Material Sciences 材料科学, 2022, 12(5), 419-428
Published Online May 2022 in Hans. http://www.hanspub.org/journal/ms
https://doi.org/10.12677/ms.2022.125044

纳米$Mg(OH)_2$在酸性气体捕集和水中污染物去除的研究进展

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收稿日期：2022年4月8日；录用日期：2022年4月28日；发布日期：2022年5月5日

摘要

纳米$Mg(OH)_2$是一类新型绿色环保无机纳米材料，广泛应用于环境、材料、增强、阻燃等多个领域。目前致力于研究利用成本低、绿色环保和广泛可用的镁资源来合成高质量的纳米$Mg(OH)_2$材料。将低价值镁盐转化为高价值纳米$Mg(OH)_2$材料是减少资源浪费和实现可持续技术发展的有效途径之一。本文主要论述了纳米$Mg(OH)_2$材料常见的制备方法及其作为吸附剂在环境领域中酸性气体和水中污染物的应用。

关键词

纳米$Mg(OH)_2$，制备方法，酸性气体，水中污染物

Nano $Mg(OH)_2$ in Acid Gas Capture and Pollutant Removal in Water: A Review

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Received: Apr. 8th, 2022; accepted: Apr. 28th, 2022; published: May 5th, 2022

Abstract

Nano-$Mg(OH)_2$ is a new type of green and environmentally friendly inorganic nanomaterials, which are widely used in many fields such as environment, materials, reinforcement, and flame retardancy. At present, the research is devoted to the synthesis of high-quality nano-$Mg(OH)_2$ materials using low-cost, green and widely available magnesium resources. Converting low-value magnesium salts into high-value nano-$Mg(OH)_2$ materials is one of the effective ways to reduce resource waste and achieve sustainable technological development. This paper mainly discusses

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文章引用: 龚露, 姚贞英, 朱春梅, 余达伟, 何柏, 余波. 纳米 $Mg(OH)_2$ 在酸性气体捕集和水中污染物去除的研究进展[J]. 材料科学, 2022, 12(5): 419-428. DOI: 10.12677/ms.2022.125044

the common preparation methods of nano-$Mg(OH)_2$ materials and their applications as adsorbents in acid gas and water pollutants in the environmental field.

Keywords

Nano-$Mg(OH)_2$, Preparation Method, Acid Gas, Pollutants in Water

1. 引言

当下，工业快速发展造成的环境污染、能源短缺等问题是掣肘我国乃至世界经济发展的关键。其中大气污染和水资源污染日益严重，温室效应、酸雨、赤潮、绿潮等现象引起了人们对环境问题的高度重视[1] [2] [3]。无机纳米材料具有高比表面积，无内部扩散阻力，扩散路径短，高孔隙率，增强结构性能，催化性能等特性，此外还可通过改性为特定应用定制的活性表面官能团[4] [5]。这些特性使纳米吸附剂具有更好的选择性、容量和对污染物的亲和力，也提供更快和高效的吸附过程[6]。由于这些特性使得无机纳米材料在环境领域一直受到大量关注。

纳米 $Mg(OH)_2$ 是一种新型无机纳米材料，具有表面与界面效应、小尺寸效应、量子尺寸效应等纳米材料的共性特点，此外，还具有热稳定性好、无毒无害、吸附能力强等优点，使其在环境领域受到极大的关注[7] [8]。在废气污染治理中，可用于 H_2S、SO_2、NO_x、CO_2 等酸性气体治理[9] [10] [11]。此外，纳米 $Mg(OH)_2$ 在废水污染治理中应用较为广泛，可用于赤潮、绿潮防控、重金属阳离子脱除、印染废水处理、药物废水处理等[12] [13] [14] [15]。本文以纳米 $Mg(OH)_2$ 为研究对象，归纳总结纳米 $Mg(OH)_2$ 在环境领域最新研究进展，旨在为今后纳米 $Mg(OH)_2$ 作为新型吸附剂在环境领域中的开发和利用提供参考和借鉴。

2. 纳米 $Mg(OH)_2$ 材料的制备

自然界中存在天然氢氧化镁矿，其形态多呈板块状，少数呈纤维状[16]。可直接采用机械粉碎法将矿石用超细粉碎机直接粉碎研磨成纳米粉，但现下国内的粉碎工艺加工得到的产品一般只能得到颗粒尺寸较大的初级粉体，粒度不易被控制且储量少、含量低分离提纯困难[17]。

当前合成纳米 $Mg(OH)_2$ 材料的主要制备方法是液相化学法[18] [19]，传统的液相法有沉淀法和水热法，此外还有氧化镁水化法，间接转化法等。沉淀法[20]通常是用镁盐溶液和碱($Na(OH)_2$、$NH_3{\cdot}H_2O$、CH_4N_2O)为原料，其中常用的镁盐有 $MgCl_2{\cdot}6H_2O$、$MgSO_4{\cdot}7H_2O$、$Mg(NO_3)_2{\cdot}9H_2O$。沉淀法相对于其他方法来说制备工艺简单、对过程条件要求不高、能大批量工业生产，是生产纳米 $Mg(OH)_2$ 的普遍工艺手段之一。水热法[21]是指含 Mg^{2+} 的溶液与碱性溶剂在高温高压的密闭容器中进行的反应。水热法可提高反应效率，增加产率。相比较之下，水热法合成的 $Mg(OH)_2$ 颗粒分散程度高，过程可控，不易团聚。氧化镁水化法[22]生产制备 $Mg(OH)_2$ 的起源较早，是指轻烧镁粉(MgO)在水溶液中水化转化生成 $Mg(OH)_2$。制备工艺简单，生产过程中不会额外产生其它对环境有害的物质，而且氧化镁转化率高。但是产物纯度取决于原料氧化镁的纯度。间接转化法[23] [24]一般先采用水热或常温反应法制备介稳态镁盐晶须(碱式硫酸镁或碱式氯化镁晶须)前驱体，再通过转化反应制备氢氧化镁，所得产物能保留晶须形貌。

3. 酸性气体的捕集

根据国家环境空气质量标准，酸性气体是主要的空气污染物，其中包括一氧化碳(CO)、氮氧化物(NO_x)和硫化物(SO_x、H_2S)等，主要来自于化石燃料的燃烧、工业废气或者火山爆发[25] [26]。这些酸性气体在大气中大量的存在，会导致酸雾、酸雨、雾霾等环境问题，对人类身体健康及生态系统有很高的危害[27] [28]。此外二氧化碳(CO_2)作为主要的温室气体，对全球变暖和气候变化起到了显著的作用。近年来碳中和、碳达峰被大力倡导。

捕获酸性气体可以改善空气质量，减轻空气污染，降低大气环境压力，并有利于资源再生。因此酸性气体的捕集技术被大量研究，其中 $Mg(OH)_2$ 作为一种碱性材料被广泛应用于酸性气体的去除和捕集当中。Salehi 等[29]采用化学气相沉积法将 Al_2O_3/SiO_3 陶瓷泡沫过滤器与 $Mg(OH)_2$ 浸渍，制备了吸附剂。阐述了一种烟气脱硫的综合吸收/吸附工艺对于高硫汽油/石脑油燃料的燃烧产生的烟气强化脱硫的综合吸附/吸收过程。$Mg(OH)_2$ 溶液不断回流并喷在烟气上，加强脱硫过程。结果表明，用 $Mg(OH)_2$ 溶液取代纯水时，总除硫率提高了约 3%。$Mg(OH)_2$ 对除硫效果有积极的影响。Hanif 等[10]报告了 Mg-Al 层状双氢氧化物(LDHs)，作为高容量吸附剂来减少环境中的 NO_2。对材料进行了分层处理，增加了材料活性碱性部位，分层 LDH 材料(LDH-am)的二氧化氮吸附能力为 8.52 mmol/g，显著高于其他的稳定吸附剂，如沸石(0.36~3 mmol/g)和碳基吸附剂(2~6 mmol/g)。表征发现二氧化氮同时取代 LDH 层间 CO_3^{2-} 离子，吸附在 Mg-OH 和 Al-OH 表面和层内。

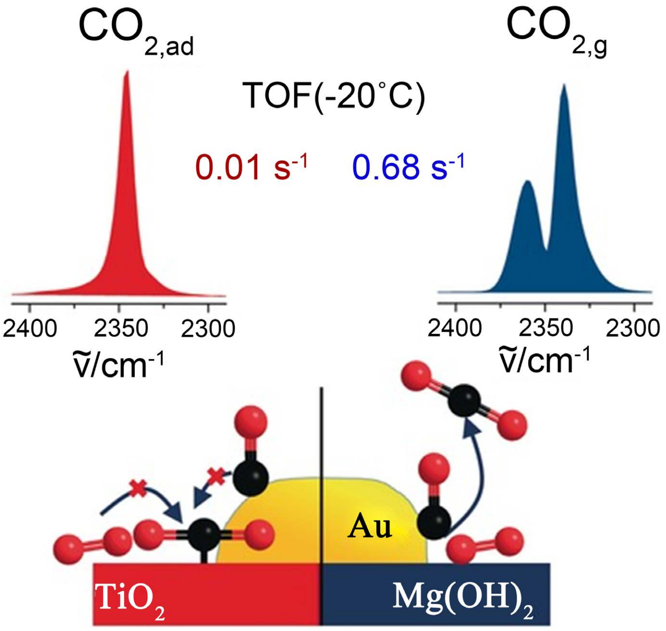

Figure 1. A Key Feature for the High Activity of Au/$Mg(OH)_2$ Catalysts in Continuous Low-Temperature CO Oxidation [30]
图 1. Au/$Mg(OH)_2$ 催化剂在连续低温 CO 氧化中高活性的关键特征

Wang 等[30]报道，Au/$Mg(OH)_2$ 催化剂在催化低温 CO 氧化(低于 0℃)中的活性远远高于已深入研究的 Au/TiO_2 催化剂。其催化机理如图 1 所示。证明了在低温反应过程中，Au/$Mg(OH)_2$ 与二氧化碳相对较弱的相互作用是催化剂性能优越的主要原因。这一特性使产品快速解吸，因此在温度远低于 0℃下连续氧化。在这些温度下，Au/TiO_2 也能催化 CO_2 的形成，但不允许 CO_2 的解吸，从而导致自我中毒。然而，在较高的温度(0℃以上)，CO_2 的形成是速率限制的，这导致在这些反应条件下 Au/TiO_2 具有更高的活性。

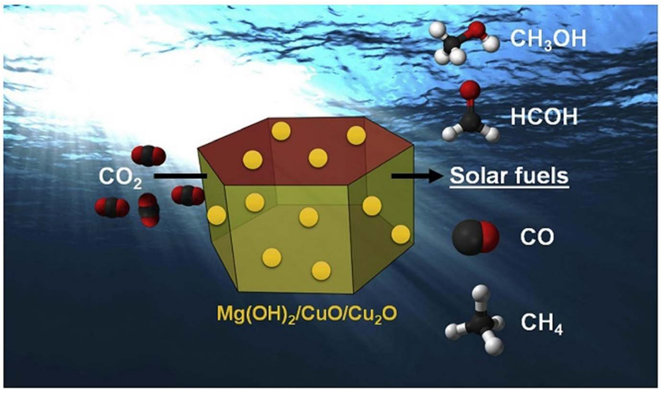

Figure 2. CO_2 adsorption and photocatalytic reduction over $Mg(OH)_2/CuO/Cu_2O$ under UV-Visible light to solar fuels [31]
图 2. 紫外可见光下对 $Mg(OH)_2/CuO/Cu_2O$ 的吸附和光催化还原

M. Flores-Flores [31]采用微波－水热法对 $Mg(OH)_2$、氧化铜和氧化铜粉末进行有效耦合，提出了在紫外－可见光照射下捕获和光催化转化二氧化碳的双功能材料，反应过程如图 2 所示。这是为了利用 $Mg(OH)_2$ 的高 CO_2 吸附能力和 CuO/Cu_2O 良好的光催化性能，在液体(甲醇和 HCOH)和气相(甲烷和 CO)中将 CO_2 还原为太阳能燃料。

纳米 $Mg(OH)_2$ 材料在捕集酸性气体时表现出了十分优异的吸附容量和吸附效率，是一种非常有前景、耐用、可扩展的酸性气体吸附剂，可用于从环境中捕获各种污染物酸气体，能有效被应用于环境改善当中，实现资源再利用。但有些吸附过程对于工艺条件要求相对较高，更加经济，节能的纳米 $Mg(OH)_2$ 材料的制备方法及捕集酸性气体的过程工艺还有待开发。

4. 水中污染物的去除

近年来，随着工业化和全球化的快速发展，越来越多的工业废水被排放到环境中，其中对环境造成污染的物质主要包括重金属离子、无机阴离子、染料废水、药物废水以及其它一些有毒有害物质[32] [33]。目前的废水处理技术难以满足无害废水排放的所有实际要求，因此迫切需要探索和开发处理各类废水的新技术[34]。纳米 $Mg(OH)_2$ 材料作为一种新的绿色无机纳米材料，被广泛应用于水中污染物的去除。

4.1. 无机物

不同的重金属离子通过涂料、金属制造、皮革、电池、合金化、电镀、采矿等各种行业的废水排放到自然水体中，如 Pb^{2+}、Cr^{6+}、Mn^{2+}、Ni^{2+}、As^{5+}、Cd^{2+}、Hg^{2+}等[35] [36]。它们是极其有害的水污染物，会对生物体造成严重的副作用。长期过量摄入重金属离子也会损害肾、肝、脑功能和神经系统，除此之外，对水生生态系统造成巨大威胁，并对现有动植物造成严重的破坏和不良影响[37]。纳米 $Mg(OH)_2$ 材料比表面积大，表面活性位点丰富，其吸附重金属离子能力强，因此纳米 $Mg(OH)_2$ 被广泛应用于工业废水中吸附重金属离子的应用。Wei-Ming 等[38]设计了一种简便的制备碳点/$Mg(OH)_2$ 复合材料的方法，能够从水中去除和回收潜在有毒的 Cd^{2+}金属。制备的碳点/$Mg(OH)_2$ 具有分层的花瓣阵列结构，并具有较大的比表面积，优化的复合材料达到 1015.4 mg/g 去除能力，循环使用 12 次，去除效率仍高达 98.6%。Xiong 等[39]研究了用一种简单的溶胶－凝胶法制备的氧化镁纳米颗粒从水溶液中去除 Cd^{2+}和 Pb^{2+}的方法。通过分批吸附实验，考察了 Cd^{2+}和 Pb^{2+}的去除效果。用 Langmuir 方程计算出的 Cd^{2+}的最大吸附能力为 2294 mg/g，Pb^{2+}的最大吸附能力为 2614mg/g。

无机阴离子污染物主要包括磷酸盐、砷酸盐、硝酸盐等广泛存在于废水中。硝酸盐、磷酸盐会使水体富营养化，造成赤潮或绿潮等环境危机，严重损害了水生生物的生存环境[40]。纳米 $Mg(OH)_2$ 表面含

有大量羟基活性基团，可以与这些无机阴离子通过表面络合进行交换，能有效使其从水中分离[41]。纳米 $Mg(OH)_2$ 去除水中无机阴离子被大量报道。Mohamed 等[42]研究了在批量体系中使用 $Mg(OH)_2$ 改性膨润土去除水溶液中磷酸盐离子的方法。研究了 pH 效应、吸附剂浓度、平衡时间、吸附等温线和热力学吸附效应。结果表明，该改性工艺对磷酸盐的去除效率有显著影响。初始磷酸离子浓度为 25 mg/L，吸附温度为 45℃，pH 为 7，吸附量为 14.33 mg/m^2。证明了氢氧化镁改性膨润土具有较好的磷酸盐吸附密度和去除率。Huan-Ping 等[43]采用综合沉淀热解法制备了生物炭支撑 $Mg(OH)_2$/膨润土复合材料(PMRB)，用于同时去除废水中的磷酸盐、铵和腐殖酸。结果表明，吸附过程不需要额外的 pH 调整，PMRB 对三种物质的吸附能力分别为磷酸盐 125.36 mg/g，铵 58.20 mg/g，腐殖酸 134.57 mg/g。Noura 等[44]采用分批吸附技术，研究了氟化物在石灰石(LS)和 $Mg(OH)_2$-LS 复合材料上的吸附动力学。$Mg(OH)_2$-LS 复合材料相对于天然 LS，F^-的吸附量显著增加。其作用机理是表面 $Mg(OH)_2$ 的氢氧化物官能团可以通过表面络合(配体交换)进行交换。

4.2. 有机物

随着药品和医疗行业的快速发展，人类和动物对抗生素的消耗有所增加。抗生素在水生环境中的普遍存在会导致耐药细菌的繁殖，严重威胁到人们的健康和抗生素药物的有效性[45]。Lu 等[46]用废弃的亚硫酸氢盐制备了 $Fe_3O_4@Mg(OH)_2$ 核壳磁性纳米颗粒，并作为改善甲硝唑(MNZ)臭氧氧化的催化剂。使用 $Fe_3O_4@Mg(OH)_2$，使 MNZ 的去除速率常数提高了 694.7%。在再利用过程中，观察到 $Fe_3O_4@Mg(OH)_2$ 纳米颗粒具有相对持久性和较高的催化活性。其吸附容量、去除率、回收过程如图 3 所示。Wang 等[47]合成了一种层状氢氧化镁并研究了其对于低浓度环丙沙星(CIP)的吸附性能。结果表明，$Mg(OH)_2$ 对环丙沙星有良好的吸附性能，最大去除率可达 91.1%。吸附机理分析表明，CIP 与纳米 $Mg(OH)_2$ 之间可能存在静电相互作用、配位和氢键。此外，$Mg(OH)_2$ 在低浓度的 CIP 溶液中是稳定的，可回收利用。

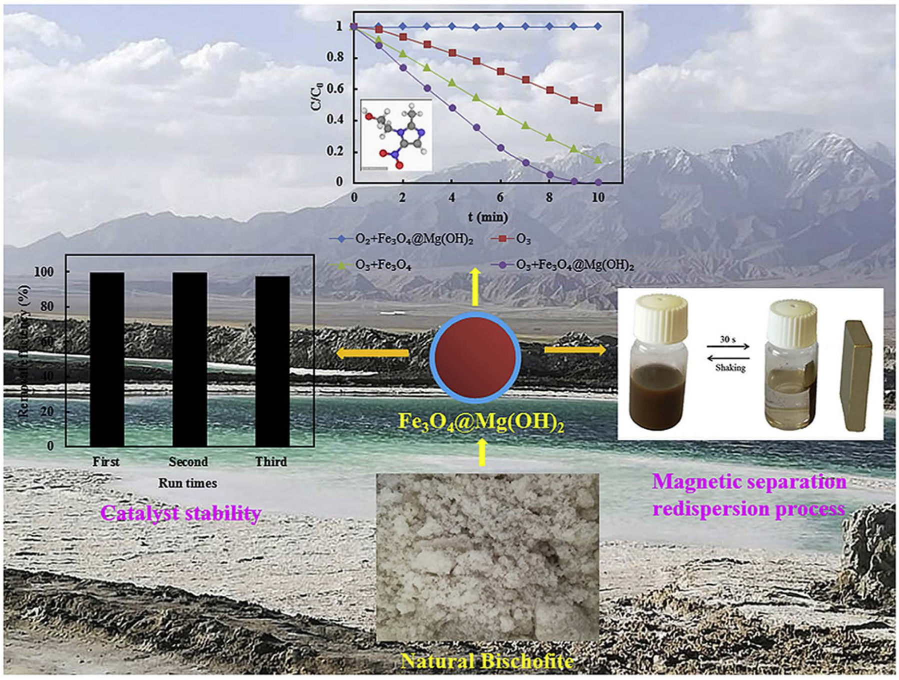

Figure 3. Enhanced ozonation of antibiotics using magnetic $Mg(OH)_2$ nanoparticles made through magnesium recovery from discarded bischofite [46]

图 3. 利用从废弃的亚硫酸氢盐中回收镁制备的磁性 $Mg(OH)_2$ 纳米颗粒增强抗生素的氧化

纺织染色工业中高浓度染料的废水的大量排放，是造成水体环境污染的主要原因之一[48]。这些着色剂成分复杂，生物降解性差、毒性高对生态系统产生巨大的影响，所造成的污染严重阻碍生态循环，并威胁环境的可持续性[49]。Jiang 等[50]采用简单的一步自组装方法制备了新型向日葵球状 $Mg(OH)_2$ 微球颗粒。分析了蒽醌类染料活性蓝 19 (RB19)和茜素红 S (ARS)的合成产物及其吸附机理在优化条件下，ARS 和 RB19 的去除率分别为 91.65%和 83.03%，最大吸附量为 349.85 mg/g，RB19 为 231.78 mg/g。分析吸附机理，RB19 和 ARS 染料通过氢键的形成在向日葵球状 $Mg(OH)_2$ 表面被吸附。Liu 等[51]采用简易水浴法成功合成了分级 $Mg(OH)_2$-MnO_2 纳米复合材料(MMNC)，并进一步应用于刚果红(CR)和甲基橙(MO)的去除。实验结果表明，纳米复合材料对 CR 和 Mo 的最大去除能力分别为 17,100 mg/g 和 7300 mg/g，吸附机理在低浓度时主要是化学吸附，高浓度时吸附、沉淀和光降解产生协同效应提高了去除效率。

4.3. 放射性元素

除了对于常见污染物的去除，纳米 $Mg(OH)_2$ 在去除放射性元素也表现出优异的吸附性能，在放射性废水处理中具有真正的应用潜力。Yan 等[52]采用简单的煅烧法成功合成了分层介孔/大孔 MgO。研究了分层介孔/大孔 MgO 在铀溶液中水解产生的介孔 $Mg(OH)_2$ 对于水溶液 U(VI)的去除。结果表明，介孔 $Mg(OH)_2$ 对于高浓度 U(VI)具有良好的吸附性能，U(VI)的最大吸附能力为 3111 mg/g，在初始铀浓度为 500 mg/L 时，最高的铀去除效率为 99%。吸附等温线符合 Freundlich 模型。在吸附过程中化学吸附是限速步骤。且介孔 $Mg(OH)_2$ 可以通过使用 1M Na_2CO_3 进行再生。Chen 等[52]采用简单的一步法，成功制备了 $Ag_2O@Mg(OH)_2$ 纳米复合材料。系统地研究了 $Ag_2O@Mg(OH)_2$ 纳米复合材料对碘化物(I^-)和 UO_2^{2+} 的去除作用。批量处理实验表明，纳米复合材料可以同时高效去除 I^-和 UO_2^{2+}。且去除能力几乎不受共存阴离子的影响，如 Cl^-，SO_4^{2-}，CO_3^{2-} 和 NO^{3-}。其复合材料形貌，对 I^-和 UO_2^{2+}的去除效率及吸附之后材料的扫描能谱如图 4 所示。

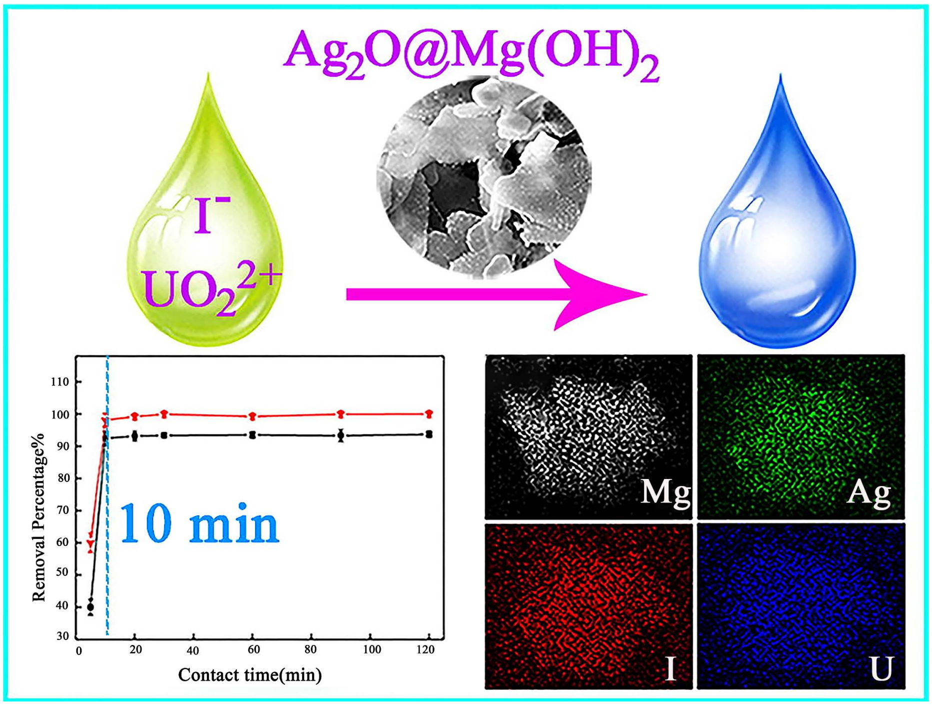

Figure 4. One-step synthesis of $Ag_2O@Mg(OH)_2$ nanocomposite as an efficient scavenger for iodine and uranium [53]
图 4. 一步合成 $Ag_2O@Mg(OH)_2$ 纳米复合材料作为碘和铀的有效吸附剂

5. 总结与展望

综上所述，纳米 $Mg(OH)_2$ 材料制备过程简单，原料廉价易得，目前已经拥有完善的生产制备工艺可以大批量工业化生产。纳米 $Mg(OH)_2$ 材料作为吸附剂在环境污染处理方面被广泛的研究，结果表明纳米 $Mg(OH)_2$ 对酸性气体如 SO_3、CO_2、NO_2 等可以有效吸附去除或转化为其它可利用资源；对于水中污染物重金属离子、无机阴离子、有机物等，纳米 $Mg(OH)_2$ 主要是通过化学吸附、静电吸附或者形成氢键等吸附去除，表现出了高吸附量以及高吸附效率，而且材料可回收循环使用，减少了固废的产生。纳米 $Mg(OH)_2$ 结构稳定，表面活性基团多，通过合理设计对其进行改性为特定应用定制的活性表面官能，可以作为一种新方法使纳米 $Mg(OH)_2$ 被推广应用。纳米 $Mg(OH)_2$ 作为一种环保型材料，在环境处理方面展现出了巨大的应用潜力。

基金项目

感谢重庆自然科学基金(No.cstc2020jcyj-msxmX0875)和重庆科技学院科技创新培训计划(No.YKJCX2020540)为这项工作提供支持。

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