Notice

DESCRIPTION CN120553696A

A method for preparing graphene composite materials based on cerium dioxide catalytic modification.

基于二氧化铈催化改性制备石墨烯复合材料的方法

[0001]

Technical Field

技术领域

[n0001]

This invention belongs to the field of graphene preparation technology, specifically relating to a method for preparing graphene composite materials based on cerium dioxide catalytic modification.

本发明属于石墨烯制备技术领域，具体涉及基于二氧化铈催化改性制备石墨烯复合材料的方法。

[0003]

Background Technology

背景技术

[n0002]

In today's era of rapid technological development, graphene, with its unique two-dimensional structure and outstanding physicochemical properties, such as ultra-high electron mobility, excellent thermal conductivity and superior mechanical strength, has shown unparalleled application potential in many cutting-edge fields such as electronics, energy storage, and optoelectronics.

在当今科技飞速发展的时代，石墨烯以其独特的二维结构和卓越的物理化学性能，如超高的电子迁移率、出色的热导率以及优异的机械强度等，在电子学、储能、光电子学等众多前沿领域展现出了无可比拟的应用潜力。

[n0003]

Chemical vapor deposition (CVD) has become one of the most promising preparation methods due to its ability to achieve large-area, high-quality graphene growth.

化学气相沉积（CVD）法凭借能够实现大面积、高质量石墨烯生长的优势，成为目前最具前景的制备方法之一。

In the process of preparing graphene by chemical vapor deposition (CVD), copper has become one of the most commonly used substrate materials due to its low carbon solubility and excellent catalytic performance for the growth of monolayer graphene.

在化学气相沉积（CVD）法制备石墨烯的过程中，铜因其低碳溶解度以及对单层石墨烯生长优异的催化性能，成为最常用的衬底材料之一。

Copper can not only effectively catalyze the decomposition of carbon sources, but also promote the nucleation and growth of graphene through its surface properties.

铜不仅能够有效催化碳源分解，还能通过其表面特性促进石墨烯的成核和生长。

However, existing copper-based CVD processes still face many challenges: the uneven distribution of active sites on the copper substrate surface leads to slow and uneven graphene growth, numerous defects, unstable graphene quality, and low thermal conductivity and high resistivity of graphene, making it difficult to meet the requirements of high-power electronic devices.

然而，现有的基于铜基底的CVD工艺仍面临诸多挑战：铜基底表面活性位点分布不均，导致石墨烯生长缓慢且不均匀，缺陷较多，石墨烯质量不稳定，且石墨烯的导热性低，电阻率高，难以满足高功率电子器件的需求。

[0006]

Summary of the Invention

发明内容

[n0004]

In view of the above-mentioned shortcomings of the existing technology, the purpose of this invention is to provide a method for preparing graphene composite materials by cerium

dioxide catalytic modification. This invention can effectively increase the number of uniformly distributed active sites on the surface of copper substrate, increase the growth rate of graphene, and ensure the uniformity of graphene growth. At the same time, it can also improve the thermal conductivity of graphene and reduce resistivity.

针对现有技术存在的上述不足，本发明的目的就在于提供二氧化铈催化改性制备石墨烯复合材料的方法，本发明能有效提高铜基底表面均匀分布的活性位点数量，提高石墨烯的生长速率，且保证石墨烯生长的均匀性，同时还能提高石墨烯的导热性能，降低电阻率。

[n0005]

The technical solution of this invention is implemented as follows:

本发明的技术方案是这样实现的：

[0009]

The method for preparing graphene composite materials based on cerium dioxide catalytic modification specifically includes the following steps:

基于二氧化铈催化改性制备石墨烯复合材料的方法，具体包括以下步骤：

[0010]

S1: Pretreatment of the copper substrate;

S1：对铜基底进行预处理；

[0011]

S2: Add cerium dioxide nanoparticles and dispersant to deionized water, then ultrasonically disperse them evenly to obtain a cerium dioxide suspension;

S2：将二氧化铈纳米颗粒和分散剂加入去离子水中，然后超声分散均匀，得到二氧化铈悬浮液；

[0012]

S3: Immerse the pretreated copper substrate from step S1 into a cerium dioxide suspension, then lift the copper substrate to form a cerium dioxide coating on the surface of the copper substrate, and then let it stand to dry for later use.

S3：将步骤S1中预处理后的铜基底浸入二氧化铈悬浮液中，然后提拉铜基底以在铜基底表面形成二氧化铈涂层，再静置干燥，备用；

[0013]

S4: Place the copper substrate with cerium dioxide coating on its surface from step S3 into the reaction chamber, then evacuate it, and then heat it to 800~1060°C. At the same time, introduce carbon source, hydrogen and protective gas into the reaction chamber and keep it at this temperature for 10-30 min to obtain graphene composite material on the surface of the copper substrate.

S4：将步骤S3表面具有二氧化铈涂层的铜基底置于反应腔室中，然后抽真空，再升温至800~1060°C，同时向反应腔室内通入碳源、氢气和保护气体，保温10-30 min，从而在铜基底表面制得石墨烯复合材料。

[n0006]

Furthermore, in step S1 pretreatment, the copper substrate is first immersed in an acidic solution for acid washing, then washed with deionized water and ethanol solution in sequence, and finally dried in an oven at 60-80°C.

进一步地，步骤S1预处理时，先将铜基底浸入酸性溶液中酸洗，再依次用去离子水和乙醇溶液清洗，最后置于60-80°C烘箱中干燥即可。

[n0007]

Furthermore, the concentration of cerium dioxide in the cerium dioxide suspension is 0.1-1 mol/L.

进一步地，所述二氧化铈悬浮液中二氧化铈浓度为0.1-1 mol/L。

[n0008]

Furthermore, the dispersant is 0.2%-1% wt of polyvinylpyrrolidone.

进一步地，分散剂为0.2%-1% wt的聚乙烯吡咯烷酮。

The polyvinylpyrrolidone used here can effectively disperse cerium dioxide, and has good film-forming and adhesive properties, few heteroatoms, and can decompose at high temperatures to serve as a solid carbon source.

这里采用的聚乙烯吡咯烷酮能有效分散二氧化铈，且成膜性和粘附性好，杂原子少，高温下可分解作为固态碳源。

[n0009]

Furthermore, in step S3, the thickness of the cerium dioxide coating is 50 nm to 500 μm.

进一步地，步骤S3中，二氧化铈涂层的厚度为50 nm~500μm。

Studies have shown that when the cerium dioxide coating is controlled within this thickness range, cerium dioxide can fully contact the copper substrate, which is beneficial for the growth of graphene.

研究表明当二氧化铈涂层控制在这个厚度范围时，二氧化铈能与铜基底完全接触，从而利于石墨烯的生长。

[n0010]

Further, in step S4, the carbon source is one or more of a solid carbon source and a gaseous carbon source. The solid carbon source is one or more of polymethyl methacrylate, polyvinylpyrrolidone, polyvinyl alcohol, polyethylene, hexamethylbenzene, biphenyl, diphenylmethane, triphenylmethane, naphthalene, tetrahydronaphthalene, sugars, activated carbon, amorphous carbon, and acetylene black. The gaseous carbon source is one or more of methane, acetylene, ethylene, ethane, carbon monoxide, and carbon dioxide.

进一步地，步骤S4中，碳源为固态碳源和气态碳源中的一种或多种，所述固态碳源为聚甲基丙烯酸甲酯、聚乙烯吡咯烷酮、聚乙烯醇、聚乙烯、六甲基苯、联苯、二苯甲烷、三苯甲烷、萘、四氢化萘、糖类、活性炭、无定形碳和乙炔黑中的一种或多种；所述气态碳源为甲烷、乙炔、乙烯、乙烷、一氧化碳和二氧化碳种的一种或多种。

[n0011]

Furthermore, when the carbon source is a gaseous carbon source, the flow rate ratio of the carbon source, hydrogen, and protective gas is 8:1:20.

进一步地，所述碳源为气态碳源时，碳源、氢气和保护气体的流量比为8:1:20。

[n0012]

Furthermore, the protective gas is nitrogen or an inert gas.

进一步地，所述保护气体为氮气或惰性气体。

[n0013]

Furthermore, when the carbon source is a solid carbon source, the solid carbon source is uniformly distributed on the surface of the copper substrate, and the thickness of the solid carbon source is 50nm~500μm.

进一步地，所述碳源为固态碳源时，所述固态碳源均匀分布在铜基体表面，且固态碳源的厚度为50nm~500μm。

Studies have shown that the thickness of the solid carbon source should not be too thick or too thin, otherwise it will not be conducive to the uniform growth of graphene.

研究表明固态碳源的厚度不能太厚或者太薄，否则不利于石墨烯的均匀生长。

In practical applications, spin coating is used to control the thickness of the solid carbon source, or a thickness scraper is used to coat different thicknesses.

在实际应用中用旋转涂覆调控固态碳源的厚度或者用厚度刮板来刮涂不同的厚度。

[n0014]

Compared with the prior art, the present invention has the following beneficial effects:

与现有技术相比，本发明具有如下有益效果：

[0023]

1. The present invention forms a cerium dioxide coating on the surface of a copper substrate. The unique crystal structure of cerium dioxide gives its surface abundant active sites.

1、本发明在铜基底表面形成二氧化铈涂层，二氧化铈独特的晶体结构使其表面存在丰富的活性位点。

In the fluorite unit cell structure, cation defects are located in symmetrical cubic lattice sites, and oxygen vacancy defects exist. These defect sites become the active centers of the reaction.

在萤石晶胞结构中，阳离子缺陷位于对称的立方格位，且存在氧空位缺陷，这些缺陷位置成为了反应的活性中心。

When a carbon source gas (such as methane) is introduced into the reaction system, methane molecules can be adsorbed on these active sites. Cerium dioxide can promote the activation of carbon-hydrogen bonds by providing additional active sites, making it easier for methane to decompose and produce carbon atoms, thereby increasing the growth rate of graphene and growing uniformly distributed graphene on the copper substrate surface.

当碳源气体（如甲烷）通入反应体系后，甲烷分子能够吸附在这些活性位点上，二氧化铈可通过提供额外的活性位点促进碳氢键的活化，使甲烷更易分解产生碳原子，从而能提高石墨烯的生长速率，在铜基底表面生长得到均匀分布的石墨烯。

[n0015]

2. The graphene composite material obtained on the copper substrate surface in this invention contains cerium dioxide. Cerium dioxide can promote the growth of graphene during the graphene growth process. At the same time, the presence of cerium dioxide nanoparticles is conducive to the heat transfer and diffusion within the composite material structure, thereby improving the thermal conductivity. Graphene has high electrical conductivity, and cerium dioxide can promote electron transport within the composite material, thereby reducing the resistivity of the composite material.

2、本发明在铜基底表面得到的石墨烯复合材料中含有二氧化铈，二氧化铈在石墨烯生长过程中能促进石墨烯生长，同时二氧化铈纳米颗粒的存在有利于复合材料内部结构热量的传递和扩散，从而提高导热性能；石墨烯具有高导电性，二氧化铈能促进复合材料内部的电子传输，从而能降低复合材料的电阻率。

[n0016]

3. This invention prepares graphene composite materials on the surface of a copper substrate, which has broad application prospects in the fields of cables, generators, motors, and batteries, and provides strong technical support for the large-scale application of graphene in high-tech industries.

3、本发明在铜基底表面制备石墨烯复合材料，在电缆、发电机、电动机、电池等领域具有广泛的应用前景，为石墨烯在高科技产业中的大规模应用提供了强有力的技术支持。

[0026]

Attached Figure Description

附图说明

[n0017]

Figure 1 - SEM images of the copper foil surface after pretreatment and after coating with cerium dioxide in Example 1.

图1-实施例1预处理后和涂覆二氧化铈后铜箔表面的SEM图。

[n0018]

Figure 2 - Optical microscope image of the graphene composite material on the surface of copper foil after coating with cerium dioxide in Example 1.

图2-实施例1涂覆二氧化铈后铜箔表面石墨烯复合材料的光学显微镜图。

[n0019]

Figure 3 - XRD pattern of graphene composite material on copper foil surface after cerium dioxide coating in Example 1.

图3-实施例1涂覆二氧化铈后铜箔表面石墨烯复合材料的XRD图。

[n0020]

Figure 4 - Measurement of carbon yield and hydrogen production rate in Example 1 and Comparative Example 1.

图4-测量实施例1和对比例1的碳收率和氢气生产率。

[n0021]

Figure 5 - Measurement of thermal conductivity and resistivity of copper foil, graphene composite material prepared in Example 1, and graphene material prepared in Comparative Example 1.

图5-测量铜箔、实施例1制得的石墨烯复合材料和对比例1制得石墨烯材料的热导率和电阻率。

[0032]

Detailed Implementation

具体实施方式

[n0022]

The present invention will now be described in further detail with reference to the accompanying drawings and specific embodiments.

下面结合附图和具体实施方式对本发明作进一步详细说明。

[n0023]

Example 1

实施例1

[0035]

The method for preparing graphene composite materials based on cerium dioxide catalytic modification specifically includes the following steps:

基于二氧化铈催化改性制备石墨烯复合材料的方法，具体包括以下步骤：

[0036]

(1) Pretreatment of copper foil with a thickness of about 0.1 mm: Immerse the copper foil in 6M HCl solution and clean for 10 minutes to remove oxides and impurities on the surface.

（1）对厚度约为0.1 mm的铜箔进行预处理：将铜箔浸入6M的HCl溶液中，清洗10分钟，以去除表面的氧化物和杂质。

The pickled copper foil was rinsed with deionized water for 10 minutes to remove residual acid, and then immersed in an ethanol solution for 10 minutes to further remove organic impurities.

将酸洗后的铜箔用去离子水清洗10分钟，以去除残留的酸液，再将铜箔浸入乙醇溶液中清洗10分钟，进一步去除有机杂质。

Place the cleaned copper foil in a 60°C oven overnight to dry, ensuring the surface is completely dry.

将清洗后的铜箔置于60°C的烘箱中干燥过夜，确保表面完全干燥。

[n0024]

(2) Preparation of CeO₂ suspension: CeO₂ nanoparticles and 0.2% wt of polyvinylpyrrolidone (PVP) were added to 100 ml of deionized water and sonicated for 10 minutes to uniformly disperse CeO₂ nanoparticles, resulting in a CeO₂ suspension with a concentration of 0.5 mol/L.

（2）CeO₂悬浮液的制备：将CeO₂纳米颗粒和0.2% wt的聚乙烯吡咯烷酮（PVP）加入100 ml去离子水中，超声处理10分钟，使CeO₂纳米颗粒均匀分散，得到浓度为0.5mol/L的CeO₂悬浮液。

[n0025]

(3) Immerse the pretreated copper foil completely in CeO₂ suspension, slowly lift the copper foil to form a uniform CeO₂ coating (thickness of 300 nm) on its surface, and then let it stand to dry.

（3）将预处理后的铜箔完全浸入CeO₂悬浮液中，缓慢提拉铜箔，使其表面形成均匀的CeO₂涂层（厚度为300 nm），然后静置干燥。

[n0026]

(4) Place the copper foil coated with CeO₂ into the chemical vapor deposition (CVD) reaction chamber.

（4）将涂覆有CeO₂涂层的铜箔放入化学气相沉积（CVD）反应腔。

Evacuate the chamber and clean it with high-purity argon (Ar) 2-3 times to ensure that there is no residual gas in the chamber.

抽真空并用高纯氩气（Ar）清洗反应腔2-3次，确保反应腔内无残余气。

The reaction chamber was then heated to 1000°C at a heating rate of 10°C/min and kept at a constant temperature of 1000°C. Hydrogen (H₂) and methane (CH₄) were introduced and the flow rate was controlled at 500 sccm. The flow ratio of Ar:CH₄:H₂ was 20:8:1.

再以10°C/min 的升温速度将反应腔加热至1000°C，保持 1000°C 恒温，通入氢气（H₂）与甲烷（CH₄），控制流速在500 sccm，Ar：CH₄：H₂的流量比为20：8：1。

The sample was grown at 1000°C for about 10 minutes, then the methane gas was turned off, and the sample was cooled to room temperature at a natural cooling rate, thus obtaining a graphene composite material on the surface of copper foil.

在1000℃下生长10分钟左右，关闭甲烷气体，以自然冷却速率使样品温度冷却至室温，从而在铜箔表面制得石墨烯复合材料。

[n0027]

Comparative Example 1

对比例1

[0041]

The method for preparing graphene materials on the surface of copper foil specifically includes the following steps:

在铜箔表面制备石墨烯材料的方法，具体包括以下步骤：

[0042]

(1) Pretreatment of copper foil with a thickness of about 0.1 mm: Immerse the copper foil in 6M HCl solution and clean for 10 minutes to remove oxides and impurities on the surface.

（1）对厚度约为0.1 mm的铜箔进行预处理：将铜箔浸入6M的HCl溶液中，清洗10分钟，以去除表面的氧化物和杂质。

The pickled copper foil was rinsed with deionized water for 10 minutes to remove residual acid, and then immersed in ethanol solution for 10 minutes to further remove organic impurities.

将酸洗后的铜箔依次用去离子水清洗10分钟，以去除残留的酸液，将铜箔浸入乙醇溶液中清洗10分钟，进一步去除有机杂质。

Place the cleaned copper foil in a 60°C oven overnight to dry, ensuring the surface is completely dry.

将清洗后的铜箔置于60°C的烘箱中干燥过夜，确保表面完全干燥。

[n0028]

(2) Place the pretreated copper foil into the chemical vapor deposition (CVD) reaction chamber.

（2）将预处理后的铜箔放入化学气相沉积（CVD）反应腔。

Evacuate the chamber and clean it with high-purity argon (Ar) 2-3 times to ensure that there is no residual gas in the chamber.

抽真空并用高纯氩气（Ar）清洗反应腔2-3次，确保反应腔内无残余气。

Heating and gas introduction: The reaction chamber is heated to 1000°C at a heating rate of 10°C/min.

加热和气体通入：以10°C/min 的升温速度将反应腔加热至1000°C。

Maintain a constant temperature of 1000°C, introduce hydrogen (H₂) and methane (CH₄), control the flow rate at 500 sccm, and the flow ratio of Ar: CH₄:H₂ is 20:8:1.

保持 1000°C 恒温，通入氢气（H₂）与甲烷（CH₄），控制流速在500 sccm，Ar：CH₄：H₂的流量比为20：8：1。

The sample was grown at 1000°C for about 10 minutes, then the methane gas was turned off, and the sample was cooled to room temperature at a natural cooling rate, thus obtaining a graphene composite material on the surface of copper foil.

在1000°C下生长10分钟左右，关闭甲烷气体，以自然冷却速率使样品温度冷却至室温，从而在铜箔表面制得石墨烯复合材料。

[n0029]

Example 2

实施例2

[0045]

This embodiment is the same as Embodiment 1, except that the CeO₂ suspension concentration is 0.1mol/L and the thickness of the CeO₂ coating is 50 nm.

本实施例同实施例1，不同之处在于，本实施例中CeO₂悬浮液浓度为0.1mol/L，涂覆的CeO₂涂层的厚度为50 nm。

This embodiment produces a uniformly distributed graphene composite material on the surface of copper foil.

本实施在铜箔表面制得均匀分布石墨烯复合材料。

[n0030]

Example 3

实施例3

[0047]

This embodiment is the same as Embodiment 1, except that the CeO₂ suspension concentration is 1mol/L and the thickness of the CeO₂ coating is 100μm.

本实施例同实施例1，不同之处在于，本实施例中CeO₂悬浮液浓度为1mol/L，涂覆的CeO₂涂层的厚度为100μm。

This embodiment produces a uniformly distributed graphene composite material on the surface of copper foil.

本实施在铜箔表面制得均匀分布石墨烯复合材料。

[n0031]

1. SEM images of the copper foil surface after pretreatment and after coating with cerium dioxide in Example 1. Figure 1(a) shows the surface morphology of the copper foil after pretreatment.

1、实施例1预处理后和涂覆二氧化铈后铜箔表面的SEM图，图1（a）为预处理后铜箔表面形貌。

It can be clearly seen that the surface of the pretreated copper foil is smooth and uniform, with no obvious oxide residue or impurity deposits, indicating that surface contaminants have been effectively removed.

可以清晰看到，经过预处理后的铜箔表面整体光滑且均匀，无明显的氧化物残留或杂质沉积，说明有效去除了表面污染物。

This provides an ideal support base for subsequent CeO₂ coating.

为后续CeO₂涂覆提供了理想的支撑基础。

Figure 1(b) shows the surface morphology of the copper foil after CeO₂ coating. As can be seen in the figure, the CeO₂ coating forms a uniformly distributed granular structure on the surface of the copper foil, with no obvious agglomeration or uncovered areas.

图1（b）展示了涂覆CeO₂后的铜箔表面形貌，图中可见，CeO₂涂层在铜箔表面形成了一层均匀分布的颗粒状结构，无明显团聚现象或未覆盖区域。

This particle distribution effectively increases the number of surface active sites, providing more sites for the adsorption and diffusion of carbon atoms during the subsequent CVD process.

这样的颗粒分布有效增加了表面活性位点的数量，为后续CVD过程中碳原子的吸附和扩散提供了更多的位点。

By comparing the SEM images of the original copper foil and the copper foil coated with cerium dioxide, it can be seen that the CeO₂ coating not only achieves uniform distribution in terms of physical coverage, but also further optimizes the surface properties through particle refinement, providing favorable conditions for subsequent graphene growth.

通过对比原始铜箔和涂覆二氧化铈后铜箔表面的SEM图像可以看出，CeO₂涂层不仅在物理覆盖上实现了均匀分布，还通过颗粒的细化进一步优化了表面特性，为后续石墨烯生长提供了有利条件。

[n0032]

2. Observe the growth of graphene on the surface of copper foil using an optical microscope system, and analyze its morphological characteristics and growth quality.

2、利用光学显微镜系统观察铜箔表面石墨烯的生长情况，并分析其形貌特征及生长质量。

As shown in Figure 2, the copper foil substrate with added cerium dioxide catalytic modification is uniformly covered with a layer of cerium dioxide graphene, indicating that cerium dioxide catalytic modification effectively improves the uniformity and quality of graphene.

从图2中可以看到，加入二氧化铈催化改性后的铜箔基底表面均匀覆盖了一层二氧化铈石墨烯，说明二氧化铈催化改性有效提升了石墨烯的均匀性和质量。

To confirm whether the material was successfully synthesized, XRD tests were performed to analyze the crystal structure.

为了确认材料是否成功合成，进行了XRD测试以分析晶体结构。

As shown in Figure 3, there are obvious material peaks on the prepared catalyst, and cerium dioxide has been successfully coated on the surface of copper foil.

如图3所示，在制备的催化剂上有明显的材料峰，二氧化铈已成功涂覆在铜箔表面。

In addition to the characteristic peaks of copper and cerium dioxide, the newly added peaks correspond to the crystal structure characteristics of carbon materials, such as the (002) crystal plane marked, indicating that carbon materials were successfully grown on Cu /CeO₂.

且除了铜和二氧化铈的特征峰外，新增的峰对应碳材料的晶体结构特征，如标注的（002）晶面，表明成功在Cu/CeO₂上生长出了碳材料。

[n0033]

3. The carbon yield and hydrogen production rate of Example 1 and Comparative Example 1 were measured. Figure 4(a) is a bar chart of carbon yield and Figure 4(b) is a graph of hydrogen production rate over time. As can be seen from Figure 4(a), the carbon yield was 10.5 when pure copper was used as the substrate; while when the copper surface was coated with cerium dioxide (Cu/CeO₂), the carbon yield increased significantly to 40.4.

3、测量实施例1和对比例1的碳收率和氢气生产率，图4（a）为碳收率柱状图，图4（b）为氢气生产率随时间的变化图，由图4（a）可知，纯铜作为基底时，碳收率为10.5；而当铜表面涂覆二氧化铈（Cu/CeO₂）后，碳收率大幅提升至40.4。

This indicates that cerium dioxide coating has a significant promoting effect on the generation of carbon materials because cerium dioxide has unique catalytic properties, which

can provide more active sites for the conversion of carbon sources, promote the adsorption, deposition and recombination of carbon atoms, thereby improving carbon yield.

这表明二氧化铈的涂覆对碳材料的生成有显著的促进作用，是因为二氧化铈具有独特的催化性能，能够为碳源的转化提供更多活性位点，促进碳原子的吸附、沉积和重组，从而提高了碳收率。

As shown in Figure 4(b), the H₂ production rate is significantly higher when copper and cerium dioxide (Cu + CeO₂) work together than when copper is on a pure copper substrate. The regulation of oxygen content and active sites by cerium dioxide creates a stable reaction atmosphere.

从图4（b）可知，铜与二氧化铈（Cu + CeO₂）共同作用时 H₂生产率明显高于纯铜基底，二氧化铈对氧含量和活性位点的调控，营造了稳定的反应氛围。

A stable environment ensures that the H₂ formation reaction can proceed continuously and stably, avoiding changes in the H₂ formation rate caused by fluctuations in reaction conditions.

稳定的环境保证了H₂生成反应能持续、稳定地进行，避免了因反应条件波动导致的H₂生成速率变化。

[n0034]

4. The thermal conductivity and resistivity of copper foil, graphene composite material prepared in Example 1 and graphene material prepared in Comparative Example 1 were measured. The specific results are shown in Figure 5(a) and Figure 5(b), respectively. As can be seen from Figure 5(a), the thermal conductivity was improved after carbon material was directly generated on the copper substrate, indicating that the introduction of carbon material improved the overall thermal conductivity of the material.

4、测量铜箔、实施例1制得的石墨烯复合材料和对比例1制得石墨烯材料的热导率和电阻率，具体结果分别如图5（a）和图5（b）所示，由图5（a）可见，在铜基底直接生成碳材料后，热导率有所提升，说明碳材料的引入改善了材料整体的热传导性能。

The thermal conductivity of the cerium dioxide-catalyzed graphene copper foil composite material (Cu/CeO₂/C) is further improved. This is because the catalytic effect of cerium dioxide optimizes the growth of graphene, and the cerium dioxide nanoparticles make the internal structure of the composite material more conducive to heat transfer and diffusion, thus enhancing the transmission efficiency.

而二氧化铈催化的石墨烯铜箔复合材料（Cu/ CeO₂/C）热导率进一步提高，这是因为二氧化铈的催化作用优化了石墨烯的生长，二氧化铈纳米颗粒使得复合材料内部结构更有利于热量的传递和扩散，增强了传输效率。

As shown in Figure 5(b), under the same temperature conditions, the graphene copper foil composite material catalyzed by cerium dioxide (Cu/CeO₂/C) has the lowest resistivity. Due to the high conductivity of graphene and the catalytic modification effect of cerium dioxide, the electron transport inside the composite material is smoother, which reduces the resistance of the material.

由图5（b）可见，在相同温度条件下，二氧化铈催化的石墨烯铜箔复合材料（Cu/CeO₂/C）电阻率最低，由于石墨烯的高导电性以及二氧化铈的催化改性作用，使得复合材料内部的电子传输更加顺畅，降低了材料的电阻。

[n0035]

Finally, it should be noted that the above embodiments of the present invention are merely examples for illustrating the present invention, and are not intended to limit the implementation of the present invention.

最后需要说明的是，本发明的上述实施例仅是为说明本发明所作的举例，而并非是对本发明实施方式的限定。

For those skilled in the field, other variations and modifications can be made based on the above explanation.

对于所属领域的普通技术人员来说，在上述说明的基础上还可以做出其他不同形式的变化和变动。

It is impossible to exhaustively list all possible implementation methods here.

这里无法对所有的实施方式予以穷举。

Any obvious variations or modifications derived from the technical solutions of this invention are still within the scope of protection of this invention.

凡是属于本发明的技术方案所引申出的显而易见的变化或变动仍处于本发明的保护范围之列。