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聚氨酯多孔吸声材料的制备、结构及性能研究进展

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摘要: 随着科学技术的迅猛发展, 噪声污染已成为人们生活的一大障碍, 高效吸声材料的研究已经成为热点研究领域。聚氨酯孔状材料具有优异的阻尼性能和发达的孔隙结构, 因此, 其作为声学材料基体在吸声降噪方面具有重要的应用价值。文中综述了国内外聚氨酯吸声材料的研究报道, 探讨了多孔材料的吸声原理, 主要介绍了聚氨酯吸声材料的制备、结构及吸声性能, 以及改性聚氨酯孔状吸声材料的研究进展, 主要包括以下方面: 填料改性聚氨酯、制备复合气凝胶、聚氨酯发泡以及形成多层发泡结构; 最后对聚氨酯在吸声领域的发展方向进行了展望, 并指出了存在的问题。基于现有分析认为, 在未来的研究中可以综合运用填料改性和结构设计双重机制对聚氨酯吸声体系性能进行调控, 以获得具有高阻尼高吸声性能的聚氨酯吸声材料, 以满足工业化、商业化应用的需求。

关键词: 聚氨酯; 吸声系数; 吸声材料; 发泡

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随着社会经济和科技的发展, 噪声污染日益严峻, 严重困扰了人们的生活。噪声不仅干扰了人们的睡眠与心理健康, 还会导致各种病症、学习困难甚至听觉损失等, 人们长时间暴露在这样的环境中, 将间接缩短寿命。为解决这一问题, 开发高效吸声材料势在必行。

目前对吸声材料的研究主要涉及吸声机理的探讨和吸声材料的制备。其中吸声机理主要通过分析材料的结构进而模拟吸声结构模型以预测材料的吸声性能, 而吸声材料的制备主要利用不同材料的结构特性实现对声能的高效吸收^[1]。吸声材料内部有大量互相贯通的微孔和间隙, 有较强的吸声降噪功能, 被广泛应用于国防军事、建筑材料、航空航天等各个领域。常见的吸声材料主要有共振吸声材料和多孔吸声材料。多孔吸声材料因其宽频吸声和密度轻等特点被广泛用于声学领域的研究。多孔吸声材料主要包括金属多孔、无机多孔和

有机多孔吸声材料。

一般情况下, 有机高分子吸声材料质轻、化学稳定性好、易加工^[2]。在众多有机高分子吸声材料中, 聚氨酯(PU)其组成单元多样且可控, 易成泡孔结构, 故以PU为原料制备的多孔材料吸声、阻尼和黏结性能优异, 被广泛应用于吸声材料的制备^[3]。但采用加发泡剂等传统工艺制备的PU多孔材料由于吸声频宽窄使其应用场景受限。因此亟待寻找新型工艺对PU材料进行改良以制备性能优异的吸声材料。鉴于此, 本文将主要通过填料改性PU、合成PU气凝胶、PU发泡以及设计多层次结构等方面对PU吸声材料的结构及性能进行详细讨论, 并展望PU吸声材料的发展方向。

1 吸声材料

1.1 吸声材料的分类

吸声即声波遇障碍物时, 被物体以结构振动或

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声学损耗等形式吸收耗散。基体材料的吸声机理主要包括黏滞损耗、波形转换和弛豫吸收^[4]。最常用的表征吸声性能的参数是吸声系数(SAC, α), α 表示吸收声能与入射总声能之比

$$\alpha = 1 - \frac{E_r + E_t}{E_i} = \frac{E_a}{E_i} \quad (1)$$

式中: E_i ——入射总声能; E_r ——反射声能; E_a ——吸收声能; E_t ——透射声能。同一种吸声材料在不同频率下 SAC 并不同, 因此常用平均吸声系数(ASAC)或特定频率下的最高吸声系数表征材料的吸声性能。

吸声材料一般分为两大类——共振吸声材料和多孔吸声材料。共振材料的低频吸声性能优异, 但有效吸收频带通常较窄^[5]。多孔吸声材料由于结构阻尼及介质孔隙中的热效应和黏性效应而具有宽吸声频带和优异的吸声性能, 且密度小、比强度高、取材范围广^[6]。常见的吸声材料及其相关性能如 Tab.1 所示。

多孔吸声材料根据组成为金属、无机和有机类。金属多孔吸声材料开孔率低、内外孔连通性差、孔隙分布不均, 质量较重且不易加工^[18]。无机多孔

Tab. 1 Common sound absorption materials and related properties

Classification	Matrix	Type	SAC	Characteristics of composite materials	Reference
Organic Synthesis	Polyurethane (PU)	Foam	Peak: 0.98 average: 0.71 (300~6400 Hz, thickness 10 mm)	Excellent viscoelasticity, damping, mechanical and sound absorption properties, developed pore structure, light weight, easy processing and poor flame resistance	[7]
	Polypropylene (PP)		Peak: 0.95 (100~2500 Hz, thickness 40 mm)	Excellent mechanical, chemical and thermal properties and easy to deform	[8]
	Melamine	Foam	Peak: 0.96 (250~6300 Hz, thickness 20 mm)	Excellent sound insulation and absorption performance, light weight and high cost	[9]
	Epoxy resin (EP)		Peak: 0.55 (1000~6300 Hz, thickness 2.71 mm)	Easy to cure, special chemical stability, poor toughness and low heat resistance	[10]
	Polystyrene (PS)	Fibrous membranes	Peak: 0.97 average: 0.47 (80~6300 Hz)	Light weight, good sound absorption ability, poor chemical stability and corrosion resistance	[11]
	Ethylene vinyl acetate (EVA)		Peak: 0.91 average: 0.72 (500~6400 Hz, thickness 40 mm)	Good chemical stability, aging resistance and ozone resistance	[12]
	Xylogen		Average: 0.12±0.025 (500~6400 Hz)	Renewable, degradable, good adhesion and surface activity, poor water solubility and difficult separation	[13]
Natural	Chitosan	Aerogel	Average: 0.59 (1000~6300 Hz, thickness 30 mm)	Renewable, good biocompatibility and microbial degradation, poor flame retardancy and weak mechanical properties	[14]
	Glass ceramics	Foam	Average: 0.40 (100~4000 Hz, thickness 10 mm)	Broadband sound absorption, light weight, high strength and complex structure	[15]
Inorganic	Pervious concretes		Average: 0.44 (100~4000 Hz)	High porosity, strong energy absorption capacity, good skid resistance and high cost	[16]
	Silica	Aerogel	Peak: 0.88 (100~6300 Hz)	High specific surface area, high mesoporosity, ultra- low thermal conductivity and low mechanical strength	[17]

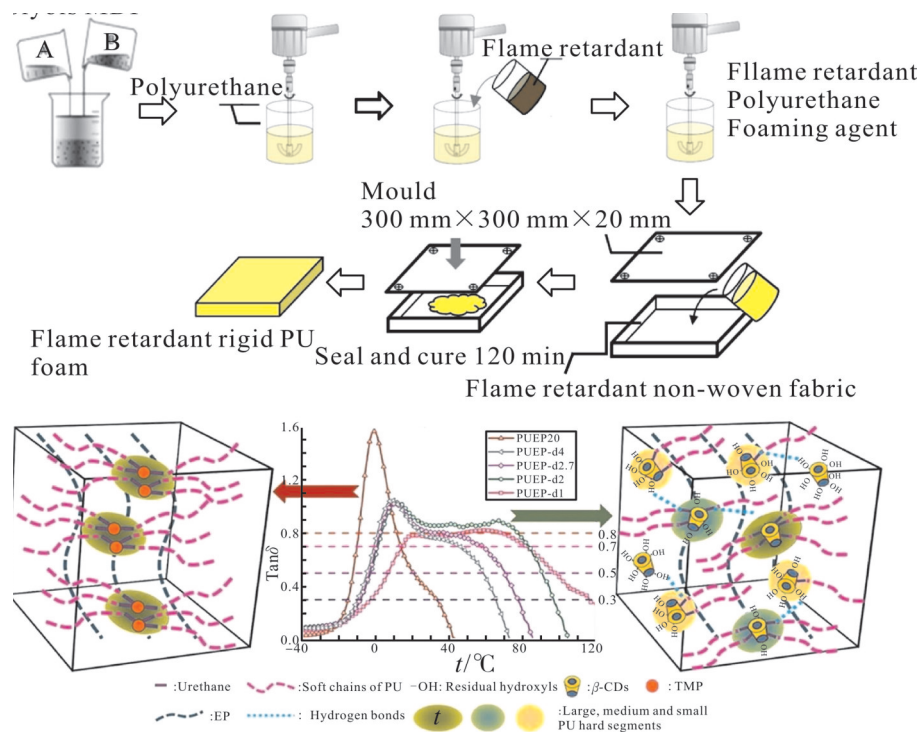


Fig.1 (a) Preparation technology of polyurethane foam^[24]; (b) damping properties of polyurethane composites^[25]

吸声材料阻燃和吸声性能优良、使用寿命较长^[16],但在应用中易破碎形成粉尘,且制备能耗高。而有机多孔吸声材料的弹性高、可塑性强、耐磨、耐腐蚀、气密性好、密度小、易加工,且吸声性能优越^[19]。

1.2 聚氨酯吸声材料

在常见的有机高分子材料中,PU因其组分中异氰酸酯和多元醇的比例可控、易成泡孔结构,且易于加工成型,常被用作多孔吸声材料的基体。PU孔状材料由于其优异的阻尼性能(见 Fig.1(b))、发达的与外界连通的内部孔道展现出良好的吸声性能,且质轻、加工性好,性能可调范围广,广泛用于工业、办公室和录音室等需要降噪的场所^[3,20]。Kim等^[21]研究发现,减小泡孔尺寸,增加泡沫弯曲路径,可提高PU泡沫的阻尼性能,进而改善吸声性能;SAC与气流阻力呈现正相关。Bhingare等^[22]研究发现,添加PU树脂的椰壳纤维最高吸声系数可达0.93。游峰等^[23]以PU为基体、磷酸三甲酚酯(TCP)为增塑剂、添加中空玻璃微珠(HGB)和偶氮二甲酰胺(AC发泡剂),采用熔融共混法制备了软质PU发泡材料,实验验证这种PU多孔材料具有良好的力学性能和吸声减振效果。

目前,研究者们不断探索制备PU吸声材料的新

方法与新工艺,以进一步提高PU材料的吸声性能。如 Fig.1(a)所示,采用加发泡剂、合成过程中发泡等传统发泡工艺制备PU复合吸声材料吸声效果增强并不明显。因此,如何有效地对PU进行改性来大幅提高其吸声性能已成为热点研究方向。

2 改性聚氨酯吸声材料

2.1 聚氨酯多孔吸声材料

目前,多孔PU吸声材料制备方法主要有加发泡剂、合成PU过程中发泡、形成互穿网络结构发泡及加中空孔隙结构材料等4种方法。

2.1.1 聚氨酯加发泡剂:发泡剂可分为化学发泡剂、物理发泡剂和表面活性剂。传统发泡工艺产生的泡孔孔径均一性差,吸声效果较差。王康等^[26]发现,先利用物理发泡剂产生部分孔结构,再使用化学发泡剂产生更多的孔结构,并能将部分孔结构涨破,制备的多孔材料具有丰富的开孔结构(见 Fig.2(a)和 Fig.2(b))。

Liu等^[27]采用自由发泡法在PU泡沫中添加不同含量的气相二氧化硅制备了不同负载质量分数的PU泡沫塑料。随着气相二氧化硅含量增加,PU泡

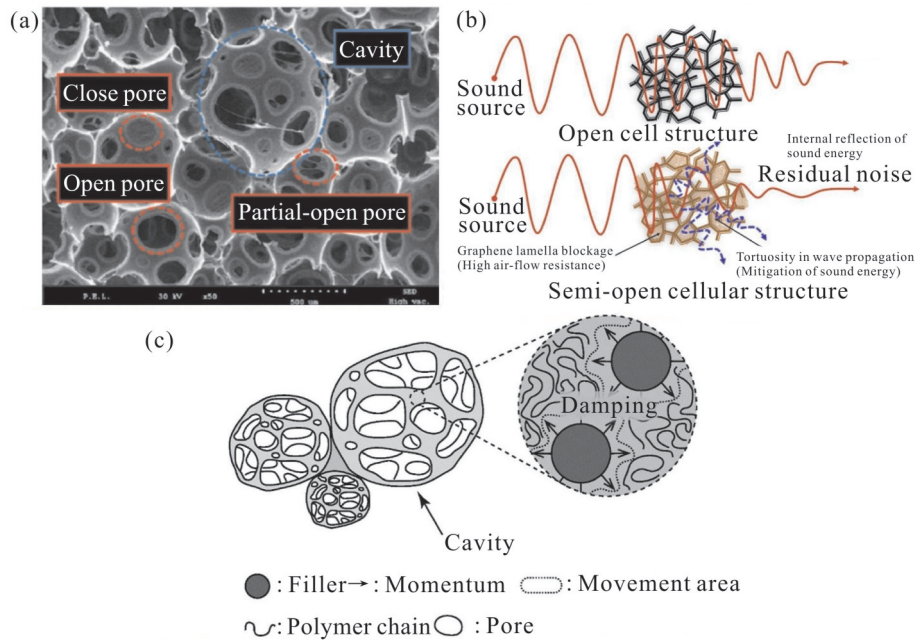


Fig.2 (a) SEM image defining areas for different three types of pores (open, partial-open and close) [29]; (b) schematic diagram of acoustic propagation through open and semi open structures [30]; (c) schematic illustration of mechanical damping effects of polyurethane foams [28]

沫的吸声峰向低频处移动,且最佳ASAC相比纯PU泡沫提高了5%左右。Sung等[28]在PU泡沫中加入氢氧化镁填料,增强了复合泡沫的阻尼运动(见Fig.2(c))和开孔的数量,制备的PU泡沫最佳开孔孔隙率达63%,最高SAC达0.53。

Gwon等[31]研究了二月桂酸二丁基锡(DBTDL)和三乙烯二胺(DABCO)2种凝胶催化剂对PU泡沫材料的影响。发现使用DBTDL制造的软质PU泡沫具有更多的小孔,吸声效果更好。Büyükakinci等[32]发现,将棉、竹、毛纤维发泡剂加入PU泡沫塑料中可增强材料的强度。

以上研究表明,在PU中加入合适的发泡剂,通过控制发泡剂的含量、类型可改变孔隙率、泡孔数量、尺寸及结构,从而制备出吸声性能优异的多孔吸声材料。

2.1.2 合成聚氨酯过程中的发泡:通过改变PU组成的结构及发泡方式,可改善PU热、力学和吸声性能。异氰酸根可与水反应生成 CO_2 ,故合成PU发泡吸声材料常以水为发泡剂[33]。反应后期体系黏度增大,水难以均匀混入其中,进而影响材料吸声性能,故需加入其他填料改性。

Zhu等[34]研究发现,在PU泡沫中加入不同含量和硬度的三元乙丙橡胶(EPDM)颗粒后,泡沫材料孔隙变小,密度增大,PU复合材料在中低频区域展现出较好的吸声性能。Soto等[35]以桐油衍生的甘油(gLY)或羟基化甲酯(HMETO)为多功能多元醇、废轮胎颗粒(WTP)为填料制备了柔性PU泡沫塑料(FPF),发现其有开孔和闭孔结构,泡孔尺寸减小,正入射SAC在500 Hz以内可达到0.5。

通过使用水为发泡剂并加入其他催化剂,可实现PU泡沫的自发泡成型,进而制备出孔隙均匀、贯通性好、声学性能优异的PU吸声材料。这种方法简便易行,有助于提高生产效率,降低生产成本。

2.1.3 互穿网络结构:PU分子链中含有大量的活性基团,有优异的弹性及耐寒性,通过调节PU软硬段间的比例可获得不同的力学性能。但单一PU材料的耐水性、力学强度不足,不同链段间的热力学稳定性也存在差异。

互穿聚合网络(IPNs)是一种将起强迫互溶和协同效应的聚合物相互贯穿形成交织网络聚合物的技术。如图3所示,各组分链间互相缠接,使相组织微细化,提高了相间结合力,这为制备综合性能优异

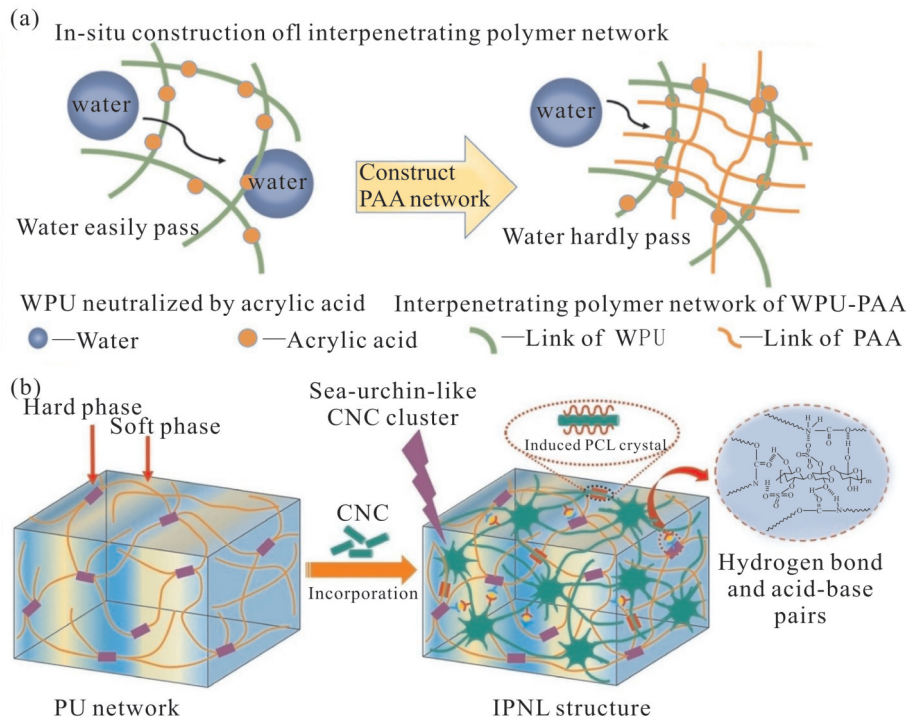


Fig.3 (a) Schematic diagram for the construction of PU IPN^[36]; (b) schematic diagram for the strengthening mechanism of class IPN (ipnl) on polyurethane composites (CPCs)^[37]

的PU吸声材料提供了一种有效途径^[36,37]。

Moradi等^[38]采用聚合法制备了PU与聚甲基丙烯酸甲酯(PMMA)不同配比的互穿网络泡沫。发现当IPN形成时,玻璃化转变温度(T_g)向高温区移动,阻尼温度范围增加。随着PMMA含量的增加,共振频率向低频方向移动。

庄建煌^[39]将模量较高、耐腐蚀性好的EP与PU合成IPN膜,发现当PU/EP质量比为7:3时,材料的损耗因子 $\tan \delta$ 可达0.96,IPN膜可显著提高材料的阻尼、热力学和低频吸声性能、力学及黏结强度。

综上所述,采用IPNs技术可将PU与其他化合物的优点结合,获得声学及阻尼性能优异的PU复合材料。

2.1.4 中空孔隙结构填料:中空结构质轻、多孔、比表面积大、化学稳定性高且声学性能优异^[40]。林健等^[41]使用硅藻土和PU成功制备出具有多孔径连通孔道的硅藻土/PU吸声材料,发现其特定频率下SAC可达0.9以上。Yu等^[42]采用浇铸法将表面改性的316L不锈钢空心球加入PU中制备出金属-空心球/聚氨酯(MHSP)复合材料,发现声波在PU的孔隙、空

心球的空腔和界面中的摩擦、黏性和反射使声能转化为热能而耗散(见Fig.4)。当空心球体积分数为60%,直径为2.5 mm时,MHSP复合材料在1518 Hz时的SAC达0.97。此外,中空结构可改变PU的弹性模量,导致材料和空气的共振频率向低频移动。以上研究表明,将中空结构填充到PU泡沫中,可同时实现吸声和力学协同增强的效果。此类研究为吸声材料的进一步发展和应用提供了参考。

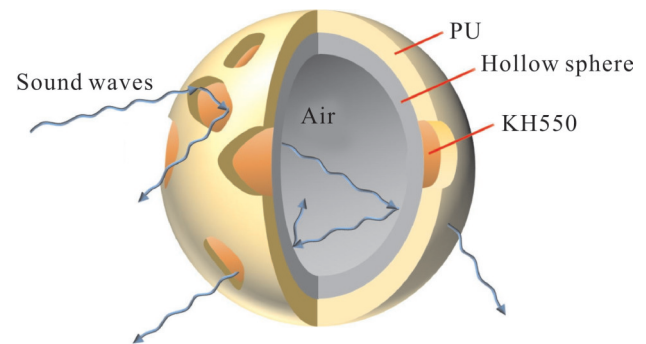


Fig.4 Sound absorption diagram of MHSP composite material^[42]

2.2 聚氨酯气凝胶

除了上述PU发泡工艺,还可添加填料制备气凝胶以改善吸声性能。制备气凝胶通常是先溶胶-凝

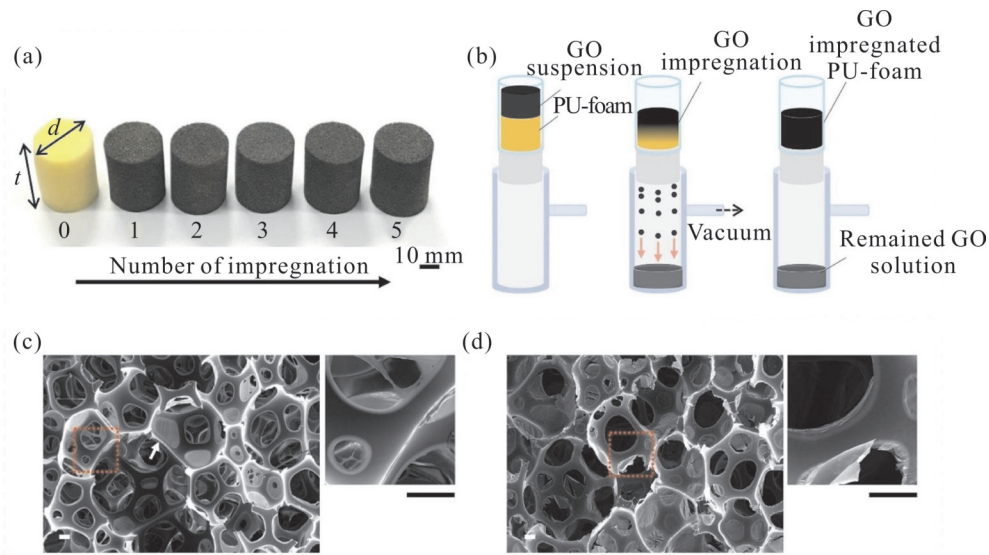


Fig.5 (a) Photo of graphene-oxide impregnated polyurethane foam (GO-PU32) with $d=30$ mm, $t=30$ mm, with number of treatments increasing from 0 to 5; (b) illustration of vacuum assisted impregnation of GO into PU foam; (c) SEM image of pristine PU32; (d) image of PU32 after five impregnations: GO-PU32 (22.8%)^[43]

胶,后干燥处理。气凝胶是目前世界上最轻的固体,有高孔隙率(约为99%)、高比表面积、低密度(最低密度可达 0.16 kg/m^3)和良好的声阻抗(100 m/s 的低声速),这些特点使气凝胶在吸声领域展现出巨大的优势和潜力^[17]。

2.2.1 石墨烯改性聚氨酯气凝胶:石墨烯比表面积大、密度低、模量高、有自修复功能,可作为提高气凝胶力学性能的填料^[43,44]。

如图5所示, Lee等^[43]采用真空辅助法将氧化石墨烯(GO)浸渍到PU泡沫中。发现GO可填充PU泡沫的大量孔洞。控制GO的浸渍密度可在特定频率范围内实现吸声性能最优化。在体积密度为 51 kg/m^3 、厚度为 5 mm 的PU泡沫中加入质量分数15%的GO, ASAC($800\sim 6300 \text{ Hz}$)提高了4倍以上。

Lee等^[44]提出了一种多层GO浸渍PU(GO-PU)的泡沫结构。优化后 54.6 mm 厚的GO-PU多层结构在 500 Hz 以上的ASAC提高到0.9以上。Oh等^[45]制备了定向拮抗GO-PU复合气凝胶,发现控制GO层的内晶胞尺寸和排列结构可显著改善低频吸声性能,这种改善归因于入射和反射波在多孔表面的多次散射及PU泡沫和GO网络间互联结构中的空气黏滞阻尼。

此外,研究表明,调整聚醚多元醇、异氰酸酯的

分子结构和相对分子量等可获得弹性模量、气密性与分子弛豫不同的PU材料,再与碳纳米管/石墨烯/类石墨烯结合可形成不同结构的性能优异的复合材料。

综上所述,添加石墨烯填料、控制石墨烯浸渍密度、改变GO的结构、PU模量与分子弛豫均可制备吸声性能优异的气凝胶,同时可改善材料的力学性能。

2.2.2 纤维素改性聚氨酯气凝胶:纤维素是一种天然的高分子聚合物,可降解可循环。以其为填料制备的气凝胶具有可持续性、环境友好、孔隙率高、比表面积大、堆积密度低及阻燃和吸声性能优异^[46]等优点,使得纤维素基气凝胶广泛用于声学等诸多领域。

Çelebi等^[47]研究发现,向软质泡沫中添加24%的茶纤维增强剂可使复合材料的吸声特性提高60% ($2500\sim 6300 \text{ Hz}$)。Ekici等^[48]研究发现,在PU泡沫中加入8-16%的茶纤维可提高所有频率范围内的吸声值。Bahrambeygi等^[49]在PU泡沫中加入聚丙烯腈或PU纳米纤维层,获得了吸声性能优异的吸声材料。Tao等^[50]开发了稻草纤维填充PU泡沫(RPUF)和麦草纤维填充PU泡沫(WPUF),发现开孔数量与纤维含量成正相关,当添加量为10%时, WPUF和RPUF的ASAC均显著提高。

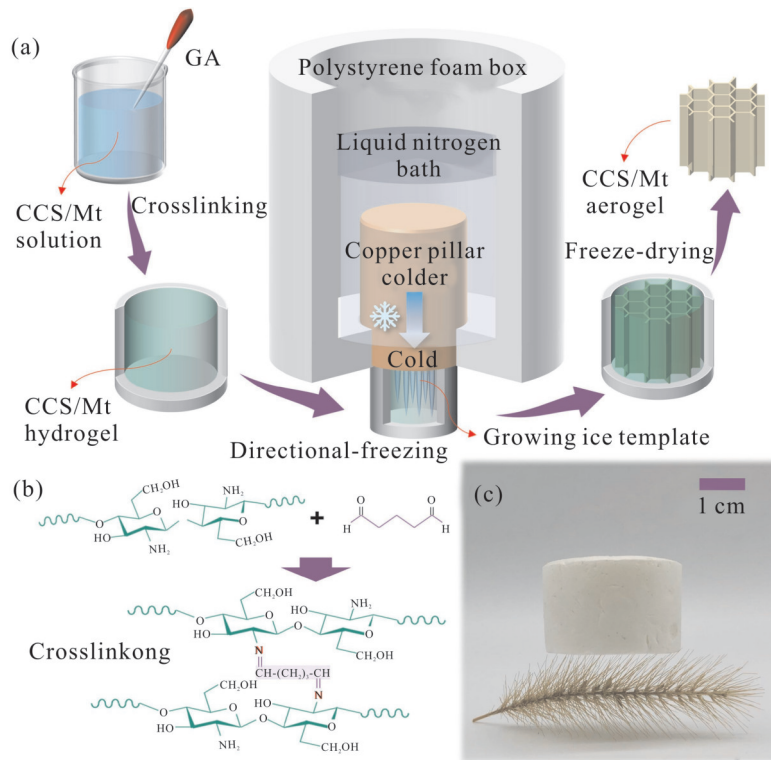


Fig.6 (a) Preparation route for CCS/Mt. aerogel; (b) crosslinking reaction of CCS and GA; (c) as-prepared CCS/Mt^[14]

以上研究表明,添加纤维素或者生物质纤维填料、控制纤维含量和种类可制备孔隙率高、阻燃及吸声性能优异的PU复合气凝胶。

2.2.3 其他改性聚氨酯气凝胶:除了通过添加石墨烯和纤维素来制备改性PU气凝胶,还可通过添加其他比表面积大、化学稳定性优异、易制备的填料改善PU气凝胶的吸声性能,其中蒙脱土(Mt)较为常见。Mt是一种多层状物质,有较大的改性空间,且比表面积较高、长径比大、化学稳定性优异。可通过有机改性增加其层间距,有利于PU进入Mt片层中改进吸声性能^[51]。

本课题组通过调节羧甲基壳聚糖(CCS)和Mt配比调控孔隙结构进而改善了气凝胶低频吸声性能。如图6所示,通过定向冷冻法和冷冻干燥工艺获得了有超低密度(33.85~38.27 mg/cm³)和高孔隙率(98.20%)的分层多孔结构。其在250~2000 Hz范围内ASAC可达0.59^[14]。此外,Liu等^[51]还发现在PU中加入改性Mt可改善PU材料的耐火及阻燃性能。

以上研究表明,在PU中加入不同配比的Mt,通

过控制气凝胶厚度可提高PU孔状材料的阻燃性,使吸声效果增强,这为制备高阻尼性、高阻燃性、高孔隙率、吸声性能优异的多孔气凝胶提供了新的研究方向。

2.3 聚氨酯层状发泡复合吸声材料

研究表明,结构会对吸声性能产生显著影响,单一的PU多孔材料存在低频吸声弱的缺点,许多报道表明,通过发泡PU层状结构设计可提高其宽频带的吸声性能。本课题组Jiang等^[52]将钛酸钡/丁腈橡胶(BT/NBR)与PU泡沫复合制备了多层交替结构的吸声材料,其具有良好的低频吸声性能。这种交变多层结构提供了更多的界面,使声音传播路径变长。Qi等^[7]将海藻酸钠(SA)和PU泡沫与3D间隔织物相结合设计出B&M-L复合材料。强共振与多孔结构相结合的方式可显著改善材料吸声性能(见Fig.7)。Li等^[53]以PU泡沫为中间层,针刺复合织物为上层,尼龙织物为底层,构建了PU泡沫夹层复合材料。其在450 Hz时SAC最大值为0.78。织物-泡沫界面的存在有利于PU泡沫夹层复合材料的低频吸声。

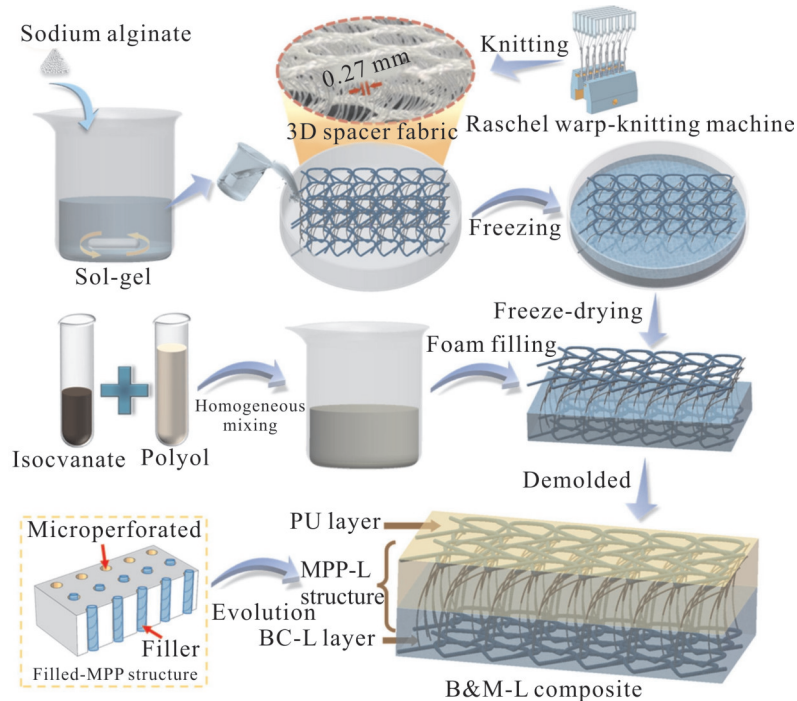


Fig.7 Process of preparing B&M-L composite^[7]

3 结语

近年来,伴随着科学技术和现代工业的发展,吸声材料广泛应用于陶粒吸声板制作、民用建筑、飞机和汽车制造等行业。PU孔状复合材料由于优异的阻尼和声学性能、质轻等特点被广泛应用于吸声材料的制备。本文结合课题组的研究工作重点总结了PU吸声材料的制备、结构及吸声性能的关系,比较了不同PU孔状吸声材料的特点。结合研究现状,改性PU复合吸声材料的研究应从下述几个方面改进:(1)在PU单体中添加氮、磷元素,通过调控氮、磷元素比例,在提高PU吸声性能的同时提升材料的阻燃性能。(2)改变PU软硬段组成,从而调控PU的 T_g 及模量等,增加PU吸声材料的黏弹性和力学性能。(3)通过多层次结构设计形成能够在特定温度条件下可调控的具有高阻尼高吸声性能的复合材料。(4)通过添加中空结构进行整体优化设计制备性能优异的PU复合吸声材料,增加声波吸收时的路径,使声能通过阻尼损耗转换为热能而耗散。

尽管PU吸声材料发展迅速并且取得了一定的成果,然而仍然存在一些问题:(1)无法通过一种混合改性方式同时解决PU的基体阻燃性能较差、不同软硬段之间存在差异性等问题。(2)目前所制备的

吸声材料均只能在一定频率内保持高吸声性能,制备在所有频率范围内都具有高SAC、同时保持材料的最小厚度和轻质的PU吸声材料仍是一个巨大的挑战^[1]。(3)PU多孔吸声材料废弃物占用较大的空间资源($20\sim 50\text{ kg/m}^3$),且PU可在垃圾填埋场中进一步水解产生有毒胺,导致环境污染^[54]。(4)改性PU吸声材料大多还在实验室探索阶段,需要大量实验寻找最佳改性条件,成本高、工艺复杂,从实验室研究到实际生产还有一定的距离。解决这些问题将进一步提高PU吸声材料的性能,具有极大的研究价值和广阔的应用前景。

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Progress in Research of Preparation, Structure and Properties of Porous Polyurethane Sound-Absorbing Materials

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ABSTRACT: With the rapid development of science and technology, noise pollution has become a major barrier for residents' happy life, and the research of efficient sound-absorbing materials has become a hot research field. Polyurethane porous material has excellent damping properties and developed pore structure. Therefore, polyurethane has important application value in sound absorption and noise reduction as acoustic material matrix. In this work, the research reports of polyurethane sound-absorbing materials at home and abroad were reviewed and the sound-absorbing principle of porous materials was discussed. The paper mainly introduced the preparation, structure and sound-absorbing properties of polyurethane sound-absorbing materials. In addition, the research progress of modified porous polyurethane sound-absorbing materials was introduced, which mainly included the following aspects: modified polyurethane with fillers, preparation of composite aerogel, polyurethane foam and formation of multi-layer foam structure. Finally, the problems concerning the development direction of polyurethane in the field of sound absorption were briefly prospected. Based on the existing analysis, it is concluded that filler modification and structural design should be comprehensively used to regulate the performance of polyurethane sound absorption system in the future research, in order to obtain polyurethane sound absorption materials with high damping and high sound absorption performance, which could meet the requirements of industrial and commercial applications.

Keywords: polyurethane; sound absorption coefficient; sound-absorbing materials; foaming