Notice

DESCRIPTION CN119591095A

An apparatus and method for preparing graphene by plasma-assisted flash Joule heating

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一种等离子体辅助闪蒸焦耳热制备石墨烯的设备及方法

[0001]

Technical Field

技术领域

[n0001]

This invention belongs to the field of composite material manufacturing, and particularly relates to the preparation of flash graphene.

本发明属于复合材料制造领域，尤其涉及一种闪蒸石墨烯的制备。

[0003]

Background Technology

背景技术

[n0002]

Graphene is a two-dimensional material composed of carbon atoms with sp² hybrid orbitals. Its unique structure gives it outstanding mechanical, electrical, and chemical properties.

石墨烯是一种由碳原子以sp²杂化轨道组成的二维材料，特殊的结构使得其表现出相当出色的机械、电学、化学性质。

These properties make it a promising field in materials science, energy, and biomedicine.

这些属性使其在材料科学、能源、生物医学等领域有着广阔的前景。

Traditional methods for synthesizing graphene are divided into top-down exfoliation and bottom-up growth. The high defects caused by the top-down method and the high cost required by the bottom-up method have limited the large-scale production of graphene.

石墨烯的传统合成方法分为自上而下剥离和自下而上生长，自上而下方法导致的高缺陷与自下而上方法所需的高成本都限制了石墨烯的大规模生产。

Flash joule heating technology is an innovative technique for directly synthesizing large-scale graphene from low-cost carbon sources. It can convert various carbon sources into high-purity graphene in less than 100 milliseconds. This technology is highly efficient, pollution-free, and produces high-purity products, and is hailed as a revolutionary technology.

闪蒸焦耳热技术是一种将低成本碳源直接合成大规模石墨烯的创新技术，可在不到100毫秒的时间内将各种碳源转化为高纯度石墨烯。 该技术效率高、无污染、产物纯度高，被誉为革命性技术。

[n0003]

The carbon sources selected for flash Joule heating equipment typically contain many impurities. The traditional method for removing impurities is low-voltage discharge

pretreatment. Although this method can quickly remove most of the impurities from the carbon source, it still has the following shortcomings: First, the resistivity of the sample after low-voltage discharge pretreatment is significantly lower than that before pretreatment. The excessively low resistivity makes it difficult for the sample to rise to the corresponding temperature during flash Joule heating, thus making it difficult for the selected carbon source to be converted into graphene. Second, the energy generated by low-voltage discharge is relatively low, which may result in incomplete removal of impurities. In particular, for impurities that are tightly bound to the sample or difficult to decompose, low voltage may not provide enough energy to decompose or detach them from the sample.

闪蒸焦耳热设备所选取的碳源通常含有许多杂质，传统的除杂方法为低电压放电预处理，虽然该方法可以快速除去碳源中大部分杂质，但仍有以下不足之处：第一，低电压放电预处理后的样品电阻值较预处理前有明显的下降，过低的电阻值会导致样品在闪蒸焦耳热过程中难以升高至相应温度，从而使所选定的碳源难以转化为石墨烯；第二，低电压放电产生的能量相对较低，可能导致杂质去除不够彻底，尤其是对于那些与样品结合紧密或难以分解的杂质，低电压可能无法提供足够的能量来使其分解或脱离样品。

[n0004]

Plasma technology is an applied science that involves the fourth state of matter – plasma.

等离子体技术是一种涉及物质第四态-等离子体的应用科学。

Plasma is composed of ions, electrons, and neutral particles. It is highly reactive and can produce various impurity removal effects under the influence of electromagnetic fields.

等离子体由离子、电子和中性粒子组成，具有高度活性，能在电磁场作用下产生各种除杂处理效果。

[n0005]

Compared to traditional low-voltage discharge pretreatment for impurity removal, plasma impurity removal is highly efficient, does not cause surface damage, and does not change the sample resistance value.

相较于传统的低电压放电预处理除杂，等离子体除杂效率高、不会造成表面损伤，且不改变样品电阻值。

Therefore, it is proposed to use plasma purification treatment instead of traditional low-voltage discharge pretreatment.

因此，拟采用等离子体除杂处理代替传统的低电压放电预处理。

However, plasma impurity removal technology is limited to the sample surface. For some powdery or granular samples, plasma is difficult to effectively remove impurities. Furthermore, combining plasma purification with flash evaporation and Joule heating would create a more complex process and prolonged contact with air, negatively impacting the final purity of the graphene.

然而，等离子体除杂技术仅限于样品表面，对于一些粉末状或颗粒状样品，等离子体难以对样品进行有效的除杂处理。 此外，若将等离子体除杂处理与闪蒸焦耳热分部进行，更复杂的操作过程以及与空气的长时间接触都会对最终石墨烯的纯度造成负面影响。

[n0006]

Therefore, this patent abandons the traditional graphene preparation method and innovatively improves the flash evaporation Joule heating equipment, combining it with plasma technology. This overcomes the problem that plasma purification technology cannot effectively remove impurities from powdered samples, and designs a device for directly synthesizing high-purity graphene from carbon sources with many impurities.

因此，本专利摒弃了传统的石墨烯制备方法，创新性地对闪蒸焦耳热设备进行改进，使之与等离子体技术相结合，克服了等离子体除杂技术不能对粉末状样品进行有效除杂处理的问题，设计出一种从杂质较多的碳源直接合成高纯度石墨烯的设备。

[0009]

Summary of the Invention

发明内容

[n0007]

To address the problems existing in the prior art, this invention provides a device and method for preparing graphene by plasma-assisted flash Joule heating. This method can effectively remove impurities from the sample by fully contacting the powdered sample with plasma without changing the initial sample resistance value, thus achieving an ideal pretreatment effect.

针对现有技术存在的问题，本发明提供一种等离子体辅助闪蒸焦耳热制备石墨烯的设备及方法，能够在不改变初始样品电阻值的情况下，利用等离子体与粉末状样品进行充分接触，有效除去样品中的杂质，达到理想的预处理效果。

Meanwhile, the equipment can continuously perform flash evaporation after plasma treatment, keeping the sample in a vacuum state during the reaction stage, effectively isolating it from the external environment, and ensuring the purity and preparation efficiency of the sample.

同时该设备可以在等离子体处理后不间断地进行闪蒸步骤，使样品在反应阶段处于真空状态，有效地与外界环境相隔离，保证了样品的纯度和制备效率。

Furthermore, the tubular rotary reaction device used in this invention has the characteristics of simple structure and easy manufacturing, and can be modified as needed.

此外，本发明使用的管式旋转反应装置具有结构简单、易于制造的特点，可按需求进行改装。

[n0008]

This invention is achieved through the following technical solution:

本发明通过下述技术方案实现：

[n0009]

A device and method for preparing graphene by plasma-assisted flash Joule heating, characterized in that it includes a central control system, a vacuum system, a signal acquisition system, a power supply system, and a tubular rotary reaction device;

一种等离子体辅助闪蒸焦耳热制备石墨烯的设备及方法，其特征在于：包括中央控制系统、真空系统、信号采集系统、供电系统、管式旋转反应装置；

[n0010]

The central control system plays the role of transmitting commands, real-time monitoring, signal acquisition, and alarm processing in the entire equipment. It connects to the spectrometer, voltage and current sensors, and power supply system of the signal acquisition system.

中央控制系统在整个设备中起到传递命令、实时监控、信号采集、报警处理的作用，连接信号采集系统的光谱仪、信号采集系统的电压和电流传感器、供电系统；

[n0011]

The vacuum system includes a vacuum chamber, a vacuum pump, and a vacuum gauge. The vacuum chamber houses the tubular rotary reactor and has two exhaust valves on its outer wall. These valves allow the internal atmospheric pressure to be adjusted to match the external atmospheric pressure after the reaction. The vacuum chamber's excellent airtightness isolates the internal atmosphere from the external atmosphere, preventing the introduction of external impurities. The vacuum pump is connected to an exhaust valve on one side of the vacuum chamber's outer wall to remove impurities and maintain the required

vacuum level during the reaction. The vacuum gauge is located above the vacuum chamber and provides real-time feedback on the vacuum level inside the chamber.

真空系统包括真空箱，真空泵，真空表；真空箱用于放置管式旋转反应装置，外壁设有两个排气阀，可在反应后将箱内大气压调节至与外界大气压相平衡，真空箱良好的气密性可将箱内气氛与外界气氛相隔离，避免了外界杂质气体的引入；真空泵连接真空箱外壁一侧的一个排气阀，用于将箱内杂质气体抽去，并保持反应过程中所需的真空度；真空表位于真空箱上方，用于实时反馈箱内真空度；

[n0012]

The signal acquisition system is used to acquire temperature, voltage, and current signals during the reaction process and transmit them to the central control system. The signal acquisition system includes an infrared thermometer, voltage and current sensors. The infrared thermometer is embedded in the top groove of the vacuum chamber in the vacuum system and is connected to the central control system. The voltage and current sensors can measure the voltage on both sides of the sample and the current passing through the sample, and thus measure the resistance value of the sample. The voltage and current sensors are also connected to the two copper electrodes of the tubular rotary reaction device and the central control system.

信号采集系统用于采集反应过程中的温度、电压、电流信号，并将其传输至中央控制系统，信号采集系统包括红外测温仪、电压和电流传感器，红外测温仪嵌入真空系统中真空箱的顶部凹槽之中，与中央控制系统相连，电压和电流传感器可以测出样品两侧的电压和通过样品的电流，进而可以测出样品的电阻值，电压和电流传感器同时连接管式旋转反应装置的两个铜电极与中央控制系统；

[n0013]

The power supply system is used to provide electrical energy to the tubular rotary reactor and is connected to the tubular rotary reactor and the central control system.

供电系统用于为管式旋转反应装置提供电能，供电系统连接管式旋转反应装置、中央控制系统；

[n0014]

The tubular rotary reaction device includes a base plate, slide rail, double-ended studs, double-ended stud fixing device, electrode fixing device, copper electrode, transmission fixing device, inner and outer tube fixing device, inner tube, outer tube, inductor coil, and inductor coil fixing device.

管式旋转反应装置包括底板、滑轨、双头螺柱、双头螺柱固定装置、电极固定装置、铜电极、传动固定装置、内外管固定装置、内管、外管、电感线圈、电感线圈固定装置；

[n0015]

The base plate is used to support other components in the tubular rotary reactor and is bonded to the slide rail, double-headed stud fixing device, transmission fixing device, and inner and outer tube fixing device. The base plate is made of wood, but the material can be selected according to actual needs.

底板用于承载管式旋转反应装置中的其他部件，与滑轨、双头螺柱固定装置、传动固定装置、内外管固定装置相互粘连，底板为木制，也可根据实际需要选定材质；

[n0016]

The slide rail allows the electrode fixing device to slide within a certain range, preventing the electrode fixing device from falling off the slide rail and from colliding with the transmission fixing device or the inner and outer tube fixing device.

滑轨使电极固定装置在一定范围内滑动，使电极固定装置无法从滑轨中脱落且无法与传动固定装置、内外管固定装置相碰撞；

[n0017]

The double-ended stud is used to control the position of the electrode fixing device. The geometric orientation of the double-ended stud is fixed by the double-ended stud fixing device. The two ends of the double-ended stud have threads in different directions, which mesh with the left and right electrode fixing devices respectively. It can rotate under the drive of the motor inside the transmission fixing device.

双头螺柱用于控制电极固定装置的位置，双头螺柱的几何方位由双头螺柱固定装置所固定，双头螺柱两端具有方向不同的螺纹，分别于左右两个电极固定装置相互咬合，在传动固定装置内部电机的带动下可以旋转；

[n0018]

The electrode fixing device is used to fix the copper electrode. It consists of a trapezoidal bracket, a sector-shaped chuck, a handle, and a spring. By rotating the handle, the precise height of the copper electrode can be adjusted so that the copper electrode and the inner tube remain coaxial.

电极固定装置用于固定铜电极，由梯形支架、扇形卡盘、手柄、弹簧组成，通过转动手柄，可调节铜电极的精确高度，使铜电极与内管保持同轴心；

[n0019]

The transmission fixing device consists of a fixed bracket, a motor, and bearings. The bearings are embedded in the round hole at the top of the fixed bracket, and the motor is located inside the fixed bracket. The transmission fixing device can completely fix the outer tube and make the inner tube rotate axially under the drive of the motor.

传动固定装置由固定支架、电动机、轴承组成，轴承嵌入固定支架上部的圆孔之中，电动机位于固定支架内部，传动固定装置可使外管完全固定，并使内管在电动机的带动下沿轴向转动；

[n0020]

The inner and outer tube fixing device consists of a fixing bracket and bearings. The bearings are embedded in the round holes at the top of the fixing bracket. The bearings in the inner and outer tube fixing device are at the same height and coaxial with the bearings of the transmission fixing device. The inner tube is used to place the sample and serves as the actual reaction site of the sample. It is made of quartz. The two ends of the inner tube are embedded in two horizontal bearings on both sides. The two ends of the outer tube are respectively embedded in the arc-shaped grooves of the transmission fixing device and the inner and outer tube fixing device.

内外管固定装置由固定支架、轴承组成，轴承嵌入固定支架上部的圆孔之中，内外管固定装置中的轴承与传动固定装置轴承处于同一高度且同轴心，内管用于放置样品，作为样品的实际反应发生地，材

质为石英，内管两端嵌入两侧水平的两个轴承内，外管两端分别嵌入传动固定装置、内外管固定装置的弧形凹槽之中；

[n0021]

The inductor coil is used to generate plasma, and the inductor coil fixing device is used to fix the inductor coil. Its two ends are respectively connected to the transmission fixing device and the inner and outer tube fixing device.

电感线圈用于产生等离子，电感线圈固定装置用于电感线圈的固定，其两端分别连接传动固定装置与内外管固定装置。

[n0022]

An apparatus and method for preparing graphene by plasma-assisted flash Joule heating, characterized by the following steps:

一种等离子体辅助闪蒸焦耳热制备石墨烯的设备及方法，其特征在于采用以下步骤：

[n0023]

Step 1: Select a suitable carbon source, such as carbon black, anthracite, metallurgical coke, or biomass;

步骤一：选择合适的碳源，如炭黑、无烟煤、冶金焦、生物质；

[n0024]

Step 2: Place the carbon source in the middle of the inner tube of the tubular rotary reactor;

步骤二：将碳源放入管式旋转反应装置的内管中部位置；

[n0025]

Step 3: Move the electrode fixing device to both sides, put the copper electrode into the electrode fixing device, and adjust the height of the copper electrode by rotating the handle to make it coaxial with the inner tube.

步骤三：将电极固定装置移动至两侧，将铜电极放入电极固定装置中，通过旋转手柄调节铜电极的高度，使之与内管同轴心；

[n0026]

Step 4: Place the tubular rotary reaction apparatus into the vacuum chamber, turn on the infrared thermometer, and adjust the position of the tubular rotary reaction apparatus so that the light spot of the infrared thermometer passes through the outer tube and illuminates the middle position of the inner tube, so as to measure the temperature of the reaction sample.

步骤四：管式旋转反应装置放入真空箱中，打开红外测温仪，调整管式旋转反应装置的位置，使红外测温仪的光点透过外管，照射在内管中部位置，以达到对反应样品温度的测量；

[n0027]

Step 5: Open the exhaust valve on the outer wall of the vacuum chamber that is connected to the vacuum pump, close the exhaust valve on the outer wall of the vacuum chamber that is connected to the outside, close and lock the vacuum chamber door, start the vacuum pump, and turn off the vacuum pump when the pressure reaches about 10 mmHg. Then close the exhaust valve connected to the vacuum pump.

步骤五：打开真空箱外壁与真空泵相连的排气阀，关闭真空箱外壁与外界相连的排气阀，关闭并锁紧真空箱舱门，启动真空泵，抽至约10mmHg时关闭真空泵，关闭与真空泵相连的排气阀；

[n0028]

Step Six: By issuing commands through the central control system, high-frequency alternating current is supplied to the inductor coils in the tubular rotary reactor using the power supply system to generate plasma;

步骤六：通过中央控制系统施加命令，利用供电系统对管式旋转反应装置中的电感线圈提供高频交流电，产生等离子体；

[n0029]

Step 7: By issuing a command through the central control system, the power supply system supplies power to the motor of the tubular rotary reactor, causing the inner tube containing the powder sample to rotate for a period of time, so that the powder sample can come into full contact with the plasma.

步骤七：通过中央控制系统施加命令，利用供电系统对管式旋转反应装置的电动机供电，使装有粉末样品的内管旋转并持续一段时间，粉末样品可与等离子体充分接触；

[n0030]

Step 8: Stop supplying power to the inductor coil of the tubular rotary reactor, and use the power supply system to supply power to the motor of the tubular rotary reactor. Push the clamping device to press the electrodes at both ends together. When the resistance is adjusted to 1 ohm by controlling the pressure, stop supplying power to the motor.

步骤八：停止对管式旋转反应装置的电感线圈供电，利用供电系统对管式旋转反应装置的电动机供电，推动压紧装置将两端电极压紧，通过控制压力调整电阻至1欧姆时，停止对电动机供电；

[n0031]

Step 9: Input the corresponding parameters of flash Joule heating into the central control system, and issue a command to the power supply system through the central control system to supply power to the copper electrodes in the tubular rotary reactor. The sample is instantaneously heated by Joule heating at 120V, so that the sample rises to a high temperature of about 3000°C in 0.1 seconds and cools to room temperature at an extremely fast rate.

步骤九：向中央控制系统输入闪蒸焦耳热的相应参数，通过中央控制系统对供电系统施加命令，使供电系统对管式旋转反应装置中的铜电极供电，在120V电压下对样品进行瞬时的焦耳加热，使样品在0.1秒内升至约3000°C的高温，并以极快的速度冷却至室温；

[n0032]

Step 10: Turn off the power supply system and observe the sample temperature measured by the infrared thermometer. When the temperature reaches about 25°C, open the exhaust valve on one side of the vacuum chamber that connects to the outside. When the air pressure inside the vacuum chamber is balanced with the outside air pressure, open the vacuum chamber door and then take out the sample.

步骤十：关闭供电系统，观察红外测温仪所测出的样品温度，当温度达到约25°C时，打开真空箱一侧与外界相连的排气阀，待真空箱内气压与外界气压相平衡时打开真空箱舱门，随后取出样品。

[n0033]

Compared with the prior art, the present invention has at least the following advantages and effects:

本发明对于现有技术，至少具备如下优点及效果：

[n0034]

This invention combines plasma technology with flash Joule heating technology, allowing the reaction raw materials to undergo plasma treatment before undergoing Joule heating

reaction, replacing the traditional low-voltage discharge pretreatment process. This results in more thorough impurity removal and effectively improves the problem of uncontrollable sample resistance before flash evaporation.

本发明利用等离子体技术与闪蒸焦耳热技术相结合，使反应原料先进行等离子体与处理，后续进行焦耳热反应，代替了传统的低电压放电预处理工艺，使杂质去除更为彻底，有效改善了闪蒸前样品电阻值不可控的问题；

[n0035]

This invention keeps the entire reaction process in a vacuum, effectively isolating the reaction sample from the outside world and avoiding the influence of external conditions on the purity of the product.

本发明使整个反应过程都处于真空状态，使反应样品有效地与外界相隔离，避免了外界条件对产物纯度的影响；

[n0036]

This invention utilizes a transmission and fixing device to rotate the inner tube, causing the sample inside the inner tube to continuously tumble and achieve full contact with the plasma.

本发明利用传动固定装置使内管转动，使内管里的样品持续翻动，达到与等离子体充分接触的效果；

[n0037]

This invention utilizes a clamping device to press the copper electrode against the sample. By adjusting the resistance through the applied pressure, the subsequent flash evaporation Joule heating step can be carried out uninterrupted after plasma treatment, thus achieving continuous reaction and significantly improving experimental efficiency.

本发明利用压紧装置使铜电极压紧样品，通过施加的压力调节电阻，可在等离子体处理后不间断地进行接下来的闪蒸焦耳热步骤，实现了反应过程的持续进行，显著改善了实验效率；

[n0038]

The invention has a relatively simple structure and the technical means are simple and easy to implement, and it has outstanding substantial progress compared with the prior art.

本发明构造相对简单，技术手段简便易行，相对于现有技术具有突出的实质性进步。

[0042]

Attached Figure Description

附图说明

[n0039]

Figure 1 is a schematic diagram of the overall structure of a device for preparing graphene by plasma-assisted flash Joule heating;

图1是一种等离子体辅助闪蒸焦耳热制备石墨烯的设备整体结构示意图；

[n0040]

In Figure 1, the numbers represent: 1-Central control system; 2-1-Vacuum chamber; 2-2-Vacuum pump; 2-3-Vacuum gauge; 2-4-Exhaust valve; 3-1-Infrared thermometer; 3-2-Voltage and current sensor; 4-Power supply system; 5-Tube rotary reaction device.

图1中编号表示：1-中央控制系统；2-1-真空箱；2-2-真空泵；2-3-真空表；2-4-排气阀；3-1-红外测温仪；3-2-电压电流传感器；4-电源系统；5-管式旋转反应装置；

[n0041]

Figure 2 is a process flow diagram of the equipment and method for preparing graphene by plasma-assisted flash Joule heating;

图2是一种等离子体辅助闪蒸焦耳热制备石墨烯的设备及方法工艺流程图；

[n0042]

Figure 3 is a cross-sectional view of a tubular rotary reactor used in a plasma-assisted flash Joule heating method for preparing graphene.

图3是一种等离子体辅助闪蒸焦耳热制备石墨烯的设备及方法管式旋转反应装置的剖视图；

[n0043]

The numbers in Figure 3 represent: 5-1-base plate, 5-2-slide rail, 5-3-double-ended stud, 5-4-double-ended stud fixing device, 5-5-electrode fixing device, 5-6-copper electrode, 5-7-transmission fixing device, 5-8-inner and outer tube fixing device, 5-9-inner tube, 5-10-outer tube, 5-11-inductor coil, 5-12-inductor coil fixing device;

图3中编号表示：5-1-底板、5-2-滑轨、5-3-双头螺柱、5-4-双头螺柱固定装置、5-5-电极固定装置、5-6-铜电极、5-7-传动固定装置、5-8-内外管固定装置、5-9-内管、5-10-外管、5-11-电感线圈、5-12-电感线圈固定装置；

[n0044]

Figure 4 is an enlarged view and a cross-sectional view of the electrode fixing device in a tubular rotary reactor, which is part of an equipment and method for preparing graphene by plasma-assisted flash Joule heating.

图4是一种等离子体辅助闪蒸焦耳热制备石墨烯的设备及方法管式旋转反应装置中的电极固定装置的放大视图与剖视图；

[n0045]

The numbers in Figure 4 represent: 5-5-1-trapezoidal bracket, 5-5-2-fan-shaped chuck, 5-5-3-handle, 5-5-4-spring;

图4中编号表示：5-5-1-梯形支架、5-5-2-扇形卡盘、5-5-3-手柄、5-5-4-弹簧；

[n0046]

Figure 5 is an enlarged view and a cross-sectional view of the transmission and fixing device in a tubular rotary reactor, which is part of an equipment and method for preparing graphene by plasma-assisted flash Joule heating.

图5是一种等离子体辅助闪蒸焦耳热制备石墨烯的设备及方法管式旋转反应装置中的传动固定装置的放大视图与剖视图；

[n0047]

In Figure 5, the numbers represent 5-7-1-fixed bracket, 5-7-2-motor, and 5-7-3-bearing.

图5中编号表示5-7-1-固定支架、5-7-2-电动机、5-7-3-轴承。

[0052]

Detailed Implementation

具体实施方式

[n0048]

The technical solutions of the embodiments of the present invention will be further described in detail below with reference to the accompanying drawings.

下面将结合附图对本发明实施例的技术方案做进一步的详细描述。

[n0049]

As shown in Figure 1, this invention discloses an apparatus and method for preparing graphene by plasma-assisted flash Joule heating, including a central control system 1, a vacuum system 2, a signal acquisition system 3, a power supply system 4, and a tubular rotary reaction device 5.

如图1所示，本发明公开了一种等离子体辅助闪蒸焦耳热制备石墨烯的设备及方法，包括中央控制系统1、真空系统2、信号采集系统3、供电系统4、管式旋转反应装置5；

[n0050]

The central control system 1 plays the role of transmitting commands, real-time monitoring, signal acquisition, and alarm processing in the entire equipment. It connects to the spectrometer 3-1 of the signal acquisition system, the voltage and current sensors 3-2 of the signal acquisition system, and the power supply system 4.

中央控制系统1在整个设备中起到传递命令、实时监控、信号采集、报警处理的作用，连接信号采集系统的光谱仪3-1、信号采集系统的电压和电流传感器3-2、供电系统4；

[n0051]

The vacuum system includes a vacuum chamber 2-1, a vacuum pump 2-4, and a vacuum gauge 2-3. The vacuum chamber 2-1 is used to house the tubular rotary reaction apparatus 5. Two exhaust valves 2-4 are installed on its outer wall to adjust the atmospheric pressure inside the chamber to balance with the external atmospheric pressure after the reaction. The excellent airtightness of the vacuum chamber 2-1 isolates the internal atmosphere from the external atmosphere, preventing the introduction of external impurities. The vacuum pump 2-2 is connected to the vacuum chamber 2-1 and is used to remove impurities from the chamber and maintain the required vacuum level during the reaction. The vacuum gauge 2-3 is connected to the vacuum chamber 2-1 and is used to provide real-time feedback on the vacuum level inside the chamber.

真空系统包括真空箱2-1，真空泵2-4，真空表2-3；真空箱2-1用于放置管式旋转反应装置5，外壁设有两个排气阀2-4，可在反应后将箱内大气压调节至与外界大气压相平衡，真空箱2-1良好的气密性可

将箱内气氛与外界气氛相隔离，避免了外界杂质气体的引入；真空泵2-2连接真空箱2-1，用于将箱内杂质气体抽去，并保持反应过程中所需的真空度；真空表2-3连接真空箱2-1，用于实时反馈箱内真空度；

[n0052]

The signal acquisition system 3 is used to acquire temperature, voltage, and current signals during the reaction process and transmit them to the central control system 1. The signal acquisition system 3 includes an infrared thermometer 3-1 and a voltage and current sensor 3-2. The infrared thermometer 3-1 is embedded in the top groove of the vacuum chamber 2-1 in the vacuum system and is connected to the central control system 1. The voltage and current sensor 3-2 can measure the voltage on both sides of the sample and the current passing through the sample, and thus measure the resistance value of the sample. The voltage and current sensor 3-2 is also connected to the two copper electrodes 5-6 of the tubular rotary reaction device and the central control system 1.

信号采集系统3用于采集反应过程中的温度、电压、电流信号，并将其传输至中央控制系统1，信号采集系统3包括红外测温仪3-1、电压和电流传感器3-2，红外测温仪3-1嵌入真空系统中真空箱2-1的顶部凹槽之中，与中央控制系统1相连，压和电流传感器3-2可以测出样品两侧的电压和通过样品的电流，进而可以测出样品的电阻值，电压和电流传感器3-2同时连接管式旋转反应装置的两个铜电极5-6与中央控制系统1；

[n0053]

Power supply system 4 is used to provide electrical energy to tubular rotary reactor 5. Power supply system 4 is connected to tubular rotary reactor 5 and central control system 1.

供电系统4用于为管式旋转反应装置5提供电能，供电系统4连接管式旋转反应装置5、中央控制系统1；

[n0054]

The tubular rotary reaction device 5 includes a base plate 5-1, a slide rail 5-2, a double-ended stud 5-3, a double-ended stud fixing device 5-4, an electrode fixing device 5-5, a copper electrode 5-6, a transmission fixing device 5-4, an inner and outer tube fixing device 5-8, an inner tube 5-9, an outer tube 5-10, an inductor coil 5-11, and an inductor coil fixing device 5-12.

管式旋转反应装置5包括底板5-1、滑轨5-2、双头螺柱5-3、双头螺柱固定装置5-4、电极固定装置5-5、铜电极5-6、传动固定装置5-4、内外管固定装置5-8、内管5-9、外管5-10、电感线圈5-11、电感线圈固定装置5-12；

[n0055]

The base plate 5-1 is used to support other components in the tubular rotary reaction device 5 and is bonded to the slide rail 5-2 and the double-headed stud fixing device 5-4. The base plate 5-1 is made of wood, but the material can be selected according to actual needs.

底板5-1用于承载管式旋转反应装置5中的其他部件，与滑轨5-2、双头螺柱固定装置5-4相互粘连，底板5-1为木制，也可根据实际需要选定材质；

[n0056]

The slide rail 5-2 allows the electrode fixing device 5-5 to slide within a certain range, preventing the electrode fixing device 5-5 from falling off the slide rail 5-2 and from colliding with the transmission fixing device 5-7 and the inner and outer tube fixing device 5-8.

滑轨5-2使电极固定装置5-5在一定范围内滑动，使电极固定装置-5无法从滑轨5-2中脱落且无法与传动固定装置5-7、内外管固定装置5-8相碰撞；

[n0057]

The double-ended stud 5-3 is used to control the position of the electrode fixing device 5-5. The geometric orientation of the double-ended stud 5-3 is fixed by the double-ended stud

fixing device 5-4. The two ends of the double-ended stud 5-3 have threads in different directions, which mesh with the left and right electrode fixing devices 5-5 respectively. It can rotate under the drive of the motor 5-7-2 inside the transmission fixing device.

双头螺柱5-3用于控制电极固定装置5-5的位置，双头螺柱5-3的几何方位由双头螺柱固定装置5-4所固定，双头螺柱5-3两端具有方向不同的螺纹，分别于左右两个电极固定装置5-5相互咬合，在传动固定装置内部电机5-7-2的带动下可以旋转；

[n0058]

The electrode fixing device 5-5 is used to fix the copper electrode 5-6. It consists of a trapezoidal bracket 5-5-1, a fan-shaped chuck 5-5-2, a handle 5-5-3, and a spring 5-5-4. By rotating the handle 5-5-3, the precise height of the copper electrode 5-6 can be adjusted so that the copper electrode 5-6 and the inner tube 5-9 remain coaxial.

电极固定装置5-5用于固定铜电极5-6，由梯形支架5-5-1、扇形卡盘5-5-2、手柄5-5-3、弹簧5-5-4组成，通过转动手柄5-5-3，可调节铜电极5-6的精确高度，使铜电极5-6与内管5-9保持同轴心；

[n0059]

The transmission fixing device 5-7 consists of a fixed bracket 5-7-1, a motor 5-7-2, and a bearing 5-7-3. The bearing 5-7-3 is embedded in the round hole at the top of the fixed bracket.

The motor 5-7-2 is located inside the fixed bracket 5-7-1. The transmission fixing device 5-7 can completely fix the outer tube 5-10 and make the inner tube 5-9 rotate axially under the drive of the motor 5-7-2.

传动固定装置5-7由固定支架5-7-1、电动机5-7-2、轴承5-7-3组成，轴承5-7-3嵌入固定支架上部的圆孔之中，电动5-7-2机位于固定支架5-7-1内部，传动固定装置5-7可使外管5-10完全固定，并使内管5-9在电动机5-7-2的带动下沿轴向转动；

[n0060]

The inner and outer tube fixing device 5-8 consists of a fixing bracket 5-7-1 and a bearing 5-7-3. The bearing 5-7-3 is embedded in the round hole at the top of the fixing bracket 5-7-1. The bearing 5-7-3 in the inner and outer tube fixing device is at the same height and coaxial with the bearing 5-7-3 in the transmission fixing device. The inner tube 5-9 is used to place the sample and serves as the actual reaction site of the sample. It is made of quartz. Both ends of the inner tube 5-9 are embedded in two horizontal bearings 5-7-3 on both sides. Both ends of the outer tube 5-10 are embedded in the arc-shaped grooves of the transmission fixing device 5-7 and the inner and outer tube fixing device 5-8, respectively.

内外管固定装置5-8由固定支架5-7-1、轴承5-7-3组成，轴承5-7-3嵌入固定支架5-7-1上部的圆孔之中，内外管固定装置中的轴承5-7-3与传动固定装置轴承5-7-3处于同一高度且同轴心，内管5-9用于

放置样品，作为样品的实际反应发生地，材质为石英，内管5-9两端嵌入两侧水平的两个轴承5-7-3内，外管5-10两端分别嵌入传动固定装置5-7、内外管固定装置5-8的弧形凹槽之中；

[n0061]

Inductor coil 5-11 is used to generate plasma, and inductor coil fixing device 5-12 is used to fix inductor coil 5-11. Its two ends are respectively connected to transmission fixing device 5-7 and inner and outer tube fixing device 5-8.

电感线圈5-11用于产生等离子，电感线圈固定装置5-12用于电感线圈5-11的固定，其两端分别连接传动固定装置5-7与内外管固定装置5-8。

[n0062]

A specific process for a plasma-assisted flash Joule heating method for preparing graphene includes the following steps:

一种等离子体辅助闪蒸焦耳热制备石墨烯的设备及方法的具体流程，包括以下步骤：

[n0063]

Step 1: Weigh 0.1g of carbon black.

步骤一：称取0.1g炭黑。

[n0064]

Step 2: Place the carbon black into the middle of the inner tube of the tubular rotary reactor.

步骤二：将炭黑放入管式旋转反应装置的内管中部位置。

[n0065]

Step 3: Move the electrode fixing device to both sides, put the copper electrode into the electrode fixing device, and adjust the height of the copper electrode by rotating the handle to make it coaxial with the inner tube.

步骤三：将电极固定装置移动至两侧，将铜电极放入电极固定装置中，通过旋转手柄调节铜电极的高度，使之与内管同轴心。

[n0066]

Step 4: Place the tubular rotary reaction apparatus into the vacuum chamber, turn on the infrared thermometer, and adjust the position of the tubular rotary reaction apparatus so that the light spot of the infrared thermometer passes through the outer tube and illuminates the middle position of the inner tube, so as to measure the temperature of the reaction sample.

步骤四：管式旋转反应装置放入真空箱中，打开红外测温仪，调整管式旋转反应装置的位置，使红外测温仪的光点透过外管，照射在内管中部位置，以达到对反应样品温度的测量。

[n0067]

Step 5: Open the exhaust valve on the outer wall of the vacuum chamber that connects to the vacuum pump, close the exhaust valve on the outer wall of the vacuum chamber that connects to the outside, close and lock the vacuum chamber door, start the vacuum pump, and turn off the vacuum pump when the pressure reaches about 10 mmHg. Then close the exhaust valve connected to the vacuum pump.

步骤五：打开真空箱外壁与真空泵相连的排气阀，关闭真空箱外壁与外界相连的排气阀，关闭并锁紧真空箱舱门，启动真空泵，抽至约10mmHg时关闭真空泵，关闭与真空泵相连的排气阀。

[n0068]

Step Six: By issuing commands through the central control system, high-frequency alternating current is supplied to the inductor coils in the tubular rotary reactor using the power supply system to generate plasma.

步骤六：通过中央控制系统施加命令，利用供电系统对管式旋转反应装置中的电感线圈提供高频交流电，产生等离子体。

[n0069]

Step 7: By issuing a command through the central control system, the power supply system supplies power to the motor of the tubular rotary reactor, causing the inner tube containing the powder sample to rotate for a period of time, so that the powder sample can come into full contact with the plasma.

步骤七：通过中央控制系统施加命令，利用供电系统对管式旋转反应装置的电动机供电，使装有粉末样品的内管旋转并持续一段时间，粉末样品可与等离子体充分接触。

[n0070]

Step 8: Stop supplying power to the inductor coil of the tubular rotary reactor, and use the power supply system to supply power to the motor of the tubular rotary reactor. Push the clamping device to press the electrodes at both ends together. When the resistance is adjusted to about 1 ohm by controlling the pressure, stop supplying power to the motor.

步骤八：停止对管式旋转反应装置的电感线圈供电，利用供电系统对管式旋转反应装置的电动机供电，推动压紧装置将两端电极压紧，通过控制压力调整电阻至1欧姆左右时，停止对电动机供电。

[n0071]

Step 9: Input the corresponding parameters of flash Joule heating into the central control system, and issue a command to the power supply system through the central control system to supply power to the copper electrodes in the tubular rotary reactor. The sample is instantaneously heated by Joule heating at 120V, so that the sample rises to a high temperature of about 3000°C within 0.1 seconds and cools to room temperature at an extremely fast rate.

步骤九：向中央控制系统输入闪蒸焦耳热的相应参数，通过中央控制系统对供电系统施加命令，使供电系统对管式旋转反应装置中的铜电极供电，在120V电压下对样品进行瞬时的焦耳加热，使样品在0.1秒内升至约3000°C的高温，并以极快的速度冷却至室温。

[n0072]

Step 10: Turn off the power supply system and observe the sample temperature measured by the infrared thermometer. When the temperature reaches about 25°C, open the exhaust valve on one side of the vacuum chamber that connects to the outside. When the air pressure inside the vacuum chamber is balanced with the outside air pressure, open the vacuum chamber door and then take out the sample.

步骤十：关闭供电系统，观察红外测温仪所测出的样品温度，当温度达到约25°C时，打开真空箱一侧与外界相连的排气阀，待真空箱内气压与外界气压相平衡时打开真空箱舱门，随后取出样品。