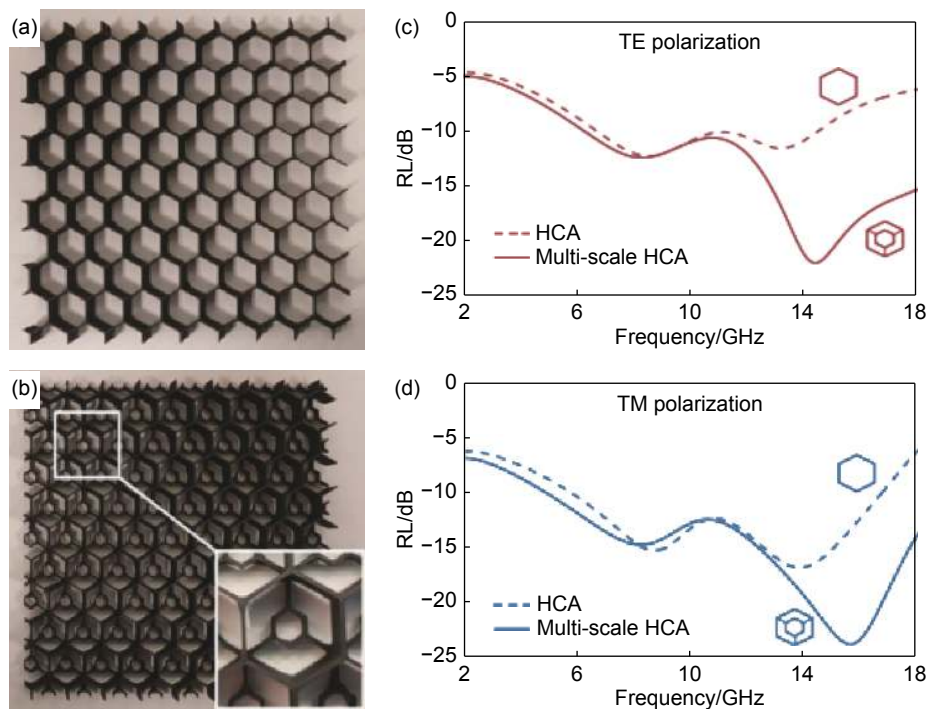


图 5 D 打印 PLA-C 蜂窝结构吸波材料^[51] (a)蜂窝结构吸波材料;(b)3D 打印多尺寸 PLA-C 蜂窝吸波材料(拓扑结构);(c)、(d)在 2~18 GHz、入射角 20°、TE/TM 极化时蜂窝结构和多尺寸蜂窝结构反射率对比图

Fig. 5 3D-printed honeycomb microwave absorbers in PLA-C^[51] (a) honeycomb microwave absorbers;(b) 3D-printed multi-scale honeycomb microwave absorbers in PLA-C(topological structure);(c), (d) comparison of measured reflection coefficients of honeycomb microwave absorbers and multi-scale honeycomb microwave absorbers at 2~18 GHz frequency band, incident angle 20° and TE/TM polarization

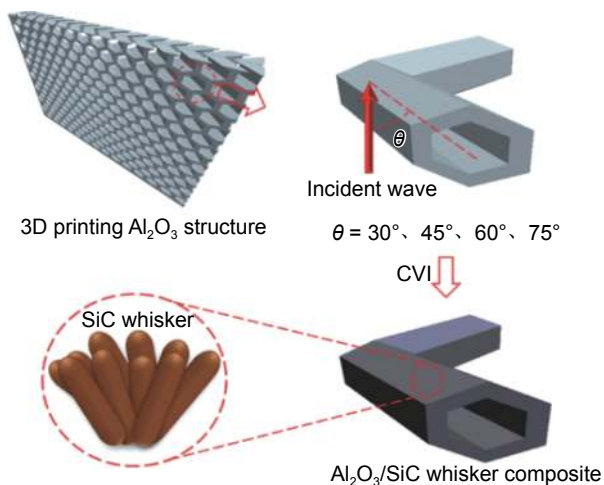


图 6 3D 打印 Al₂O₃/SiC 晶须复合材料的工艺流程示意图^[63]
Fig. 6 Schematic diagram of processing procedure for 3D printed Al₂O₃/SiC whiskers composites^[63]

与直接 3D 打印多孔陶瓷基不同, Meng 等^[64]利用 3D 打印方法间接制备 SiC 导电屏/石英陶瓷多孔吸波材料。首先用 3D 打印技术制备网格结构的 PLA 模板, 然后通过逆复制模板法制备石英多孔结构, 再通过浆料浸渍法制得 ISCS/SO 陶瓷基结构吸波材料。通过 PLA 模板的结构来调整成型结

构的尺寸, 可实现材料整体结构电性能灵活设计。对浸渍后的 ISCS/SO 材料分别进行 800 °C、900 °C、1000 °C、1100 °C 下的后处理烧结。结果显示, 随着烧结温度的增加, 材料常温电性能降低; 在高温 800 °C 长时间可稳定工作, 实现电磁波的吸收, 短时 900 °C 材料可用(如图 7 所示)。

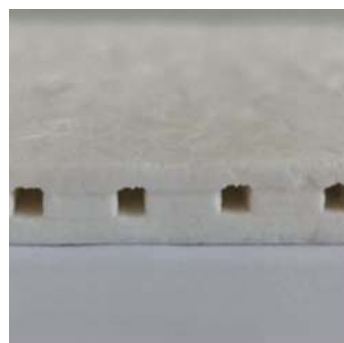


图 7 PLA 模板法打印陶瓷吸波体^[64]
Fig. 7 3D printed ceramics absorber by PLA template^[64]

Zhang 等^[65]使用 3D 打印技术首先制备出含短切碳纤维的 SiC 基陶瓷复合材料, 然后通过液态硅渗透法在 1650 °C 的真空环境下填充打印空隙, 使材料致密化, 最后进行整体烧结。打印原料为海藻

硅酸钠与陶瓷浆料的混合物,其中陶瓷浆料含有 SiC 陶瓷粉 ($D_{50} = 0.5 \mu\text{m}$)、炭黑 ($D_{50} = 200 \mu\text{m}$)、短切碳纤维 ($d=7\sim 10 \mu\text{m}$),通过调整成分占比和含量优化打印的浆料黏度和流变特性。结果表明,成型的 SiC 基陶瓷复合材料最大弯曲强度为 300 MPa,烧结线性收缩小于 2%。通过微观形貌观察,可以看到短切碳纤维在打印 Y 轴方向定向分散,根据碳纤维电磁损耗的原理^[66],材料沿纤维轴向的电磁波损耗最大(如图 8 所示)。

4 其他 3D 打印微波吸波材料

3D 打印技术还可用于制造其他类型的微波吸收材料。Zuo 等^[67]采用数字光处理(digital light processing, DLP) 3D 打印技术制备了石墨烯/羰基铁粉/PMMA 吸波体。当石墨烯含量为 1.0%,羰基铁粉含量为 47.8%,材料厚度 2.1 mm 时,制备的吸波体最佳吸波性能为 -54.4 dB,有效吸波带宽

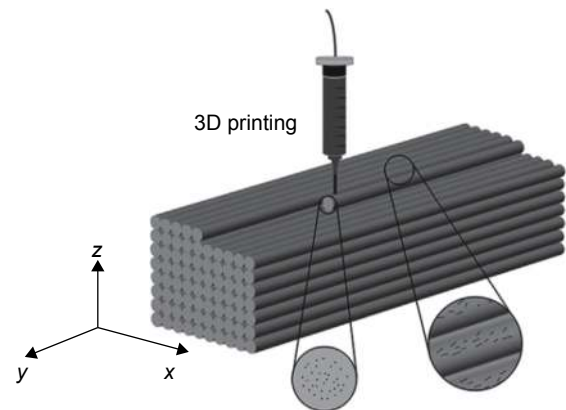


图 8 3D 打印 SiC 陶瓷复合材料示意图^[65]

Fig. 8 Schematic diagram of 3D printed SiC ceramic composites^[65]

($RL < -10 \text{ dB}$) 为 3.41 GHz。研究表明,数字激光处理打印工艺可提高石墨烯/羰基铁粉与纳米复合材料的分散性,进而提高系列体系的吸波效能。其 DLP 打印机理图和吸波机理图如图 9 所示。

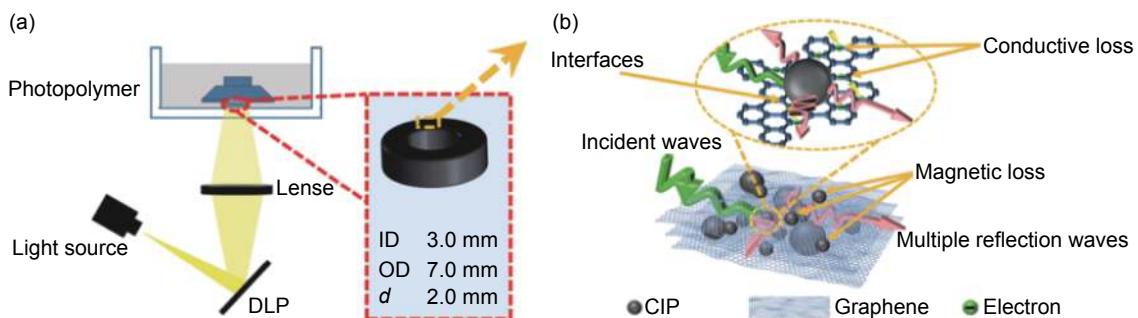


图 9 数字光处理 3D 打印制备吸波体^[67] (a)DLP 打印原理图;(b)石墨烯/羰基铁粉/PMMA 吸波材料吸波机理图

Fig. 9 3D printed absorber by DLP^[67] (a) schematic diagram of the DLP printing; (b) schematic diagram of microwave absorption mechanism of graphene/CIP/PMMA composites

Ren 等^[46]利用 3D 打印技术制备了一种由椭圆介电谐振器组成的光栅周期结构吸波体。打印材料为炭黑/ABS 混合物。基于谐振器的结构模式,吸波体在低频出现 3 个谐振峰。通过优化介电谐振器的尺寸,使得材料在 3.9~12 GHz 拥有宽频的吸收,吸收率大于 90%。同时材料在大角度斜入射时仍具有较好的吸波性能。

Liu 等^[47]利用 3D 打印技术制备一种阶梯尖劈形状的吸波体。尖劈结构为常见的吸波体设计形状,这种结构有利于材料对入射电磁波的阻抗匹配。通过优化常规尖劈结构,引入阶梯状结构,进一步提高了该结构的吸波性能。吸波体打印材料为羰基铁粉和 PLA 的混合物,打印喷嘴温度为 220 °C,平台设定温度为 50 °C,喷嘴口径为 0.6 mm,

打印速率为 20 mm/s。当吸收剂填充量为 70% 时材料的有效频带范围 ($RL < -10 \text{ dB}$) 为 12.9~18 GHz,当吸收剂填充量为 100% 时,材料的有效频带范围为 9.5~18 GHz,材料在斜入射角度为 70° 时,反射率在 12~18 GHz 时 $\leq -20 \text{ dB}$,显示了优异的斜入射吸收性能。

5 总结与展望

在微波吸收材料领域中,3D 打印制造工艺可以提高吸收剂与树脂基或陶瓷基的分散性,易加工不规则尺寸或周期性尺寸,同时因为成型特点,可实现快速高效制造,降低制造成本与周期。为工业化精准生产复杂结构体或特殊部位吸波材料提供

了一条可尝试的路线。但在这个新兴的制造领域,仍存在一些亟需研究解决的问题。

(1)打印材料的局限性。目前主要应用于吸波材料的介电打印材料为尼龙、ABS、PLA等,该类材料的玻璃化转变温度有时会低于材料应用环境温度,无法满足温度适应性。故需探索多种打印材料的开发,以提高制备材料对应用场景的丰富匹配性。

(2)缺少材料力学性能、微观结构方面的测试分析。各项研究都只针对图型设计和打印工艺实现进行研究,处于3D打印吸波体制造初期,对于成型材料的力学性能,微观无损检测等尚未全面开展,而吸波材料真正应用于工程领域时,除去电性能以外的其他物理性能也尤为重要且不可或缺。

(3)为应对未来小型化、多功能、智能化的趋势,3D打印吸波材料的尺寸精度和制造工艺应进一步提高。

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Progress of 3D printed microwave absorbers

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Abstract: In recent years, as the 3D printing technology growing maturity and commercialization, the researchers have attempted to apply this emerging manufacturing technology to the design and fabrication of wave-absorbing materials. In this paper, the recent progress of 3D printing technology in fabrication of microwave absorbing materials, including 3D printing FSS and metamaterial absorbing materials, 3D printing honeycomb absorbing materials, 3D printing ceramics and other 3D printing microwave absorbing materials are reviewed. Furthermore, the limitations of 3D printing materials, the lack of mechanical properties of materials, the problems of testing and analysis of microstructure of 3D printing technology in microwave absorption materials manufacturing are also systematically expounded, at the same time, the future developing trend of 3D printing technology in the manufacturing field of microwave absorption materials, such as miniaturization, multi-function and intelligent is also prospected.

Key words: 3D printing technology; microwave absorption; metamaterial absorbing; honeycomb microwave absorbers; ceramic matrix composites absorbers

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