基于磁性纳米流体的中温太阳能吸收与光热转换性能

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摘要 纳米流体基直接式吸收器被认为是进一步提高太阳能热利用效率的重要替代方案, 但针对具有较高能源品位的中温太阳能吸收与光热转化性能的研究却较少. 为促进中温太阳能光热的规模化应用, 本文提出一种较为实用的中温磁性纳米流体基直接式吸收器吸收体系的使用方法. 首先, 通过“两步法”制备一种导热油基Co@NC中温磁性纳米流体, 并利用其优异的光学吸收性能, 在大功率模拟太阳光照射下获得较高温度的流体; 然后, 基于磁分离技术将Co@NC纳米颗粒与导热油基液分离, 获得具有较高温度的纯基液; 再经过换热器热交换过程得到所需要的各种较高温液体, 在5个太阳辐射下, 纳米流体最高温度可达120.37°C. 该方法实现了较高的光热转换效率, 同时可有效解决因纳米流体的不稳定性而引发沉降的问题, 为促进纳米流体在太阳能中温热利用中的新发展提供了一种方案.

关键词 太阳能光热利用, 磁性纳米流体, 磁分离, 光热转换

太阳能作为一种清洁、无污染的可再生能源已被广泛应用于光伏发电^[1]、光热利用^[2]、光化学催化^[3]等领域. 其中, 太阳能光热利用是人们利用太阳能最为简单、直接有效的途径之一. 按照其工作温度范围, 可分为低温热利用(<80°C)、中温热利用(80~250°C)和高温热利用(>250°C)三大领域. 目前, 低温热利用技术已经较为成熟, 主要用于生活热水; 高温热利用主要为太阳能热发电技术, 在国外已有规模化商业应用; 中温热利用技术则发展相对迟缓, 可用于建筑供暖、海水淡化及部分工业用热等, 是解决当前能源短缺、环境污染等问题的有效技术之一. 因此, 突破中温太阳能热利用在工业运用中的技术瓶颈, 有望使其成为未来发展

的一个重要方向^[4,5].

在太阳能热利用中最关键的问题是如何提高太阳能集热器的集热效率. 近年来, 纳米流体基直接式吸收器被认为是进一步提高太阳能热利用效率的重要替代方案. 研究表明, 利用纳米流体直接吸收太阳辐射能有效提高太阳能集热器的集热效率^[6-8]. 然而, 由于纳米颗粒的存在, 纳米流体吸收的能量必须通过热交换器换热过程后才能使用(图1(a))^[9,10]. 为获得较高的传热效率, 研究者开发设计了一系列新型换热器, 在一定程度上提高了太阳能的热利用率^[11,12]. 但纳米流体在长期的冷热交变过程中易于沉降, 将会沉积在换热器的管道壁面上, 甚至引发堵塞, 造成热交换过程中热阻

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图1 (网络版彩色) 纳米流体基直接式吸收器. (a) 传统直接式吸收器使用方法; (b) 提出的假设工作模型

Figure 1 (Color online) Nanofluids-based direct absorber. (a) The traditional working mechanism of direct absorber; (b) the proposed hypothetical working mechanism model

增大和热损失严重^[13]. 此外, 由于纳米流体基液及纳米颗粒的特性限制, 传统的水、乙二醇基纳米流体只能在低温领域得以应用. 为解决以上问题, 本文提出了一种基于磁分离技术的纳米流体光热吸收方案. 该纳米流体具有较高的光热转换率, 能实现太阳光的高效捕获和转换, 获得中温的纳米流体; 在其进入换热器前实现基液与纳米颗粒的完全分离, 获得较高温度的纯基液, 再经过换热器热交换过程得到所需要的各种较高温液体(图1(b)). 同时, 研究了不同光照下纳米流体的光热转化特性. 实验结果表明, 在5个太阳辐射下, 纳米流体最高温度可达120°C.

磁性纳米流体主要由磁性纳米颗粒和基液两部分组成. 作为一种新型功能材料, 磁性纳米流体兼具磁性固体材料的磁特性和液体的流动性, 在航空航天、机械、生物医药、电子、化工、环保^[14,15]等多个领域具有较大的应用潜力.

1 实验部分

(i) 磁性纳米颗粒的制备. 纳米级ZIF-67的制备方法与文献^[16]中的制备方法相似. 7.7 g六水合硝酸钴和110 g 2-甲基咪唑分别溶解到50和400 mL去离子水中. 将两种溶液迅速混合到一起, 并在室温下持续搅拌6 h. 反应后的悬浮液, 经离心得到紫色固体. 将紫色粉末用甲醇反复超声、离心、洗涤3次, 最后将产物置于80°C真空干燥箱中干燥, 得到ZIF-67. 将制备的ZIF-67粉末均匀地分散在陶瓷坩埚中, 放入管式炉, 然后在室温下暴露于氮气流中1 h. 管式炉以10°C/min的升温速度升至目标温度900°C, 保持恒温2 h后冷却至室温.

(ii) 磁性光热纳米流体的制备. 磁性光热纳米流体采用“两步法”制备. 首先将制备好的Co@NC磁性纳米颗粒按照一定的比例添加到导热油(Oil)中; 然后通过超声作用, 使Co@NC磁性纳米颗粒均匀稳定地分散在导热油中形成Co@NC-Oil磁性光热纳米流体.

(iii) 表征方法. 用场发射扫描电子显微镜(scanning electron microscope, SEM)和透射电子显微镜(transmission electron microscope, TEM/HRTEM)表征Co@NC磁性纳米颗粒形貌. 磁性纳米颗粒的晶体结构利用粉末X射线衍射仪(X-ray diffractometer, XRD)测定. 样品的磁特性由振动样品磁强计(vibrating sample magnetometer, VSM)在室温下表征. 采用紫外可见近红外分光光度计(ultraviolet-visible-near infrared spectrophotometer, UV-Vis NIR)测量Co@NC纳米颗粒的光学性质.

2 结果与讨论

2.1 Co@NC磁性纳米颗粒的形貌结构

从图2(a), (b)可以看出, Co@NC前驱体ZIF-67呈现出标准的十二棱面体形状; 碳化后得到的Co@NC在一定程度上保持了模板ZIF-67的形貌, 但颗粒尺寸变小, 表面变得更加粗糙, 同时金属钴粒子热还原成具有铁磁性的钴单质颗粒. 从图2(c)可以看出, 金属颗粒(Co)被碳素结构均匀包覆. 从图2(d)可以看出, 钴纳米颗粒只呈现出一种晶格条纹, 间距约为0.20 nm, 对应于fcc构型单质钴的(111)晶面. 以上结果说明, 所有Co@NC纳米颗粒都具有磁性, 从而保证纳米颗粒可以通过磁分离技术完全分离.

图2 (网络版彩色)Co@NC形貌图。(a) 前驱体ZIF-67晶体SEM图；(b) Co@NC纳米颗粒SEM图；(c) Co@NC纳米颗粒TEM图；(d) Co@NC纳米颗粒HRTEM图

Figure 2 (Color online) Co@NC morphology. (a) SEM image of as-prepared ZIF-67 crystals; (b) SEM image of Co@NC nanoparticles; (c) TEM image of Co@NC nanoparticles; (d) HRTEM image of Co@NC nanoparticles

2.2 Co@NC磁性纳米颗粒的晶体结构

如图3(a)所示, 制备的Co@NC前驱体ZIF-67晶体特征衍射峰与PDF标准卡所对应的峰位完全吻合, 表明Co@NC具有较高的纯度, 几乎没有杂质。从图3(b)可以看出, 经过高温碳化后, 所有的ZIF-67样品都经历了结构转变, 并表现出相似的XRD峰位, 仅峰强度有所差异。在 $2\theta=26.5^\circ$ 时出现一个较宽的XRD峰, 表明ZIF-67前驱体经碳化后形成了碳素结构。其他3组峰($2\theta=44.28^\circ$, 51.63° 和 75.97°)分别归属于金属钴(111), (200)和(220)晶面。这些结果表明, 在碳化过程中钴离子脱离ZIF-67骨架并被热还原成具有铁磁性质的钴单质颗粒。

2.3 Co@NC磁性纳米颗粒的磁性能

图4所示为Co@NC在室温下的磁性能。合成的Co@NC纳米颗粒表现出典型的滞后环, 说明它们具有铁磁性特征。其饱和磁值达到53.9 emu/g, 具有较高的磁性, 可以保证纳米颗粒通过磁分离技术完全分离。

2.4 磁性纳米流体的稳定性

由图5(a)可以看出, 随着浓度增加, 磁性纳米流体的颜色越来越深, 表明其吸光能力越来越强, 与透射光谱数据一致。静置2周后, 发现无明显的沉淀, 说明制备的Co@NC磁性纳米流体具有良好的稳定性。

图3 (网络版彩色)XRD图。(a) 前驱体ZIF-67; (b) Co@NC纳米颗粒

Figure 3 (Color online) XRD patterns of as-prepared ZIF-67 (a) and Co@NC nanoparticles (b)

图4 (网络版彩色)Co@NC纳米颗粒的磁化-磁滞曲线. $1 \text{ emu/g} = 1 \text{ (A m}^2\text{)/kg}$, $1 \text{ Oe} = 80 \text{ A/m}$

Figure 4 (Color online) Magnetization hysteresis curves of Co@NC nanoparticles. $1 \text{ emu/g} = 1 \text{ (A m}^2\text{)/kg}$, $1 \text{ Oe} = 80 \text{ A/m}$

2.5 磁性纳米流体的光学性能

图6为纳米流体的透射光谱图及消光系数图. 由图6(a)可知, 纳米流体的透过率远低于基液Oil的透过率, 说明添加少量的Co@NC纳米颗粒可以大幅度地提高流体的光吸收性能. 为进一步研究其光学性质, 纳米流体的消光系数由式(1)得出^[17~19]:

$$T(\lambda) = \exp(-K_{el}d). \quad (1)$$

其中, K_{el} 为纳米流体的消光系数; $T(\lambda)$ 为纳米流体的透过率; d 为光路长度, 取 1.0 cm .

由图6(b)可知, 纳米流体随浓度增加, 吸收太阳光的能力逐渐增强, 与透射光谱结果一致.

2.6 磁性纳米流体光热转化性能评价

本文通过闷晒实验对不同浓度纳米流体在不同光

图5 磁性纳米流体照片. (a) 分散后; (b) 放置2周后. $1 \text{ ppm} = 0.001\%$
Figure 5 Photographs of magnetic nanofluids. (a) Fresh prepared; (b) after being stored for 2 weeks. $1 \text{ ppm} = 0.001\%$

强下的光热转化性能进行评价, 实验装置如图7所示. 采用CEL-PE300-3A均匀光氙灯(北京中教金源科技有限公司)作为光源模拟太阳光照, 配备AM1.5光学滤波器. 用辐照仪(NOVA II博纳德精密仪器有限公司)测量平均入射热通量, 辐射热通量测量误差小于 $\pm 4\%$. 采用石英比色皿(辐照面积 4 cm^2)盛装纳米流体, 用导热系数较低的绝热气凝胶保温以减少热损. 此外, 太阳光模拟器到纳米流体接受光照的上表面距离为 20 cm , 相应的辐射热通量入射量范围为 $1000 \sim 5000 \text{ W/m}^2$. K型热电偶探头放置在接收器中心, 使用数据采集系统(34970A, 安捷伦)记录温度, 温度测量精度在 $\pm 0.1^\circ\text{C}$ 范围内, 每 2 s 记录一次温度. 所有样品在相同条件下进行测量. 其光热转化效率由式(2)计算^[20]:

$$\eta = \frac{(T_{eq} - T_{am})m_w c_m B}{AS_m}. \quad (2)$$

图6 (网络版彩色)不同浓度Co@NC纳米流体的透射光谱图(a)和消光系数图(b)

Figure 6 (Color online) UV-Vis-NIR spectra (a) and extinction coefficient (b) of Co@NC nanofluids with different concentrations

图7 (网络版彩色)光热转换测试系统示意图

Figure 7 (Color online) Diagram of the photothermal conversion experimental system

其中, η 为光热转化效率; T_{eq} 为平衡时的温度($^{\circ}\text{C}$); T_{am} 为环境温度($^{\circ}\text{C}$); m_w 为纳米流体的质量(kg); c_m 为纳米流体的比热容($\text{J}/(\text{kg K})$); B 为恒定散热率($1/\text{s}$); A 为光辐照面积(m^2); S_m 为辐照强度(W/m^2).

为评价其最大吸光能力, 将整个测试过程分为三个阶段: 升温过程、平衡过程、降温过程. 在试验开始阶段, 太阳模拟器刚刚开启, 纳米流体就开始吸收太阳光, 并将其转化为热能. 由于液体与环境的温差不明显, 热损失率低, 液体温度迅速升高, 称该阶段为升温过程. 由图8(a)~(e)可以看出, 在升温阶段随着纳米流体浓度升高, 温度上升速度也随之增加. 相较于基液导热油, 纳米流体温升速率明显增加. 结果表明, Co@NC纳米颗粒能大幅提高流体的光吸收及转化性能. 随着温差增大和损失增强, 温度上升速度逐渐减小, 最后趋于平缓, 该阶段被称为平衡阶段. 此时, 可以简单地理解为热损失量等于热产生量. 由图8(a)~(e)可以看出, 不同浓度纳米流体达到平衡时的温度关系为 $100 \text{ ppm} > 75 \text{ ppm} > 50 \text{ ppm} > 25 \text{ ppm} > \text{Oil}$. 随着光照强度增强, 达到平衡时的温度也逐渐增高. 当光照强度为 $5000 \text{ W}/\text{m}^2$, 浓度为 100 ppm 时, 最高温度可达到 120.37°C , 相较于基液导热油最高温度(90.7°C)提高了 29.67°C . 待温度达到平衡后, 关闭太阳光模拟器, 整个测试体系开始降温, 该阶段被称为降温阶段. 其中降温曲线可由式(3)进行描述^[15].

$$\frac{d(T(t) - T_a)}{dt} = -B(T(t) - T_a). \quad (3)$$

其中, t 为关闭太阳光模拟器的时间; $T(t)$ 为关闭太阳光模拟器后 t 时对应的瞬时温度; T_a 为环境温度.

对式(3)进行积分, 得

$$\ln(T(t) - T_a) = -Bt + C. \quad (4)$$

由太阳光模拟器 $t=0$ 时 $T(t)=T_{eq}$, 可知恒定散热率(如

图8(f)所示)

$$B = -\frac{\ln\left(\frac{T(t) - T_a}{T_{eq} - T_a}\right)}{t}. \quad (5)$$

光热转化效率作为衡量纳米流体光热性能的重要指标可以由式(2)求得. 从图9可以看出, 在同一光强下随纳米流体浓度增加, 其光热转化效率相应增加. 当光照强度为 $1000 \text{ W}/\text{m}^2$, 浓度为 100 ppm 时, 光热转化效率最大, 为 65.23% . 但在同一浓度下随光照强度增强, 虽然所得最终纳米流体的温度增加, 但光热转化效率却明显降低. 当光照强度为 $5000 \text{ W}/\text{m}^2$, 浓度为 100 ppm 时, 得到的最大光热转化效率仅为 33.84% . 造成这一现象的主要原因是, 随着光照强度增加, 纳米流体中纳米颗粒吸光能力很快达到饱和, 导致部分太阳光无法被吸收. 此外, 光照越强, 则穿透能力越强. 试验中使用 1 cm 厚比色皿, 且纳米流体最大浓度为 100 ppm , 相对较低, 部分光线透过整个纳米流体层, 无法被完全吸收, 在宏观层面上表现为光热转化效率随光照强度增加而降低.

2.7 不确定度分析

在本实验中, 磁性纳米流体的消光系数 K_{el} 按式(1)计算. 由该式可知, 消光系数 K_{el} 是透光率 T 和光路 d 的函数. 在测量过程中, 波长的测量误差为 $\pm 0.02 \text{ nm}$, 杂散光 $\leq 0.00007\%$. 消光系数不确定度可简化为

$$\frac{U_{K_{el}}}{K_{el}} = \left[\left(\frac{U_d}{d} \right)^2 + \left(\frac{U_{T(\lambda)}}{T(\lambda)} \right)^2 + \left(\frac{U_\lambda}{\lambda} \right)^2 \right]^{0.5}. \quad (6)$$

其中, $\frac{U_d}{d} = 0.1\%$, $\frac{U_{T(\lambda)}}{T(\lambda)} \leq 0.00007\%$, $\frac{U_\lambda}{\lambda} = 0.0071\%$,

$$\frac{U_{K_{el}}}{K_{el}} = 0.1003\%.$$

由光热转化效率公式可知, 纳米流体的光热转换性能 η 是流体质量 m 、比热 C_p 、温度增量 $(T_s - T_i)$ 、辐照面积 A 、光强度 G 和恒定散热率 B 的函数. 其中, 恒定散热率 B 由辐照时间 t 决定; A 由上表面边长 d 决定; m , C_p , t (温度), d , G , t (时间)的测量误差分别为 $\pm 0.01 \text{ g}$, $\pm 1\%$, $\pm 0.1^{\circ}\text{C}$, $\pm 0.1 \text{ mm}$, $\pm 4\%$ 和 $\pm 0.1 \text{ s}$. 因此, 光热转化效率不确定度为

$$\frac{U_\eta}{\eta} = \left[\left(\frac{U_m}{m} \right)^2 + \left(\frac{U_{C_p}}{C_p} \right)^2 + \left(\frac{U_{T_s - T_i}}{T_s - T_i} \right)^2 + \left(\frac{U_G}{G} \right)^2 + \left(\frac{U_N}{\Delta t} \right)^2 + \left(\frac{U_A}{A} \right)^2 \right]^{0.5}. \quad (7)$$

图8 (网络版彩色)不同光强下Co@NC纳米流体升温曲线。(a) 1000 W/m²; (b) 2000 W/m²; (c) 3000 W/m²; (d) 4000 W/m²; (e) 5000 W/m²; (f) 在太阳光模拟器关闭后绘制的 $\ln \frac{(T(t)-T_a)}{(T_{eq}-T_a)}$ 与 t 的关系曲线

Figure 8 (Color online) Temperature rises of Co@NC nanofluids with different irradiation intensity. (a) 1000 W/m²; (b) 2000 W/m²; (c) 3000 W/m²; (d) 4000 W/m²; (e) 5000 W/m²; (f) the relationship curve between $\ln \frac{(T(t)-T_a)}{(T_{eq}-T_a)}$ and t after the solar simulator was shut down

$$\text{其中, } \frac{U_m}{m} = \frac{0.01}{100} = 0.01\%, \quad \frac{U_{c_p}}{c_p} \leq 1\%, \quad \frac{U_{T_s-T_i}}{T_s-T_i} = \frac{0.1}{95} \\ = 0.105\%, \quad \frac{U_G}{G} = \frac{0.04}{5000} = 0.0008\%, \quad \frac{U_{\Delta t}}{\Delta t} = \frac{1}{5000} = 0.02\%, \\ \frac{U_A}{A} = \left(2 \left(\frac{U_d}{d} \right)^2 \right)^{0.5} = \left(2 \left(\frac{0.1}{100} \right)^2 \right)^{0.5} = 0.14\%, \quad \frac{U_\eta}{\eta} = 2.69\%.$$

3 结论

为获得较高品位的能量, 同时有效避免换热器堵塞问题, 进一步促进太阳能中温热利用大规模工业化应用, 本文提出一种较为实用的中温磁性纳米流体集热系统, 并探究不同光强对纳米流体光热性能的影响。

主要结论如下:

(1) 选用ZIF-67为前驱体, 经高温碳化制备出一种具有高光热转换效率的Co@NC磁性纳米颗粒. 磁性金属钴单质被碳素结构均匀包覆, 保证磁性纳米颗粒可以通过磁分离技术与纳米流体基液完全分离, 为解决换热器堵塞问题提供了一个较好的方案.

(2) 添加磁性纳米颗粒后, 磁性光热纳米流体的光热转化效率明显高于基液. 当光照强度为 5000 W/m^2 , 浓度为 100 ppm 时, 最高温度可达到 120.37°C , 获得较高品位的能量.

(3) 该研究为进一步提升中温光热纳米流体的工业化应用水平提供了一个可行性方案.

图9 (网络版彩色)不同浓度Co@NC纳米流体的光热转化效率
Figure 9 (Color online) Photothermal conversion efficiencies of Co@NC nanofluids with different concentrations

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Summary for “基于磁性纳米流体的中温太阳能吸收与光热转换性能”

Properties of solar energy absorption and photothermal conversion at medium temperature based on magnetic nanofluids

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As a kind of clean and renewable energy, solar energy has been widely used in photovoltaic power generation, photothermal utilization, photocatalysis and other fields. According to its working temperature range, photothermal utilization can be divided into three areas: Low-temperature heat utilization (<80°C), medium-temperature heat utilization (80–250°C) and high-temperature heat utilization (>250°C). At present, the technologies of low-temperature heat utilization and high-temperature heat utilization have been relatively mature, while the development of medium-temperature heat utilization technology is relatively slow. Furthermore, the medium-temperature heat utilization is one of the effective technologies to solve the current energy shortage, environmental pollution and other problems. The key problem in solar energy heat utilization is how to improve the efficiency of solar collector. Nanofluids-based direct absorption of solar radiation energy can effectively improve the collection efficiency of solar collectors. However, nanofluids are easy to settle in the long-term cold-hot alternating process, which will deposit on the pipe wall of heat exchanger and even cause blockage, resulting in the increase of thermal resistance and serious heat loss in the process of heat exchange.

In this paper, a way of magnetic nanofluid-based direct absorption system for medium-temperature heat utilization is proposed. Firstly, a heat-conducting oil-based nanofluid with magnetic Co@NC is prepared by “two-step” method, and high temperature fluids are obtained under high-power simulated solar irradiation by its excellent optical absorption performance. Secondly, based on magnetic separation technology, Co@NC nanoparticles are separated from the heat conducting oil matrix to obtain pure base liquids with high temperature. Under five solar radiations, the highest temperature of the nanofluids could reach to 120.37°C. Moreover, various liquids with higher temperature can be obtained by the process of heat exchange. Finally, the effects of illumination on the photothermal conversion properties of nanofluids are studied, and the mechanism is explained. This method achieves high photothermal conversion efficiency, and can effectively solve the problem of sedimentation caused by the instability of nanofluids. In addition, it provides a scheme for promoting new development of nanofluids in the use of solar energy at medium temperature.

The results show that the photothermal conversion efficiency of magnetic nanofluids is significantly higher than that of base fluids. When the illumination intensity is 5000 W/m² and the concentration is 100 ppm, the highest temperature can reach to 120.37°C, which is higher than that of pure thermal conductive oil (90.7°C). In addition, the highest photothermal conversion efficiency is 65.23% when the illumination intensity is 1000 W/m² and the concentration is 100 ppm. With the increase of illumination intensity at the same concentration, the final temperature of the nanofluids increases, but the photothermal conversion efficiency decreases significantly. When the illumination intensity is 5000 W/m² and the concentration is 100 ppm, the highest photothermal conversion efficiency is only 33.84%. The main reason for this phenomenon is that part of the sunlight can't be absorbed. With the increase of light intensity, the absorptivity of nanoparticles in nanofluids quickly reaches saturation. Moreover, the stronger the illumination is, the stronger the penetration ability is. The colorimetric dish with a thickness of 1 cm is used, and the maximum concentration of nanofluids is 100 ppm, which is relatively low. Some light passes through the whole nanofluidic layer and can't be fully absorbed. Therefore, the results show that the photothermal conversion efficiency decreases with the increase of light intensity.

solar energy photothermal utilization, magnetic nanofluids, magnetic separation, photothermal conversion

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