

**补充材料  
Supplementary Materials for**

**废物中的稀土元素  
Rare earth elements from waste**

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## 补充文本

### Supplementary Text

#### 补充文本1。回收率的计算。

##### Supplementary Text 1. Calculation of the recovery yield.

CFA原料 $c_{\text{total}}(\text{CFA raw})$ 中的总稀土含量是通过使用HF:HNO<sub>3</sub>消解方法的总消解来测量的(详见材料和方法, 图S6)。  
The total REE contents in CFA raw materials,  $c_{\text{total}}(\text{CFA-Raw})$ , was measured by total acid-leachable REE contents in CFA raw materials,  $c_0(\text{CFA-Raw})$ , were measured by HCl or HNO<sub>3</sub> leaching of the CFA raw materials. The REE recovery yield by acid leaching the CFA raw materials was calculated by Eq. S1,

$$Y_0 = \frac{c_0(\text{CFA Raw})}{c_{\text{total}}(\text{CFA-Raw})} \quad (\text{S1})$$

将CFA原料和炭黑混合并进行FJH活化过程。  
The CFA raw materials and carbon black were mixed and underwent the FJH activation process. The obtained solid is termed as activated CFA. The acid-leachable REE content in the activated CFA,  $c(\text{activated CFA})$ , was measured by the same acid leaching procedure of CFA after FJH. The REE recovery yield by acid leaching the activated CFA was calculated by Eq. S2,

$$Y = \frac{c(\text{activated CFA})}{c_{\text{total}}(\text{CFA Raw})} \quad (\text{S2})$$

因此,  $Y/Y_0$ 的比率由等式S3计算。

Hence, the ratio of  $Y/Y_0$  was calculated by Eq. S3,

$$\frac{Y}{Y_0} = \frac{c(\text{activated CFA})}{c_0(\text{CFA Raw})} \quad (\text{S3})$$

$Y/Y_0$ 可作为通过FJH活化过程提高CFA稀土回收率的指标。

The  $Y/Y_0$  could be used as an index of the increase of the REE recovery yield from CFA by the FJH activation process. If  $Y/Y_0 > 1$ , the activation process has a positive effect and improves the recovery yield. Similarly, for BR and e-waste, the increase of recovery yield is calculated by using Eq. S4 and Eq. S5:

$$\frac{Y}{Y_0} = \frac{c(\text{activated BR})}{c_0(\text{BR Raw})} \quad (\text{S4})$$

$$\frac{Y}{Y_0} = \frac{c(\text{activated e-waste})}{c_0(\text{e-waste Raw})} \quad (\text{S5})$$

#### 补充文本2。扩大FJH流程的策略。

##### Supplementary Text 2. Strategy for scaling up the FJH process.

焦耳加热广泛应用于许多设备和工业过程, 如电加热器和管式炉。  
Joule heating is widely used in many devices and industrial processes, such as electric heaters and tube furnace. Flash Joule heating (FJH) reported here, is intrinsically a Joule heating process. For traditional Joule heating process, a constant and long-time alternating current (AC) or direct current (DC) is used as the electrical resources. In contrast, in the FJH process, which is firstly invented by our group for the synthesis of turbostratic graphene (24), a pulsed DC provided by the discharging of capacitors is used, which provides a much higher temperature in a short duration. The FJH process is scalable. We here conduct theoretical analysis of the FJH process to identify the key parameters determining the temperature; then, we experimentally demonstrate the scaling-up of the FJH process; we also provide conceptual prototype of the continuous production; lastly, we briefly mention the ongoing industrial scale application of the FJH process on graphene synthesis, which can be easily shifted for the REE recovery purpose.

#### 标度规则的理论分析。

##### Theoretical analysis of the scaling rule.

对于放大, 当样品质量增加时, 保持恒定的温度值和温度分布至关重要。  
For scaling up, it is critical to maintain a constant temperature value and temperature distribution when the sample mass is increased.

通过等式S6计算焦耳加热产生的热量( $Q$ )。

The heat amount ( $Q$ ) produced by Joule heating is calculated by Eq. S6,

$$Q = I^2 R t \quad (\text{S6})$$

其中I是通过样品的电流，R是样品电阻，t是时间。

where  $I$  is the current passing through the sample,  $R$  is the sample resistance, and  $t$  is the time.

考慮到单位体积热量( $Q_v$ )，可以将等式修改为等式S7，

Considering heat amount per volume ( $Q_v$ ), the equation could be revised to Eq. S7,

$v=2e(S7)$ , 其中 $Q_v$ 是每体积的热量， $j$ 是电流密度， $e$ 是样品的电阻率。

$$Q_v = j^2 \rho_e t \quad (S7)$$

where  $Q_v$  is the heat per volume,  $j$  is the current density, and  $\rho_e$  is the resistivity of the sample.

根据传热公式，可通过等式S8计算温度变化。

According to the heat transfer formula, the change of temperature could be calculated by Eq. S8,

$=p(S8)$ , 其中 $\Delta T$ 是温度变化， $m$ 是样品质量， $C_p$ 是比热容。

$$Q = C_p m \Delta T \quad (S8)$$

where  $\Delta T$  is the change of temperature,  $m$  is the mass of the sample, and  $C_p$  is the specific heat capacity. Since the specific capacity ( $C_p$ ) is a constant for a specific type of sample, the change of

temperature is determined by the heat amount.

此外，我们可以将每体积的公式S8修改为公式S9。

Furthermore, we can revise the Eq. S8 per volume to Eq. S9,

$v=p(S9)$ , 其中 $m$ 是样品的密度。

$$Q_v = C_p \rho_m \Delta T \quad (S9)$$

由于样品的密度( $\rho_m$ )和比热容( $C_p$ )恒定，温度变化与 $Q_v$ 成线性比例。换句话说，无论样品质量如何，只要 $Q_v$ 保持不变，就可以实现相同的温度。

where  $\rho_m$  is the density of the sample. Since the density ( $\rho_m$ ) and the specific heat capacity ( $C_p$ ) are constant for the sample, the change of temperature is linearly proportional to  $Q_v$ . In other word, regardless of the sample mass, the same temperature could be achieved as long as the  $Q_v$  remains the same.

然后，根据等式S7，由于样品的电阻率( $\rho_e$ )对于样品是恒定的，为了在放大样品质量( $m$ )时保持恒定的 $Q_v$ 和 $t$ ，我们需要保持恒定的 $j$ ，其由等式S10确定。

Then, according to Eq. S7, since the resistivity ( $\rho_e$ ) of the sample is constant for the sample, to keep a constant  $Q_v$  and  $t$  when scaling up the sample mass ( $m$ ), we need to maintain a constant  $j$ , which is determined by Eq. S10,

$$j = \frac{I}{S} \quad (S10)$$

其中I是通过样品的电流，S是横截面积。

where  $I$  is the current passing through the sample, and  $S$  is the cross-sectional area. The current is calculated by Eq. S11,

$$I = \frac{q}{t} \quad (S11)$$

其中 $q$ 是电荷， $t$ 是时间。

假设电容器组中的电荷在时间内放电，可以通过等式S12计算电荷，

Supposing that the charges in the capacitor bank are discharged within the time of  $t$ , the charge could be calculated by Eq. S12,

$$q = CV \quad (S12)$$

where  $C$  is the total capacitance of the capacitor bank, and  $V$  is the charging voltage.

根据等式S10至S12，电流密度由等式S13确定，

According to Eq. S10 to S12, the current density is determined by Eq. S13,

$$j = \frac{CV}{St} \quad (S13)$$

由于我们始终使用石英管，因此样品通常为圆柱形，因此样品质量通过等式S14计算。

The sample is usually cylinder-shaped since we always use a quartz tube, so the sample mass is calculated by Eq. S14,

$$m = \rho_m S L \quad (S14)$$

where  $S$  is the cross-sectional area, and  $L$  is the sample length.

根据等式S13和S14，通过等式S15确定电流密度。

According to Eq. S13 and S14, the current density is determined by Eq. S15

$$j = \frac{CV \rho_m L}{mt} \quad (S15)$$

样品的密度( $\rho_m$ )是恒定的，我们可以改变样品的横截面积以保持恒定的样品长度 $L$ 。因此，当样品的质量( $m$ )增加时，为了保持恒定的 $j$ 和 $t$ ，有两种方法：(1)增加焦耳加热电压 $V$ ；和/或(2)增加C的电容。

The density of the sample ( $\rho_m$ ) is constant, and we can change the sample cross-sectional area to maintain a constant sample length of  $L$ . Hence, to maintain a constant  $j$  and  $t$  when the mass ( $m$ ) of the sample is increased, there are two approaches: (1) to increase the Joule heating voltage  $V$ ; and/or (2) to increase the capacitance of  $C$ .

放大演示。

Scaling up demonstration.

我们在这里演示了通过使用第一种策略，增加电压来放大FJH过程。

We here demonstrated the scaling up of the FJH process by using the first strategy, increasing the voltage. In most of the experiments (table S1) with the sample mass of  $m_0 = 0.2$  g, we use a FJH voltage of  $V_0 = 120$  V and capacitance of  $C_0 = 60$  mF, and a cylinder-shape sample

直径D0=8 mm ( $S_0 \sim 50 \text{ mm}^2$ )，为了将反应放大到 $m_1=2 \text{ g}$ ，我们使用了直径为D1=16 mm ( $S_1 \sim 200 \text{ mm}^2$ )，与直径为 $D_0 = 8 \text{ mm}$  ( $S_0 \sim 50 \text{ mm}^2$ )。To scale up the reaction to  $m_1 = 2 \text{ g}$ , we used a large sample size with diameter of  $D_1 = 16 \text{ mm}$  ( $S_1 \sim 200 \text{ mm}^2$ )。According to Eq. S14 to S15, 大规模样品的电压应为 $V_1=2.5 \times V_0=300 \text{ V}$ 。我们以CFA-C为例，演示了质量高达2g的FJH过程(图S16A)。  
根据等式S14至S15，大规模样品的电压应为 $V_1=2.5 \times V_0=300 \text{ V}$ 。我们以CFA-C为例，演示了质量高达2g的FJH过程(图S16A)。  
for the large-scale sample should be  $V_1 = 2.5 \times V_0 = 300 \text{ V}$ . We used CFA-C as an example and demonstrated the FJH process with mass up to 2 g (fig. S16A). The reaction conditions are:  $m_1 = 2 \text{ g}$ ,  $D_1 = 16 \text{ mm}$ ,  $V_1 = 300 \text{ V}$ , and  $t = 1 \text{ s}$ 。The result is shown in fig. S16B. The CFA-C is successfully activated with the increase of REE recovery yields to 150% to 190%, comparable to the results of the sample with smaller mass (Fig. 2H).  
结果如图S16B所示。REE被成功激活，REE回收率增加到150%-190%，与质量较小的样品的结果相当(图2H)。

我们还可以通过增加电容(C)来扩大FJH过程。在本研究中使用的  
We can also scale up the FJH process by increasing the capacitance ( $C$ ). In our first  
第一代FJH系统中，电容器组由10个商用铝电解电容器组成，总电容为 $C_0=60\text{ mF}$ 。我们还在实验室  
generation FJH system, which is used in this work, the capacitor bank is composed of 10  
室建立了第二代FJH系统，总电容 $C_2=0.624\text{ F}$ 。我们实现了每批 $m_2=5\text{ g}$ 的样品质量。通过  
built a second generation FJH system in our lab with the total capacitance of  $C_2 = 0.624 \text{ F}$ . By  
使用 $V_2=380\text{ V}$ 的FJH电压和 $C_2=0.624\text{ F}$ 的电容，我们实现了每批 $m_2=5\text{ g}$ 的样品质量。  
using a FJH voltage of  $V_2 = 380 \text{ V}$  and capacitance of  $C_2 = 0.624 \text{ F}$ , we realized the sample mass  
实际上，我们的实验室已经将煤转化为石墨烯的FJH工艺规模扩大到每天17.6公斤左右。由于石墨烯合成的主要FJH  
of  $m_2 = 5 \text{ g}$  per batch. Actually, our lab has already scaled the FJH process for the conversion of  
工艺和稀土元素回收的主要FJH工艺基本相同，因此可以安全地假设，在我们的研究实验室中，用于稀土元素回收的FJH工艺的生产率大于10 kg  
coal to graphene to  $\sim 17.6 \text{ kg day}^{-1}$ . Since the main FJH process for graphene synthesis and for the  
 $\text{day}^{-1}$   
REE recovery is pretty the same, it is safe to presume that the FJH process for REE recovery has  
the production rate of  $> 10 \text{ kg day}^{-1}$  in our research lab.

**连续生产的概念原型。**  
*The conceptual prototype of the continuous production.*  
 在我们目前的工作中，FJH过程是分批进行的。  
 In our present work, the FJH process is done batch-by-batch. We presume that the FJH process could be done automatically in a continuous manner as we have demonstrated in our graphene production process. We provide here a conceptual prototype of the continuous production reactor (fig. S17). The continuous production process consists of four steps. The原料的混合物被装载到传送带上的腔室中。  
 mixture of CFA/CB feedstock is loaded onto the chamber on the conveyor belt. The sample is compressed to a specific resistance. The sample then undergoes the FJH reaction. Lastly, the CFA产物。  
 activated CFA product is collected. This is only one possible design. Considering the various and设备，如滚带工艺，我们的FJH工艺可以整合到其中，以回收稀土元素。  
 commercially available continuous production processes and equipment, such as rolling-belt processes, our FJH process could be integrated into them for the purpose of REE recovery.

**FJH工艺的商业规模正在扩大。**  
*The commercial scaling up of the FJH process is ongoing.*  
环球物质股份有限公司正在将FJH工艺应用于石墨烯的工业规模生产(<https://www.universalmatter.com/>)到2022年第二季度，目标产量为每天1吨，到2023年，进一步扩大到每天100吨。  
The application of the FJH process to an industrial scale for the production of graphene is ongoing by Universal Matter Inc. (<https://www.universalmatter.com/>), with the targeted production rate of 1 ton day<sup>-1</sup> by Q2 2022, and further scaling to 100 tons per day by 2023. To date, the production rate doubles every 9 weeks. The equipment and process designed and optimized for graphene production could be applied for REE recovery purpose. At the industrial scale, alternative current (AC) is a more feasible electrical resource than the direct current (DC). According to our above analysis, optimizing the voltage is one approach to scale up the FJH process. In industry, the high voltage or even ultrahigh voltage up to hundreds of kV are mature technologies, which could be applied in the REE process. Hence, the FJH process has a tried route to scalability for the REE recovery. The ongoing commercial scaling of the FJH process paves the way for future REE recovery from large-scale waste products.  
为石墨烯生产设计和优化的设备和工艺可用于稀土元素回收。  
在工业规模上，交流电(AC)比直流电(DC)更可行。  
因此，FJH过程为REE恢复的可扩展性提供了一条可行的途径。  
FJH工艺正在进行的商业规模化为今后从大规模废物中回收稀土元素铺平了道路。

### 补充文本3。能耗计算和利润估算。

### Supplementary Text 3. Energy consumption calculation and profit estimation.

电能(E)消耗通过等式S16计算

The electrical energy ( $E$ ) consumption is calculated by Eq. S16,

$$E = \frac{(V_1^2 - V_2^2) \times C}{2 \times M} \quad (\text{S16})$$

其中 $V_1$ 和 $V_2$ 分别是FJH后的起始电压和结束电压， $C$ 是电容(60 mF)， $M$ 是每批的质量。

where  $V_1$  and  $V_2$  are the start voltage and the end voltage after the FJH, respectively,  $C$  is the capacitance (60 mF), and  $M$  is the mass per batch.

在 $V_1=120\text{ V}$ ,  $V_2=0\text{ V}$ ,  $C=60\text{ mF}$  and  $M=0.2\text{ g}$ 的典型实验中，能量计算为 $E=2.16\text{ kJ g}^{-1}=6.0 \times 10^{-4}\text{ kWh g}^{-1}=600\text{ kWh ton}^{-1}$

In a typical experiment with  $V_1 = 120\text{ V}$ ,  $V_2 = 0\text{ V}$ ,  $C = 60\text{ mF}$ , and  $M = 0.2\text{ g}$ , the energy is calculated to be,

$$E = 2.16\text{ kJ g}^{-1} = 6.0 \times 10^{-4}\text{ kWh g}^{-1} = 600\text{ kWh ton}^{-1}$$

考虑到美国德克萨斯州的工业电价为 $0.02\text{ kWh}^{-1}$ 美元，激活1吨CFA的成本将为 $P(\text{电力})=12\text{ $ ton}^{-1}$ 。

Considering that the industrial electricity price in Texas, USA is  $\$0.02\text{ kWh}^{-1}$ , the cost for activating 1 ton of CFA would be  $P(\text{electricity}) = 12\text{ $ ton}^{-1}$ .

与未经活化过程的稀土元素回收相比，通过活化过程增加的回收稀土量可通过等式S17计算。

Compared with the REE recovery without the activation process, the increased recovered

REE amount by the activation process could be calculated by Eq. S17,

$$= \times \times (\text{S17}) \text{ 其中 } m \text{ 是稀土元素的质量, } m \text{ 是CFA原料的质量, } c \text{ 是CFA原料中稀土元素的可提取含量, } \Delta Y \text{ 是热活化过程后提高的稀土回收率。}$$

$$m = M \times c \times \Delta Y \quad (\text{S17})$$

where  $m$  is the mass of the REE,  $M$  is the mass of CFA raw materials,  $c$  is the extractable content of REE from CFA raw materials, and  $\Delta Y$  is the improved REE recovery yield after the thermal activation process.

对于从CFA中回收稀土元素，Sc和临界稀土元素，包括Nd、Eu、Dy、Er和Tb，贡献了80%以上的值(4)。

For REE recovery from CFA, the Sc, and critical REE, including Nd, Eu, Dy, Er, and Tb, contribute to more than 80% of the values (4). Considering 1 ton of CFA-C, the improved recovered REE amount would be:

$$m(\text{Sc}) = 23\text{ g}, m(\text{Nd}) = 29\text{ g}, m(\text{Eu}) = 2\text{ g}, m(\text{Dy}) = 5\text{ g}, m(\text{Er}) = 3\text{ g}, \text{ and } m(\text{Tb}) = 3\text{ g}$$

2019年7月，这些关键稀土元素的价格为Sc( $P=5735\text{ kg}^{-1}$ )，Nd( $P=64\text{ kg}^{-1}$ )，Eu( $P=285\text{ kg}^{-1}$ )，Dy( $P=375\text{ kg}^{-1}$ )，Er( $P=30\text{ kg}^{-1}$ )和Tb( $P=770\text{ kg}^{-1}$ )<sup>(53)</sup>。

The price of these critical REE are Sc ( $P = \$5735\text{ kg}^{-1}$ ), Nd ( $P = \$64\text{ kg}^{-1}$ ), Eu ( $P = \$285\text{ kg}^{-1}$ ), Dy ( $P = \$375\text{ kg}^{-1}$ ), Er ( $P = \$30\text{ kg}^{-1}$ ), and Tb ( $P = \$770\text{ kg}^{-1}$ ) in July 2019 (53). The values of the individual REE would be:

$$P(\text{Sc}) = \$132, P(\text{Nd}) = \$1.8, P(\text{Eu}) = \$0.6, P(\text{Dy}) = \$1.9, P(\text{Er}) = \$0.1, \text{ and } P(\text{Tb}) = \$2.3$$

采用热活化工艺的稀土元素的附加值为： $P(\text{REE}) = 139\text{吨}^{-1}$ 美元

The value added of the REEs with the thermal activation process would be:

$$P(\text{REE}) = \$139\text{ ton}^{-1}$$

在这里，我们假设直接浸出过程和活化浸出过程的稀土分离成本是恒定的。

Here, we presume that the REE separation cost is constant for the directly leaching process and the activation-leaching process. The profit percentage for the activation process is calculated by Eq. S18,

$$\text{Profit percentage} = \frac{P(\text{REE}) - P(\text{electricity})}{P(\text{electricity})} \quad (\text{S18})$$

经计算，利润率约为11%。

The profit percentage is calculated to be ~11. The profit margin could be larger considering the improved recovery of other REE.

补充文本4。

渗透液中的杂质及其去除设想。

#### Supplementary Text 4. The impurities in the leachate and the envision on impurity removal.

稀土元素分离通常分为一级分离(稀土元素与其他杂质元素的分离)和二级分离(单个稀土元素的分离)(54)。

REE separation is generally classified as primary separation (the separation of REE from other impurity elements), and secondary separation (the separation of individual REE) (54). The presence of metal impurities in the REE-containing leachate affects the subsequent REE separation efficiency by methods such as solvent extraction and ion exchange (51, 54, 55). Hence, 通常需要在稀土元素分离之前去除杂质(54)。因此，通常需要在稀土元素分离之前去除杂质(54)。在这里，分析了渗透液中的杂质，并且通常需要在稀土元素分离之前去除杂质(54)。在这里，分析了渗透液中的杂质，并且通常需要在稀土元素分离之前去除杂质(54)。Here, the impurities in the leachate are analyzed, and the removal of metal impurities and selective separation of individual REE from the leachate are envisioned.

渗透液中的杂质。

#### The impurities in the leachate.

矿石和二次废物的成分通常因来源不同而显著不同，因此渗透液中的杂质在类型和含量上也存在显著差异(54)。

The composition of ores and secondary wastes often differ significantly from one source to another, and hence the impurities in the leachate also vary significantly in types and contents (54). The major impurities include Al, Si, Fe, Ca, Mg, Zn, Co, Ni, Cr, Cu, etc. Impurities, and especially the ratio of impurity content to REE content, could affect REE recovery and precipitation efficiencies by co-precipitation with REE, consuming the reagents, emulsification, etc. (54, 55). Here, we measured the impurity contents in our REE leachates from different secondary resources.

对于CFA-F，主要杂质包括铝、硅、铁、钙(10至100 ppm)、镁(1至10 ppm)、锌、镍、铬(0.1至1 ppm)和钴(0.01至0.1 ppm)(图S18)。对于CFA-F，主要杂质包括Al, Si, Fe, Ca(10 to 100 ppm), Mg (1 to 10 ppm), Zn, Ni, Cr (0.1 to 1 ppm), and Co (0.01 to 0.1 ppm) (fig. S18). We found that ratio of  $c(\text{REE})/c(\text{Impurity})$  for the activated CFA-F is larger than that of the CFA-F raw materials for all the tested pH conditions ranging from 0 to 2 (fig. S18D)。这意味着FJH工艺有助于提高稀土元素的浸出能力，同时在一定程度上降低CFA-F的杂质浸出能力，这有利于杂质去除。

对于CFA-C，主要杂质包括铝、硅(>100 ppm)、铁、钙、镁(10至100 ppm)、锌(1至10 ppm)、镍、铬(0.1至1 ppm)和钴(0.01至0.1 ppm)(图S19)。类似地，当测试pH值为0和1时，活化CFA-C的 $c(\text{REE})/c(\text{Impurity})$ 比CFA-C原料的 $c(\text{REE})/c(\text{Impurity})$ 更大(图S19)。Similarly, the ratio of  $c(\text{REE})/c(\text{Impurity})$  for the activated CFA-C is larger than that of the CFA-C raw materials for the tested pH of 0 and 1 (fig. S19C)。

对于溴，主要杂质包括铝、铁(>100 ppm)、硅、钙(10至100 ppm)、镁、镍、铬、铜(1至10 ppm)、锌(0.1至1 ppm)和钴(0.01至0.1 ppm)(图S20A)。

For BR, the major impurities include Al, Fe (>100 ppm), Si, Ca (10 to 100 ppm), Mg, Ni, Cr, Cu (1 to 10 ppm), Zn (0.1 to 1 ppm), and Co (0.01 to 0.1 ppm) (fig. S20A)。The ratio of  $c(\text{REE})/c(\text{Impurity})$  for the BR raw materials is larger than that of the activated BR (fig. S20B)。这意味着FJH还提高了杂质浸出能力，尤其是Fe(图S20A)。This means that the FJH also enhances the impurity leachability, especially Fe (fig. S20A)。对于e-waste，主要杂质包括Cu(>100 ppm)、Al、Si、Fe、Zn(10至100 ppm)、Ca、Mg、Ni(0.1至10 ppm)以及Co和Cr(0.1至1 ppm)(图S20C)。活化电子废物的 $c(\text{REE})/c(\text{Impurity})$ 比大于电子废物原料的 $c(\text{REE})/c(\text{Impurity})$ 。The ratio of  $c(\text{REE})/c(\text{Impurity})$  for the activated e-waste is larger than that of the e-waste raw materials (fig. S20D)。

上述杂质分析将有助于设计合适的下游稀土元素净化和分离工艺。The above impurity analysis would be helpful for the design of appropriate downstream REE purification and separation processing. The impurities content and the ratio of impurities to REE could be further optimized by adjusting the leaching conditions (e.g., pulp density, pH, acid type, temperature, time, etc.)。

讨论从浸出液中去除杂质的可能性。

#### Discussion on possible removal of impurities from the leach liquors.

许多技术，包括溶剂萃取、离子交换或吸附以及选择性沉淀，已广泛用于去除浸出液中的杂质(54)。

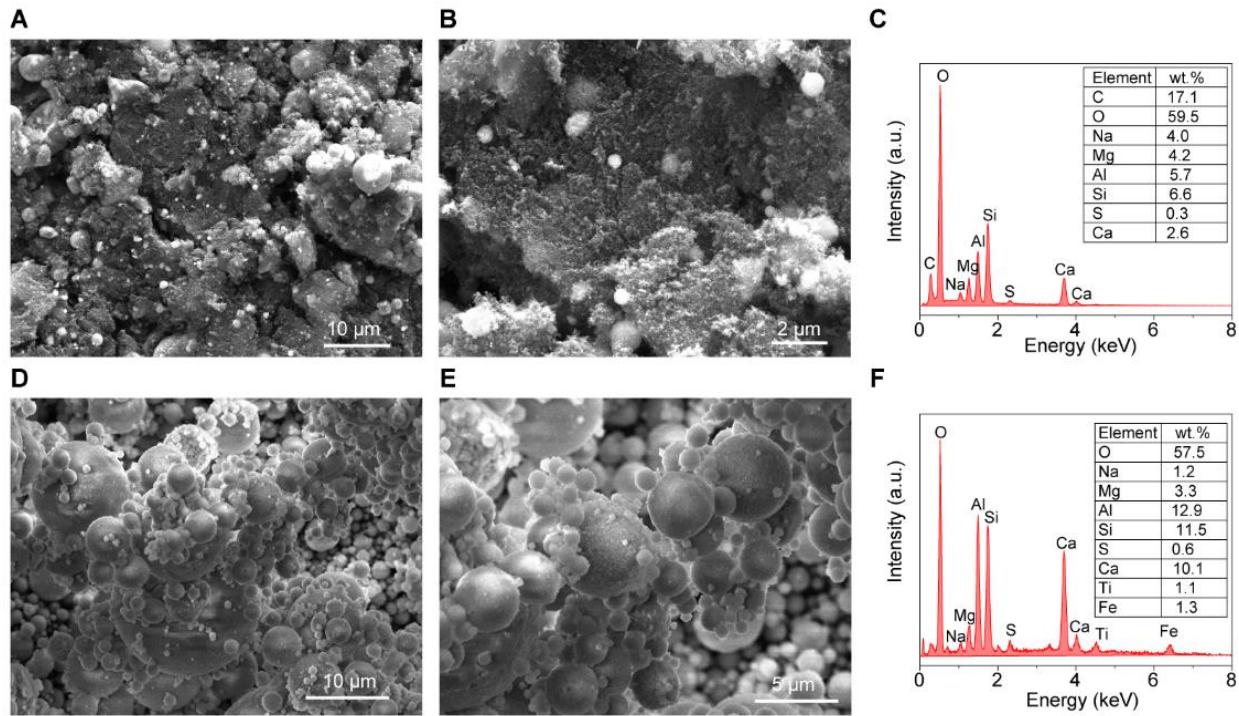
Many techniques, including solvent extraction, ion exchange or adsorption, and selective precipitation, have been widely used to remove the impurities in leach liquor (54)。The applicable route significantly depends on the impurities type and content, and the target application of REE. 在我们从CFA或BR获得的浸出液中，主要杂质为浓度>10 ppm的铝、硅、铁、镁和钙，而从e-waste获得的浸出液也应考虑铜和锌。In our leach liquor obtained from CFA or BR, the major impurities are Al, Si, Fe, Mg, and Ca with concentration >10 ppm, while Cu and Zn should also be considered for the leach liquor from e-waste. Many methods have already been widely used for removing these impurities (54)。例如，许多方法已广泛用于去除这些杂质(54)。For example, for the Fe-containing solution, acidic extractants such as di-(2-ethylhexyl)phosphoric

酸(D2EHPA)可以在适当的萃取剂浓度和有机/水比(56)下选择性地萃取稀土元素。acid (D2EHPA) can selectively extract REE with appropriate extractant concentration and organic/aqueous ratio (56). For Al impurity, a significant amount can be removed through selective precipitation by adjusting the pH of the leach liquor (57). Ca and Mg impurities usually do not co-extract with REE with the upper tolerance limit being 1500 ppm (58), which is much higher than the contents in our leach liquor (fig. S18 to 20). Thus, Ca and Mg are not particularly problematic in REE extraction. For Cu and Zn, they do not usually co-extract with REE during cation solvent extraction or ion exchange (58, 59).

#### 溶剂萃取分离稀土元素的探讨。 *Discussion on the REE separation by solvent extraction.*

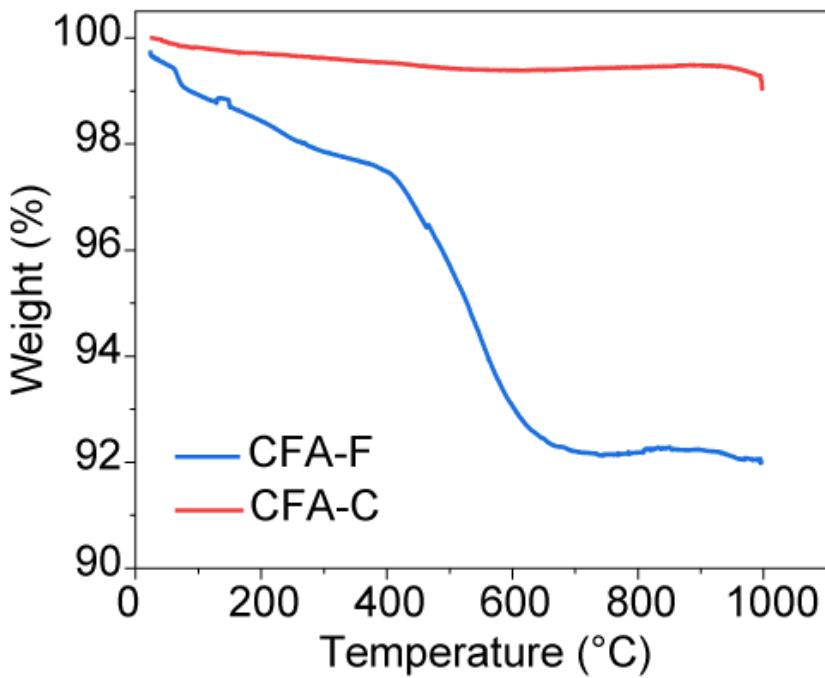
目前，溶剂萃取是最适合的稀土分离商业技术。  
Nowadays, solvent extraction is the most appropriate commercial technology for REE separation. The separation is generally done by primary separation and secondary separation. For the primary separation, D2EHPA is usually used to concentrate the REE from dilute solutions because of the high distribution coefficients. Subsequently, cation exchangers, solvation extractants, and anion exchangers are used to separate individual REE. Up to hundreds of stages of mixer and settler may be assembled to achieve the necessary separation and purity of REE, which have already been the routine scheme in many commercial REE extraction plants (51).

我们的含稀土元素的浸出液可以通过类似的程序获得单个稀土元素。  
Our REE-containing leachates could undergo similar procedures to get the individual REE. Moreover, the extraction of REE with cation exchangers is promoted by increasing the aqueous phase pH (51). Here, we realized the high REE leachability at a relatively high pH (e.g., pH 2 for CFA-F, and pH 1 for CFA-C), so the as-obtained leach liquors already have high pH. This could reduce the use of chemical agents to neutralize the leachate and be beneficial for the subsequent REE separation. Hence, existing separations technologies can be exploited to work with the REE extracts obtained through FJH. The mixtures obtained by FJH are often less cumbersome than those generated through the mining of ores, which represent another major benefit of the recycling scheme.



图S1。通过SEM和EDS对CFA的形态和成分进行表征。  
**Fig. S1. Morphology and composition characterization of CFA by SEM and EDS.** (A to B) CFA-F的SEM图像。 (C) CFA-F的EDS光谱。插图, CFA-F中主要元素的重量百分比。  
 SEM images of CFA-F. (C) EDS spectrum of CFA-F. Inset, the weight percentage of major elements in CFA-F. (D to E) CFA-C的SEM图像。 (F) CFA-C的EDS光谱。插图, CFA-C中主要元素的重量百分比。

SEM图像显示, CFA-C由粒径为几到几十米的球形颗粒组成。  
 The SEM images show that the CFA-C is composed of spherical particles with size of a few to tens of  $\mu\text{m}$ . In contrast, the size of CFA-F is smaller. The major elements in CFA-F are C, O, Na, Mg, Al, Si, S, and Ca. The major elements in CFA-C are O, Na, Mg, Al, Si, S, Ca, Ti, and Fe. The CFA-F has a high content of C while CFA-C has negligible C content. In contrast, the Ca content in CFA-C is significantly higher than that in CFA-F. Previous report showed that a higher content of Ca contributes to the higher acid extractability of REE (4)。

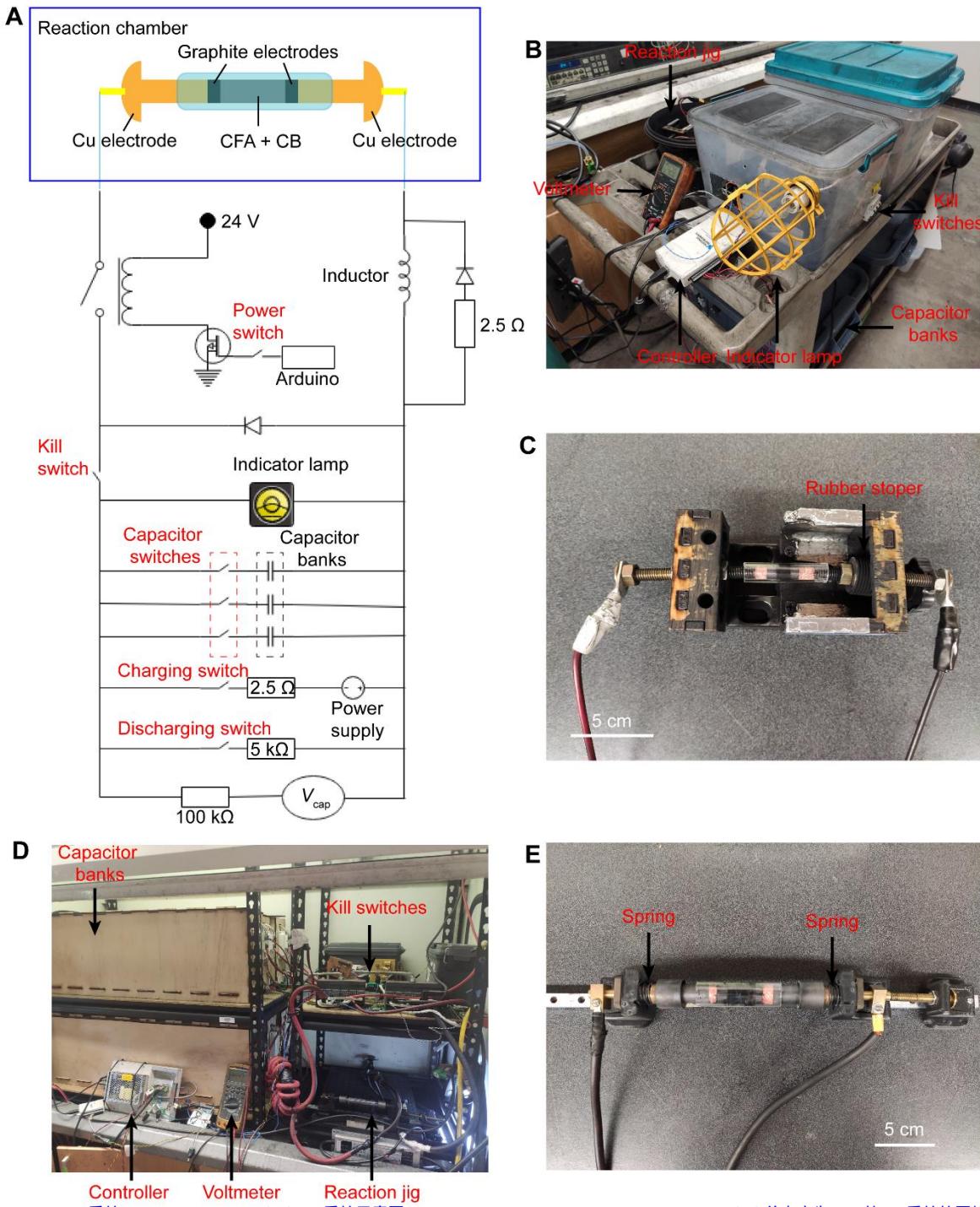


图S2。CFA在加热速率为 $10\text{ }^{\circ}\text{C min}^{-1}$ 的空气中的TGA曲线。

**Fig. S2. TGA curve of CFA in air with a heating rate of  $10\text{ }^{\circ}\text{C min}^{-1}$ .**

CFA-C的热重分析曲线显示，当温度上升到约 $700\text{ }^{\circ}\text{C}$ 时，重量损失约为8 wt%。

The TGA curve of CFA-C shows an obvious weight loss of ~8 wt% when the temperature rises to ~ $700\text{ }^{\circ}\text{C}$ . This is probably caused by the combustion of C, since there is a high C content in CFA-F according to the EDS analysis (fig. S1). In contrast, the CFA-F only shows a minor weight loss up to  $1000\text{ }^{\circ}\text{C}$ . In CFA, the major components are metal oxides, such as  $\text{CaO}$ ,  $\text{Fe}_2\text{O}_3$ , and  $\text{SiO}_2$ , which are stable in air up to  $1000\text{ }^{\circ}\text{C}$ .



**Fig. S3. The FJH system.** (A) FJH系统示意图。  
**(B)** 总电容为0.06F的FJH系统的图片。  
**(C)** 较小的FJH夹具连接样品和FJH系统，用于200 mg合成。  
**(D)** 图为总电容为0.624F的较大FJH系统。  
**(E)** 较大的FJH夹具用于连接样品和FJH系统进行2-g合成。  
在FJH过程中，(C)中的橡胶塞和(E)中的弹簧为样品提供逐渐压缩。  
The smaller FJH jigs to connect the sample and the FJH system for 200-mg synthesis.  
The larger FJH jigs to connect the sample and the FJH system for 2-g synthesis. The rubber stopper in (C) and the springs in (E) provide gradual compression to the sample during FJH.

## 电气部件:

### Electrical components:

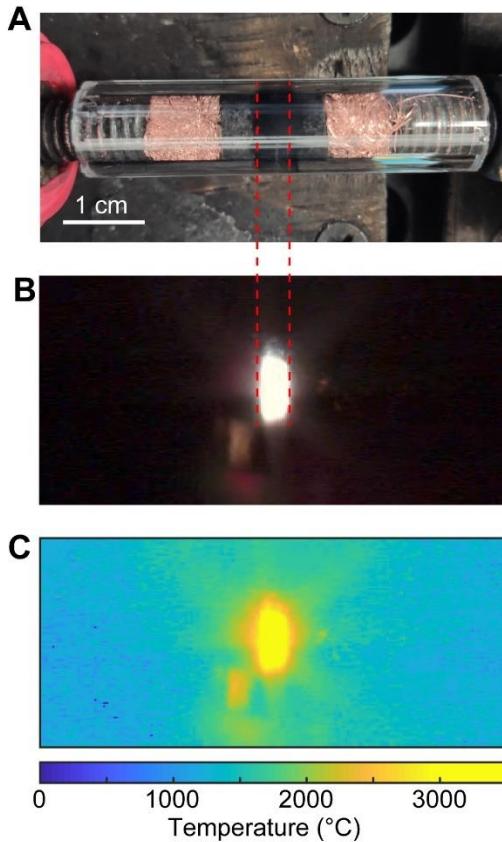
有关FJH系统中使用的电气部件的详细信息，请参阅我们之前的出版物(24)。  
The details of the electrical components used in the FJH system could be found in our previous publication (24). We also listed major of them here.

1. 电容器：铝电解电容器 (Mouser#80-PEH200YX460BQU2, 450 V, 6 mF)。  
 对于较小的FJH系统，电容器组由10个这样的电容器组成，总电容为0.06F。  
 该电容器组适用于批量为0.5g的反应。  
 对于较大的FJH系统，电容器组由104个此类电容器组成，总电容为0.624F。  
 该电容器组适用于批量达10g的反应。  
 1. Capacitors: Aluminum electrolytic capacitors (Mouser #80-PEH200YX460BQU2, 450 V, 6 mF). For the smaller FJH system, the capacitor bank is composed of 10 such capacitors with the total capacitance of 0.06 F. This capacitor bank is suitable for the reaction with batch size  $\leq 0.5$  g. For the larger FJH system, the capacitor bank is composed of 104 such capacitors with the total capacitance of 0.624 F. This capacitor bank is suitable for the reaction with batch size up to 10 g.
2. 机械继电器：900 V, 500 A (TE Connectivity LEV200A5ANA)。  
 电源：LED电源299.6 W 214-428 V 700 mA (Mouser #709-428 V 700 mA (鼠标#709-HLG320H-C700B))。
3. Power supply: LED Power Supplies 299.6 W 214–428 V 700 mA (Mouser #709-HLG320H-C700B)。
4.  $V_{cap}$ : 万用表福禄克189  
 充电和放电开关：400 V, 6 A断路器 (ABB S 282 K 6 A)
5. Charging and discharging switches: 400 V, 6 A breaker (ABB S 282 K 6 A)
6. 电容器开关：277 V, 10 A断路器 (ABB S201P-C10)
7. Kill switch: 440 V, 630 A断路器 (AAB S283 UC Z 63A)
8. 电感器：24 mH (鼠标#553-C-80U)
9. Controller: Arduino Uno with LCD display
10. Power supply: LED Power Supplies 299.6 W 214-428 V 700 mA (Mouser #709-HLG320H-C700B)

## 安全指南：

### Safety guidelines:

1. 封闭或小心绝缘电线连接。  
 所有连接和导线必须适用于高电压和高电流。
2. All connections and wires must be suitable for high voltages and currents.
3. 用户应遵守一只手规则：在系统上工作时只使用一只手，另一只手不得接触任何接地表面。
3. Users should obey the one hand rule: use only one hand when working on the system, with the other hand not touching any grounded surface.
4. 请记住，系统可以在毫秒内放电数千焦耳，这可能导致继电器等部件爆炸。  
 在电容器组上工作时，始终检查每个电容器组上的电压。
4. Keep in mind that the system can discharge thousands of Joules in milliseconds, which could cause components such as relays to explode.
5. 始终保持高压测试电压表可用。  
 在电容器组上工作时，始终检查每个电容器组上的电压。
5. Keep a voltmeter with high voltages test available at all times. When working on the capacitor banks, always check the voltage on each.
6. 使用仪器时，戴上厚厚的橡胶手套，一直戴到肘部，以防触电。
6. Wear thick rubber gloves that extend to the elbows when using the apparatus to protect from electrocution.
7. 系统的可靠性和鲁棒性应由经验丰富的电气技术人员确认。
7. The reliability and robustness of the system should be confirmed by an experienced electrical technician.



**图S4。FJH期间样品的温度图。**  
**Fig. S4. Temperature map of the sample during FJH.** (A) Optical image of the sample before FJH. (B) Optical image of the sample during FJH. (C) Temperature map of the sample during FJH. (A)之前样品的光学图像。  
(B) FJH期间样品的光学图像。  
(C) FJH期间样品的温度图。  
样品两侧的深色区域是石墨电极。  
The dark regions on each side of the sample are graphite electrodes.

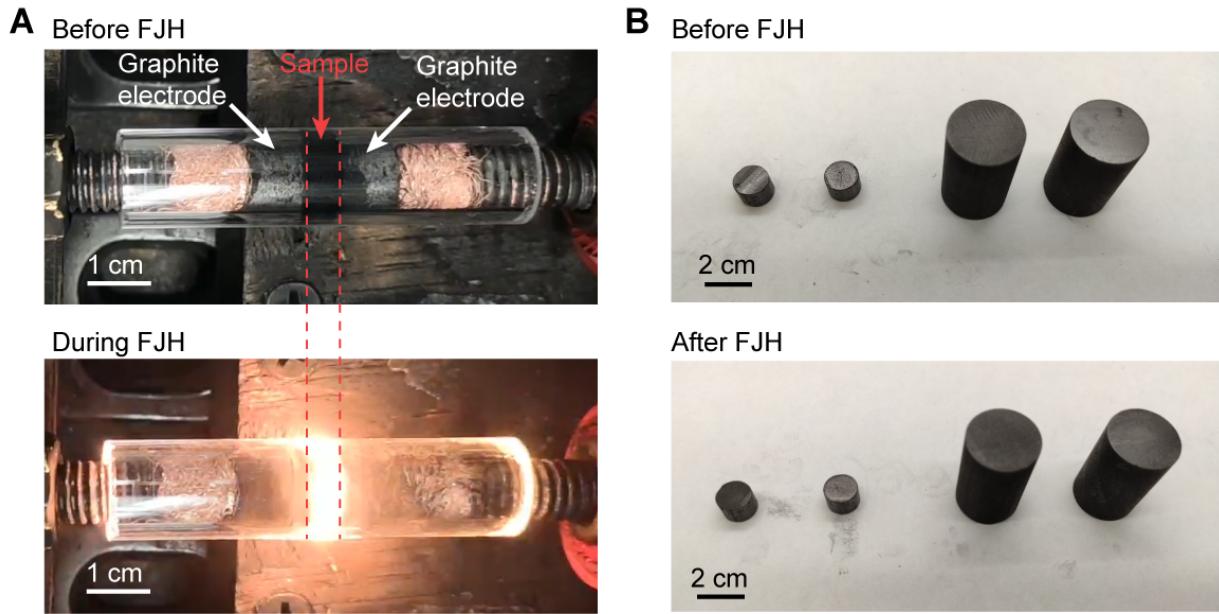
根据Stefan-Boltzmann定律，黑体辐射发射度( $j$ )与黑体热力学温度( $T$ )的四次方成正比，  
According to the Stefan-Boltzmann law, the blackbody radiant emittance ( $j$ ) is proportional to the  
fourth power of the blackbody's thermodynamic temperature ( $T$ )，

$$j = \sigma T^4 \quad (\text{S19})$$

其中 $\sigma$ 为比例常数。  
where  $\sigma$  is a constant of proportionality. Inversely, the temperature of a sample could be evaluated  
based on its radiant intensity. Experimentally, we first captured the optical image of a sample  
during FJH process using an ultrafast camera (fig. S4B). Then, the color image was converted to  
a grayscale image, which was further converted to an intensity matrix using MATLAB. The  
外温度计测量获得样品的最高温度( $T_{\max} \sim 2970^\circ\text{C}$ )(图2C)，其对应于强度矩阵中的最大值( $I_{\max}$ )。  
highest temperature ( $T_{\max} \sim 2970^\circ\text{C}$ ) of the sample was obtained according to the IR thermometer  
measurement (Fig. 2C), which corresponds to the largest value ( $I_{\max}$ ) in the intensity matrix. Hence,  
the temperature ( $T$ ) of each pixel of the image could be calculated using the intensity value ( $I$ ) in  
the intensity matrix, based on the Stefan-Boltzmann law,

$$\frac{I}{I_{\max}} = \left(\frac{T}{T_{\max}}\right)^4 \quad (\text{S20})$$

绘制了样品内的温度分布图(图S4C)。  
The temperature distribution within the sample was plotted (fig. S4C). It is found that the  
temperature is very uniform throughout the entire sample without obvious gradient. The whole  
sample could achieve a high temperature of  $\sim 3000^\circ\text{C}$ , demonstrating that the FJH process has a  
homogenous heating capability.



**图S5. FJH系统的耐久性。**  
**Fig. S5. Durability of the FJH system.** (A) Optical images of the sample before (top) and during (bottom) the FJH. (B) Optical images of the graphite electrodes before (top) and after (bottom) the FJH.

FJH装置具有良好的耐久性。

The FJH setup has good durability. A high temperature ( $\sim 3000^{\circ}\text{C}$ ) could be achieved by the FJH process, but the high-temperature region is only limited to the sample. According to the Joule heating equation,

$$Q=I^2Rt \quad (\text{S21})$$

where  $I$  is the current passing through the sample,  $R$  is the resistance of the sample, and  $t$  is the discharging time, the electrical heat is proportional to the resistance. The graphite electrodes have much smaller resistance than the sample. Hence, the heat generated by the discharging process is

如图S5A所示，在FJH过程中，样品区域的强发光受到限制，而石墨电极和FJH系统的其他部分保持在相对较低的温度。

因此，放电过程产生的热量施加在样品上。石墨电极具有良好的热稳定性。此外，FJH时间很短

温度为 $3000^{\circ}\text{C}$ ，时间为几十毫秒(图2C)。在FJH工

艺后，石墨棒没有显示出明显的变化(图S5B)，电阻保持不变。

因此，FJH工艺不会损坏电极。FJH系统的其他部分是远离高温样品的商用电气元件，如导线和电容器。

都是商业电气元件，如导线和电容器，远距离样品。在我们的实验中，相同的FJH装置被使用了数百次，没有降解。

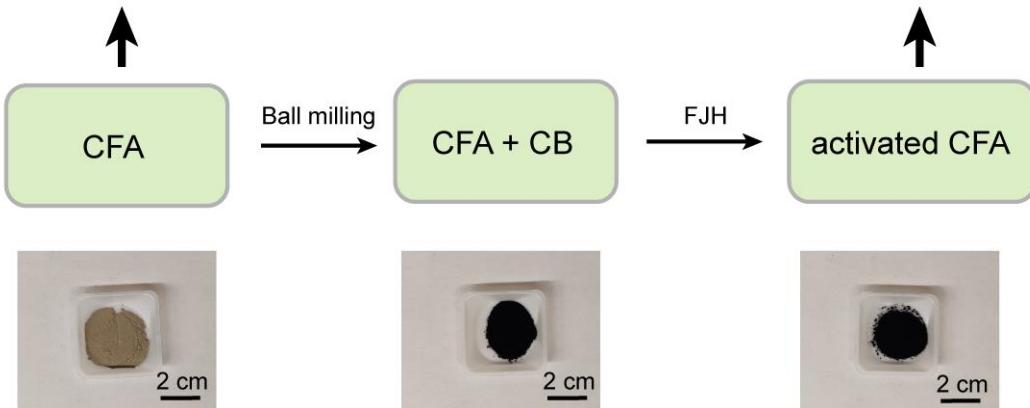
因此，我们得出结论，FJH系统具有很好的耐久性。

故此，FJH系统具有良好的耐久性。

$c_{\text{total}}(\text{CFA-Raw})$ : REE content by total quantification from CFA raw materials

$c_0(\text{CFA-Raw})$ : acid leachable REE content from CFA raw materials

$c(\text{activated CFA})$ : acid leachable REE content from CFA after FJH

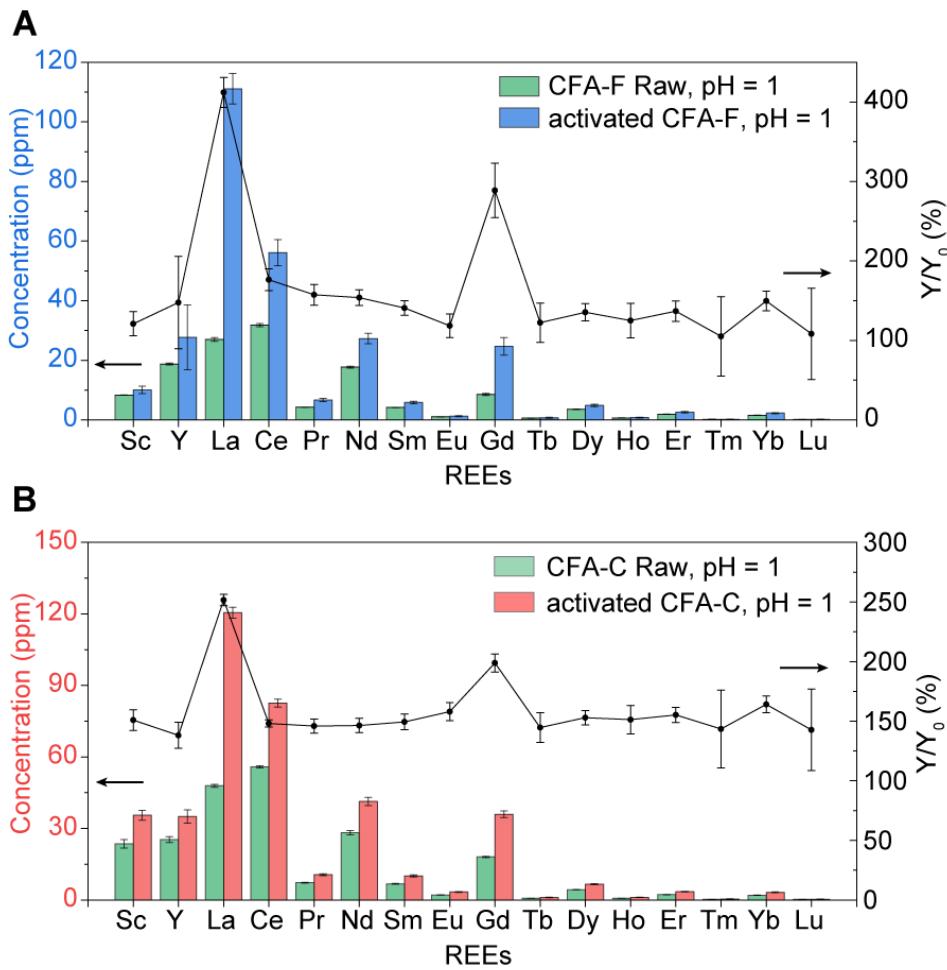


图S6。通过电热活化从二次废物中回收稀土元素的流程图。

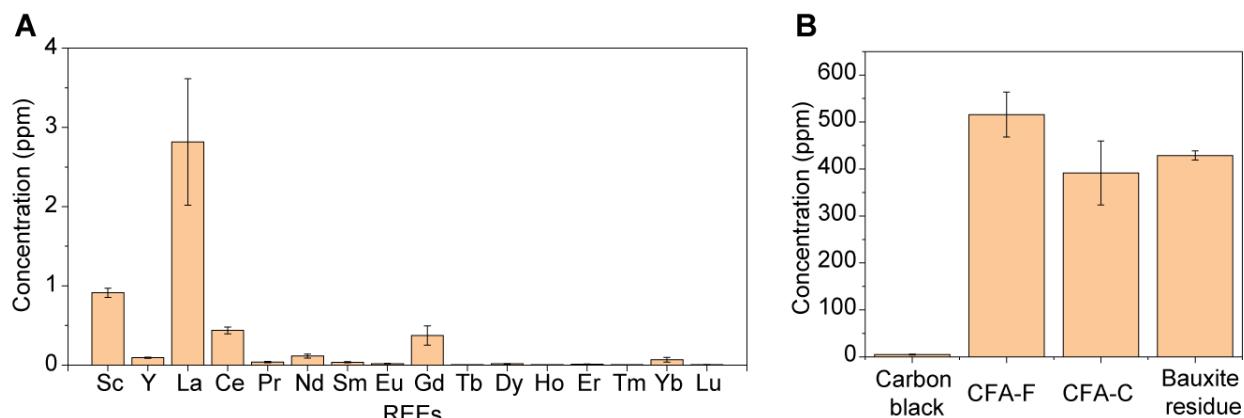
**Fig. S6. Flow chart of REE recovery from secondary wastes by electrothermal activation.**

使用HF-HNO<sub>3</sub>消解法通过全消解测量CFA原料 $c_{\text{total}}(\text{CFA raw})$ 中的总稀土含量(详见材料和方法)。  
The total REE content in CFA raw materials,  $c_{\text{total}}(\text{CFA-Raw})$ , was measured by total digestion using a HF-HNO<sub>3</sub> digestion method (see details in Materials and Methods). The acid-leachable REE contents in CFA raw materials,  $c_0(\text{CFA-Raw})$ , were measured by HCl or HNO<sub>3</sub> leaching of the CFA raw materials. The CFA raw materials and carbon black were mixed and underwent the FJH activation process. The obtained solid is termed as activated CFA. The acid-leachable REE contents in activated CFA,  $c(\text{activated CFA})$ , were measured by the same acid leaching procedure of the activated CFA. Then, the REE recovery yield by acid leaching of the CFA raw materials, and the activated CFA were calculated (Supplementary Text 1).

通过对CFA原料进行HCl或HNO<sub>3</sub>浸出，测定了CFA原料 $c_0(\text{CFA-Raw})$ 中可酸浸出的稀土元素含量。  
将CFA原料和炭黑混合并进行FJH活化过程。  
获得的固体称为活化CFA。  
采用与活化CFA相同的酸浸程序测量活化CFA中可酸浸的稀土元素含量 $c(\text{活化CFA})$ 。  
然后，通过酸浸CFA原料和活化CFA计算稀土回收率(补充文本1)。



**Fig. S7. REE leachability from CFA using 0.1 M HCl.** (A) Acid-leachable REE contents (0.1 M HCl, 85 °C) from CFA-F raw materials and the activated CFA-F, and the increase of recovery yield. (B) Acid-leachable REE contents (0.1 M HCl, 85 °C) from CFA-C raw materials and the activated CFA-C, and the increase of recovery yield.  $Y_0$  represents the REE recovery yield by 0.1 M HCl leaching the CFA raw materials, and  $Y$  represents the REE recovery yield by 0.1 M HCl leaching the activated CFA. For all the REE, the average values of  $Y/Y_0$  were >100%, indicating that the FJH process increased the recovery yields. The error bars in A and B represent the standard derivation where N = 3.

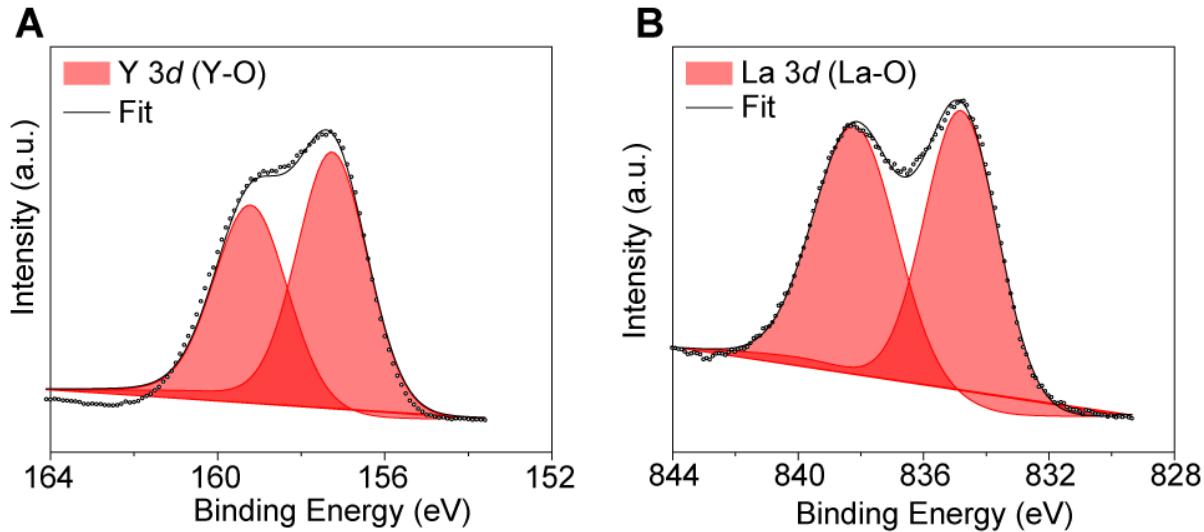


**Fig. S8. REE contents in carbon black.** (A) Individual REE content in carbon black. (B) Total REE content in carbon black, CFA-F, CFA-C, and bauxite residue (BR). All error bars in (A) and (B) represent the standard deviation where  $N = 3$ .

炭黑中稀土元素含量较低，约为CFA-F中稀土元素含量的1.0%，CFA-C中稀土元素含量的1.3%，BR中稀土元素含量的1.2%。因此，炭黑添加剂不会在我们的测量中产生显著误差。

此外，炭黑不是作为导电添加剂的唯一选择。在实际应用中，如果需要，炭黑可以用无烟煤替代，也可以用任何其他廉价的中等导电碳源替代。在我们的案例中，我们仅使用炭黑，因为其稀土元素含量较低，以排除导电添加剂引起的误差源。

In addition, carbon black is not the only choice as the conductive additive. In practical applications, the carbon black could be substituted with anthracite coal if desired, of any other inexpensive sources of mildly conductive carbon. In our cases, we used carbon black only since its REE content is low to exclude the error source induced by the conductive additive.



**Fig. S9. XPS characterization of REE oxides.** (A) XPS fine spectrum of Y in Y<sub>2</sub>O<sub>3</sub> precursor. (B) XPS fine spectrum of La in La<sub>2</sub>O<sub>3</sub> precursor.

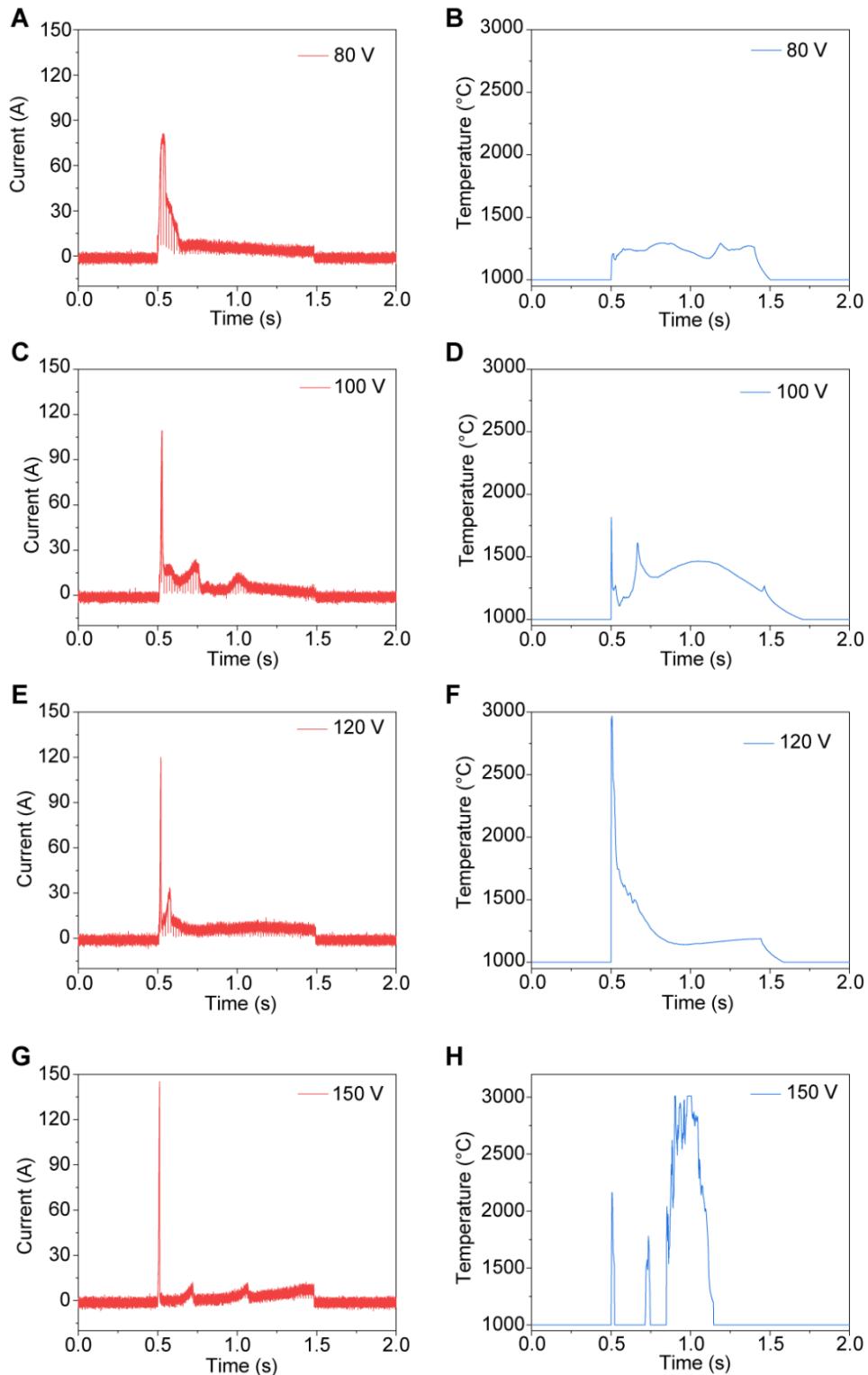
The detailed peak fittings for La<sub>2</sub>O<sub>3</sub>, La<sub>2</sub>O<sub>3</sub> after FJH, Y<sub>2</sub>O<sub>3</sub>, and Y<sub>2</sub>O<sub>3</sub> after FJH are shown in table S4. The Y 3d was split into 3d<sub>5/2</sub> and 3d<sub>3/2</sub>. In Y<sub>2</sub>O<sub>3</sub>, the peak positions for 3d<sub>5/2</sub> and 3d<sub>3/2</sub> 文献报道(35, 36)吻合良好。在Y<sub>2</sub>O<sub>3</sub>中，3d<sub>5/2</sub>和3d<sub>3/2</sub>的峰值位置分别为157.4 eV和159.4 eV，与文献报道(35, 36)吻合良好。 After FJH后，Y 3d由四个峰值拟合(图3E)。

The peaks at 157.4 eV and 159.4 eV, respectively, matching well with the literature reports (35, 36). After FJH, the Y 3d were fitted by four peaks (Fig. 3E). The peaks at 157.5 eV and 159.6 eV are assigned to 3d<sub>5/2</sub> and 3d<sub>3/2</sub> of Y in Y<sub>2</sub>O<sub>3</sub>, respectively. The peaks at 156.4 eV and 158.5 eV are assigned to Y金属中Y的3d<sub>5/2</sub>和3d<sub>3/2</sub>分别与文献报道(36)吻合良好。XPS分析 3d<sub>5/2</sub> and 3d<sub>3/2</sub> of Y in Y metal, respectively, matching well with the literature report (36). The 表明，在FJH过程中，Y<sub>2</sub>O<sub>3</sub>被还原为Y金属，而Y<sub>2</sub>O<sub>3</sub>的小比例可能来自表面氧化。

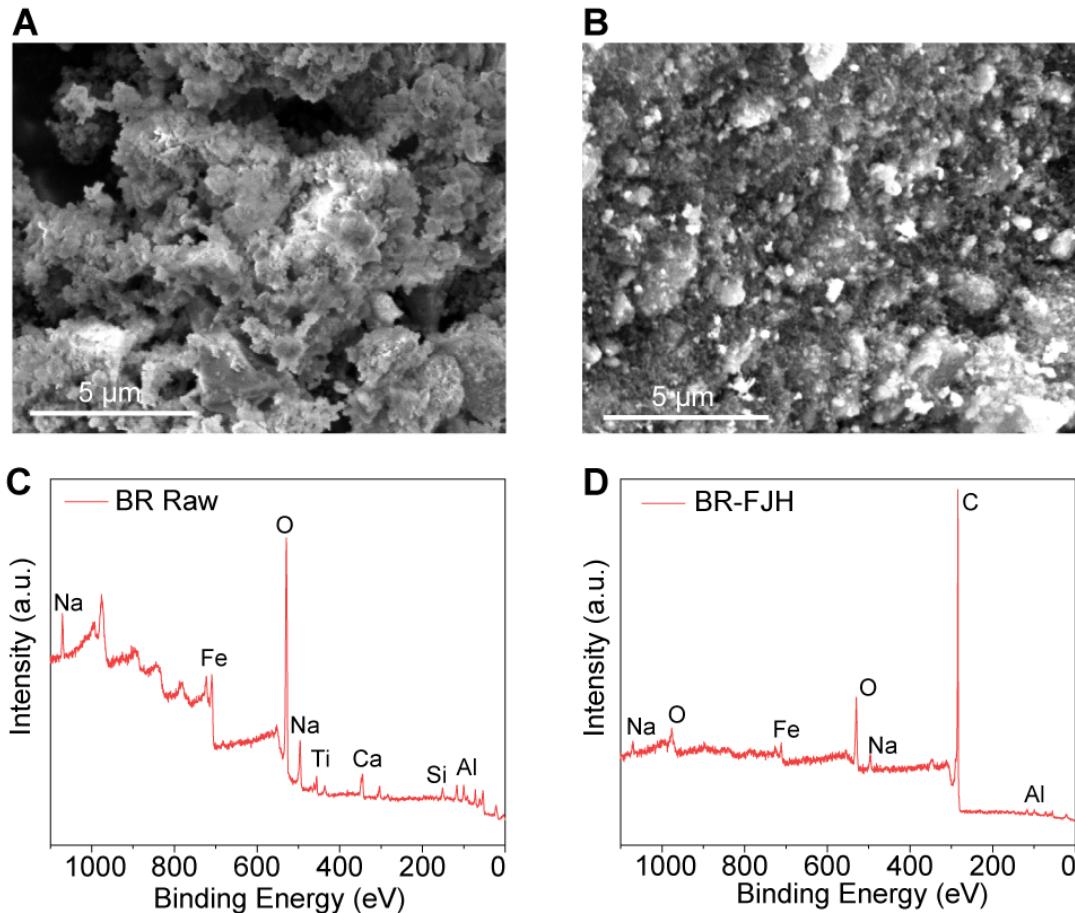
XPS analysis showed that the Y<sub>2</sub>O<sub>3</sub> is reduced to Y metal during the FJH process, while the small ratio of Y<sub>2</sub>O<sub>3</sub> might be from the surface oxidation.

对于La<sub>2</sub>O<sub>3</sub>，834.8 eV和838.2 eV处的峰值对应于3d<sub>5/2</sub>及其卫星峰值(38, 60)。

For La<sub>2</sub>O<sub>3</sub>, the peaks at 834.8 eV and 838.2 eV correspond to 3d<sub>5/2</sub> and its satellite peak (38, 60). After FJH, in addition to the peaks from La<sub>2</sub>O<sub>3</sub> at 834.8 eV and 838.1 eV (La 3d<sub>5/2</sub> and 839.6 eV时的主要成分)，the peak fitting (Fig. 3F, table S4) shows major components at 836.0 eV and 839.6 eV, which are assigned to La 3d<sub>5/2</sub> and its satellite peak from La metal, matching well with literature reports (37, 61)。XPS分析表明，在FJH过程中，La<sub>2</sub>O<sub>3</sub>被还原为La，而较小比例的La<sub>2</sub>O<sub>3</sub>可能来自表面氧化。

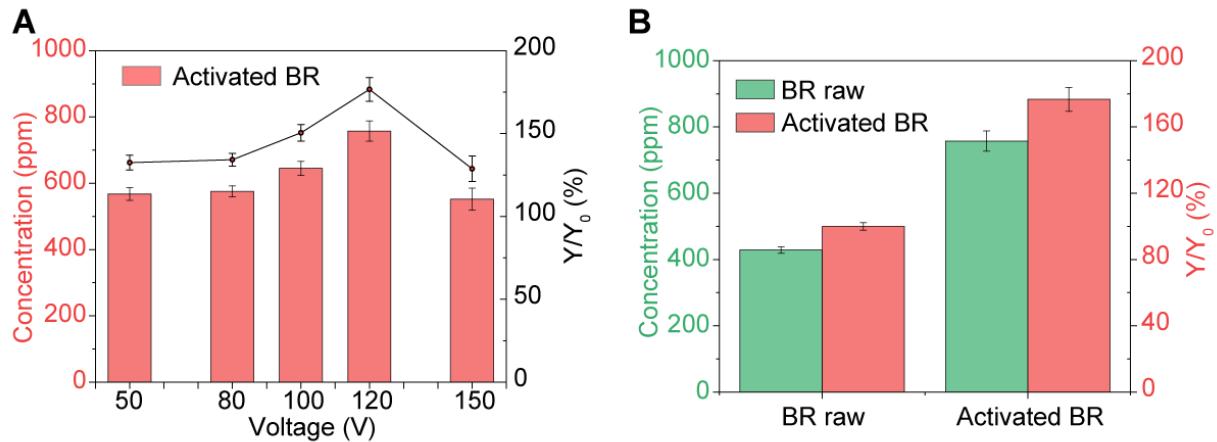


图S10。温度测量。  
**Fig. S10. Temperature measurement.** Current curves and real-time temperature curves for (A to B) 80 V for 1 s, (C to D) 100 V for 1 s, (E to F) 120 V for 1 s, and (G to H) 150 V for 1 s.



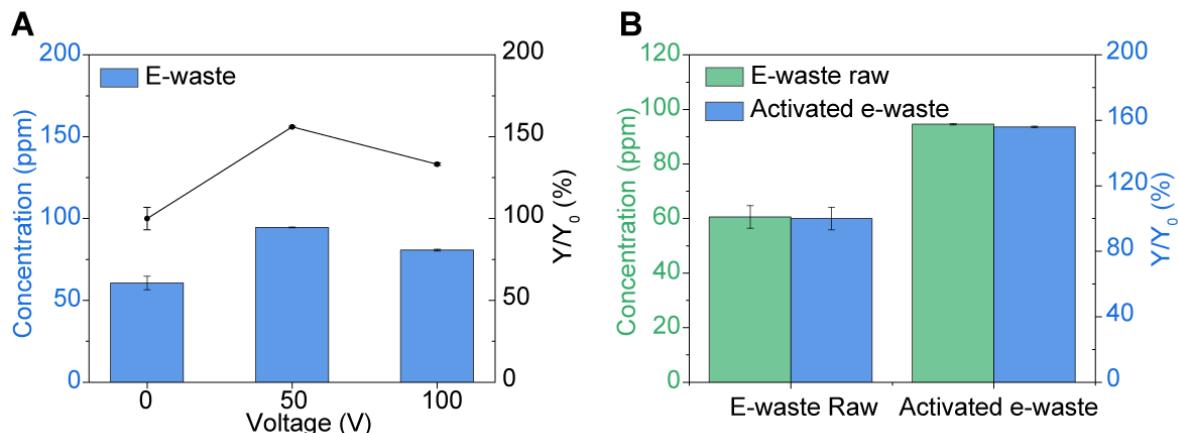
**图S11。丁苯橡胶和丁苯橡胶经FJH后的表征。**  
**Fig. S11. Characterization of BR and BR after FJH.** (A) SEM image of BR raw materials. (B) SEM image of BR after FJH. (C) XPS spectrum of BR raw materials. (D) XPS spectrum of BR after FJH.

经过FJH后，BR(图S11B)的粒径小于BR原料(图S11A)，这将有助于酸浸。  
 After FJH, the particle size of the BR (fig. S11B) becomes smaller than the BR raw materials (fig. S11A), which would be helpful for the acid leaching. According to the XPS, after FJH, the O content in BR is significantly reduced, demonstrating the carbothermic reduction of the metal components in BR. This would also contribute to the acid leaching of REE for the same reasons we have explained for the recovery of REE from CFA in the main text (Fig. 3).  
 根据XPS，经过FJH后，BR中的O含量显著降低。  
 这也有助于稀土元素的酸浸，原因与我们在正文中解释的从CFA中回收稀土元素的原因相同(图3)。



图S12. BR的FJH电压相关稀土元素回收率。

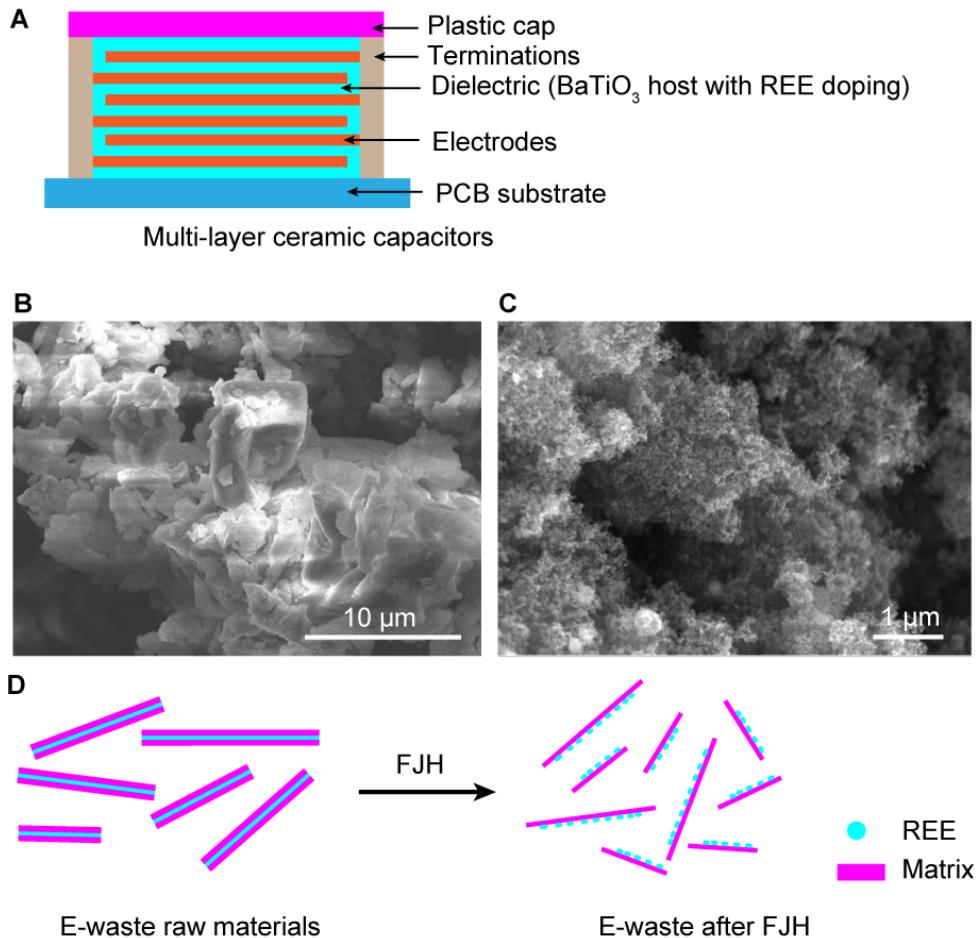
**Fig. S12. FJH voltage dependent REE recovery yield from BR.** (A) Acid leachable content of BR中的总稀土元素(0.5 M HNO<sub>3</sub>)，稀土产率的增加随FJH电压的变化而变化。  
total REE (0.5 M HNO<sub>3</sub>) from BR, and the increase of REE yield varied with FJH voltages. (B) 在120 V FJH下，可酸浸REE含量(0.5 M HNO<sub>3</sub>)和回收率的增加。The acid leachable REE content (0.5 M HNO<sub>3</sub>)，and the increase of recovery yield at 120 V FJH.  
 $Y_0$ 表示直接浸出BR原料的稀土回收率。  
 $Y$ 表示通过浸出活化溴得到的REE回收率。  
 $Y_0$  represents the REE recovery yield by directly leaching the BR raw materials.  $Y$  represents the REE recovery yield by leaching the activated BR. The error bar in A and B denotes the standard deviation where N = 3.



图S13. 通过FJH活化提高电子垃圾中稀土元素的回收率。

**Fig. S13. Improving the REE recovery yield from e-waste by FJH activation.** (A) Acid-leachable content of total REE (1 M HCl) from e-waste, and the increase of REE recovery yield varied with the FJH voltages. (B) The acid leachable content of total REE (1 M HCl), and the increase of REE recovery yield at 50 V FJH.  $Y_0$  represents the REE recovery yield by directly leaching the e-waste raw materials.  $Y$  represents the REE recovery yield by leaching the activated e-waste. The error bars in A and B denote the standard deviation where N = 3.

从电子垃圾中回收稀土元素的最佳FJH电压为50V。电压过高会导致温度过高，可能导致稀土元素的蒸发损失。  
The optimized FJH voltage for recovering REE from e-waste is 50 V. Too high voltage, which leads to too high temperature, might result in the evaporative loss of REE. The acid leachable content of total REE from the activated e-waste is ~95 ppm, corresponding to ~156% of the leachable REE content from e-waste raw materials.



图S14。FJH活化提高电子垃圾中稀土回收率的机理。

**Fig. S14. Mechanism of the improved REE recovery yield in e-waste by FJH activation.** (A)

多层陶瓷电容器的结构方案。  
(B) 电子垃圾破碎后的SEM图像。

Structure of the multi-layer ceramic capacitors. (B) SEM image of the e-waste after

(C) 电子垃圾经过FJH后的SEM图像。

(C) SEM image of the e-waste after FJH.

(D) FJH过程中电子垃圾的形态变化方案。

(D) Scheme of the morphology change of the e-waste during the FJH process.

稀土元素广泛应用于现代电子产品中，如强磁体、陶瓷电容器等。

The REE are widely used in modern electronics as strong magnets, ceramic capacitors, etc. 大多数电子设备具有层压结构，例如多层陶瓷电容器(44)(图S14A)，REE被塑料和陶瓷的隔离层和保护层覆盖。Most electronic devices have the laminated structures, e.g., the multilayer ceramic capacitors (44)

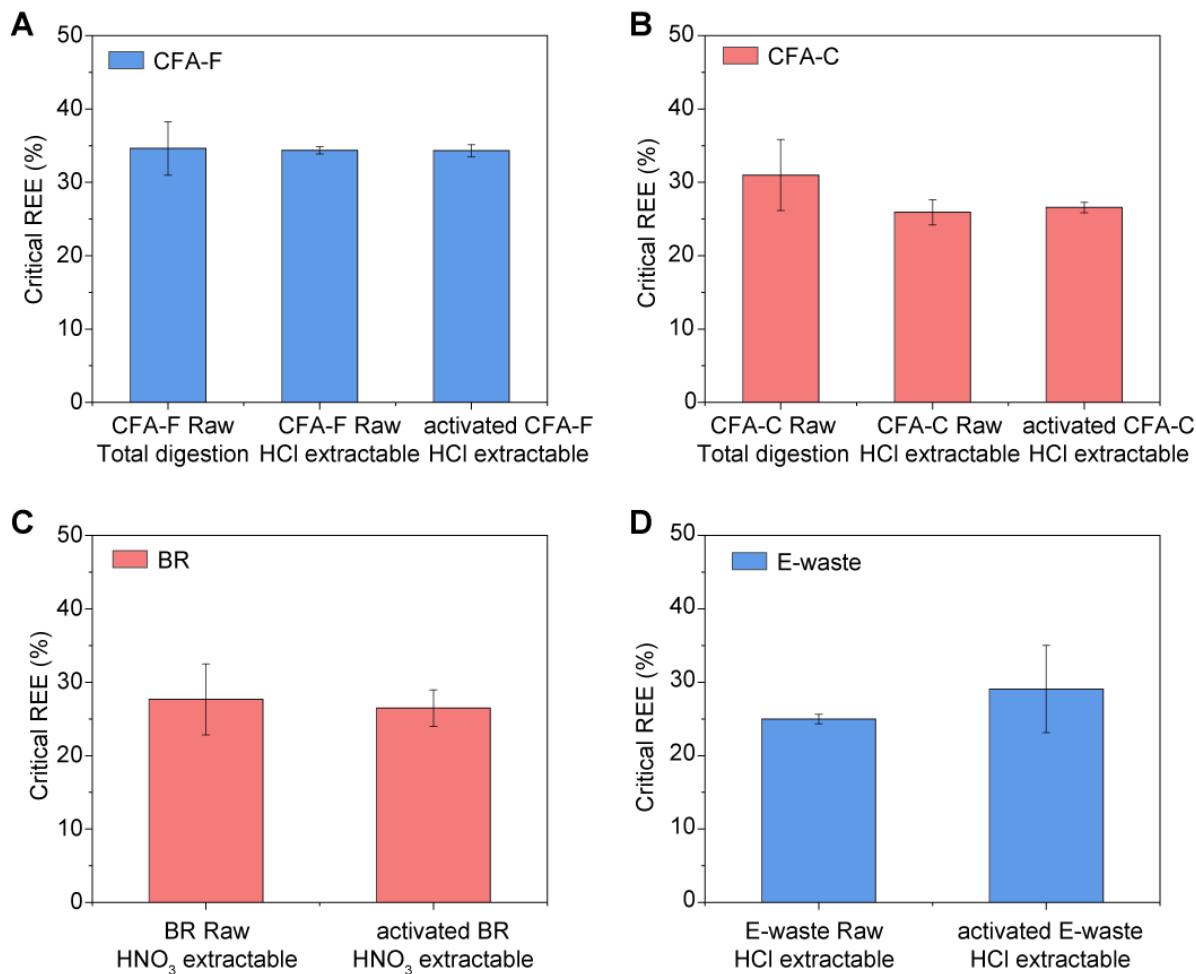
(fig. S14A), and the REE are covered by the separator and protection layer of plastics and ceramics. 层状结构通过防止稀土物种暴露于浸出剂而使稀土元素浸出变得困难。

The laminated structure makes it difficult for REE leaching by preventing the exposure of REE species to leachant. 即使在研磨后，电子垃圾的粒径也很大(图S14B)。

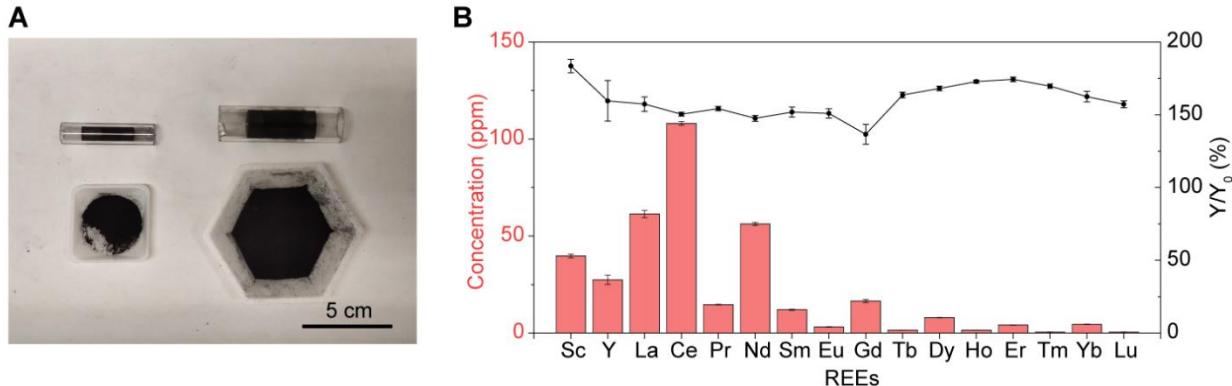
Even after grinding, the particle size of the e-waste is large (fig. S14B). After 在FJH工

艺后，层状结构被破坏(图S14C至D)，这提供了可酸浸的稀土元素。

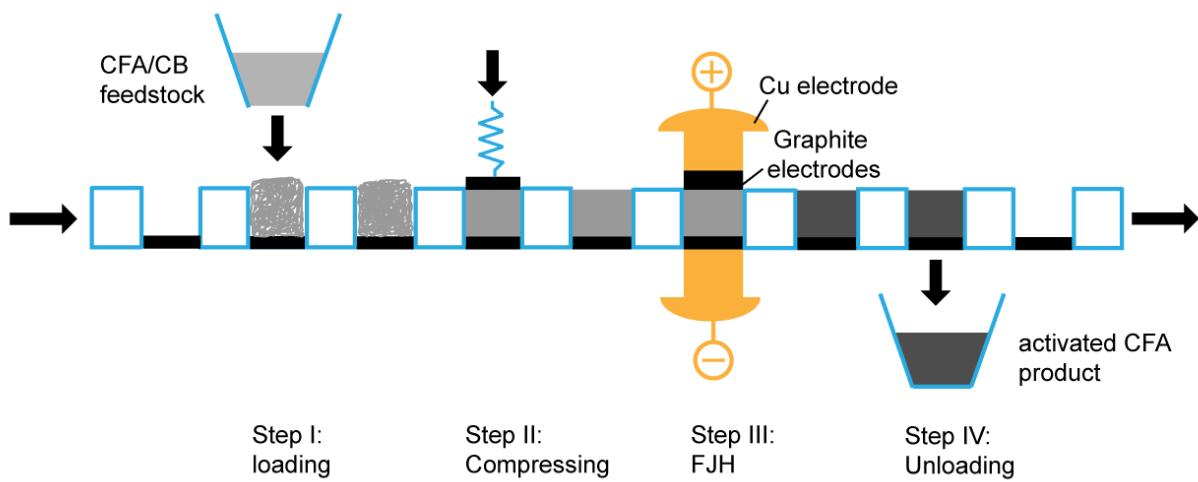
After the FJH process, the laminated structures are broken (fig. S14C to D), which affords the acid leachable REE.



图S15。临界稀土元素在总稀土元素中的百分比。  
**Fig. S15. The percentage of critical REE in total REE.** (A) Percentage of critical REE in CFA-F, including the total digestion, HCl (1 M, 85 °C) extractable REE content in CFA-F Raw materials, and HCl extractable REE content in activated CFA-F. (B) Percentage of critical REE in CFA-C, including the total digestion, HCl (1 M, 85 °C) extractable REE content in CFA-C Raw materials, and HCl extractable REE content in activated CFA-C. (C) Percentage of critical REE in BR, including the HNO<sub>3</sub> (0.5 M, RT) extractable REE content in BR Raw materials, and HNO<sub>3</sub> extractable REE content in activated BR. (D) Percentage of critical REE in e-waste, including the HCl (1 M, 85 °C) extractable REE content in e-waste Raw materials, and HCl extractable REE content in activated e-waste. The critical REE include Y, Nd, Eu, Tb, and Dy. All error bars in (A) to (D) denote standard deviation where N = 3.

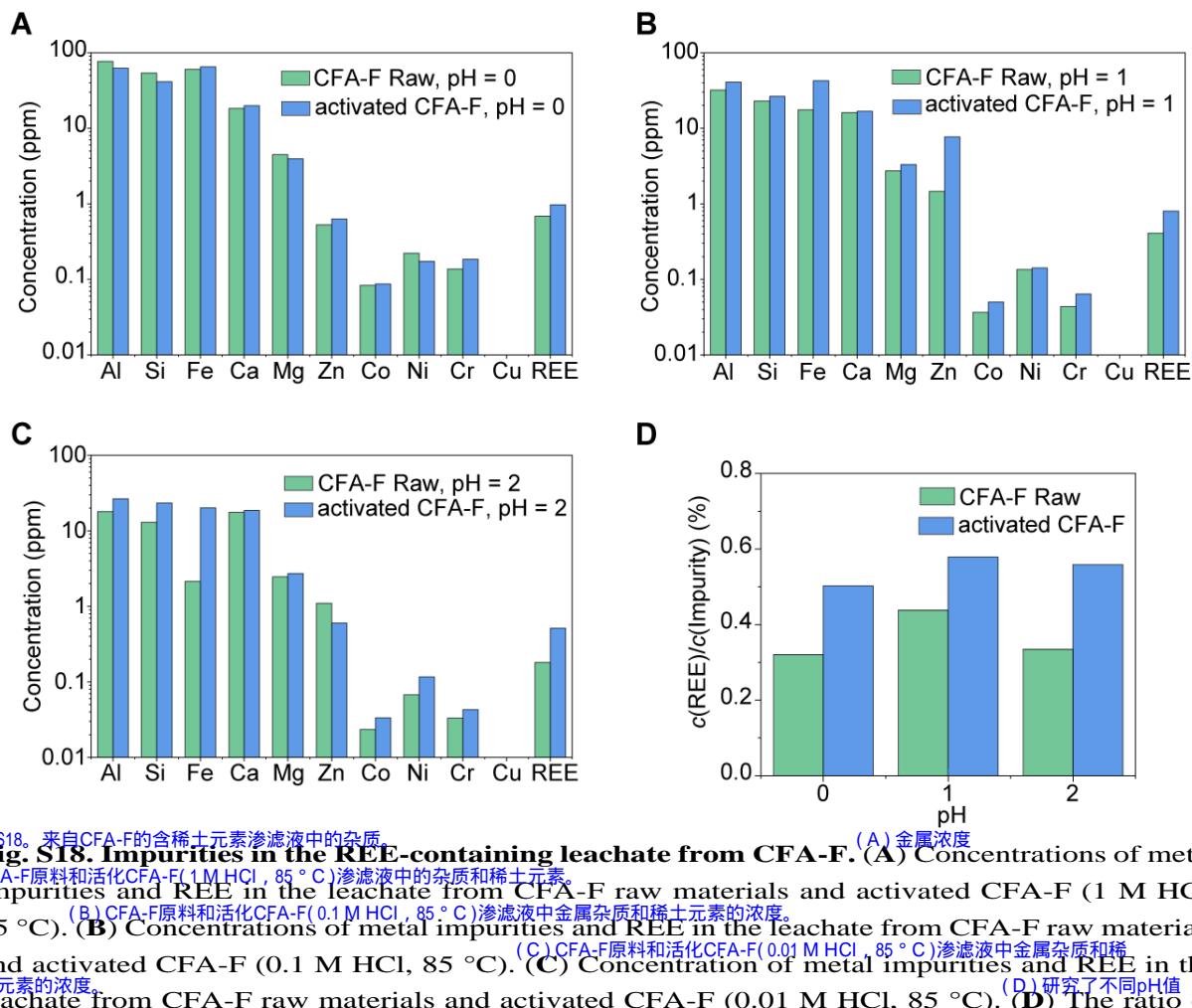


**Fig. S16. Scaling up of the electrothermal activation process by using CFA-C as an example.**  
 (A) 质量为 $m_0=200$  mg, 石英管直径为 $D_0=8$  mm(左)的小样品和质量为 $m_1=2$  g, 管直径为 $D_1=16$  mm(右)的大样品的照片。  
 (A) Picture of the small sample with mass of  $m_0 = 200$  mg and quartz tube diameter of  $D_0 = 8$  mm  
 (left), and larger sample with mass of  $m_1 = 2$  g and tube diameter of  $D_1 = 16$  mm (right). (B) Acid  
 量为 $m_1=2$  g的活化CFA-C中, 可酸浸出的稀土元素含量( $1\text{ M HCl}, 85^\circ\text{C}$ )和回收率的增加。  
 $Y_0$ 表示直接浸出CFA-C原料的稀土回收率。  
 $Y$ 表示通过浸出活化CFA-C获得的稀土回收率。  
 (B) 在质  
 量为 $m_1=2$  g的活化CFA-C中, 可酸浸出的稀土元素含量( $1\text{ M HCl}, 85^\circ\text{C}$ )和回收率的增加。  
 $Y_0$ 表示直接浸出CFA-C原料的稀土回收率。  
 $Y$ 表示通过浸出活化CFA-C获得的稀土回收率。  
 (B) 中的误  
 差线表示标准偏差, 其中 $N=3$ 。  
 bars in (B) denote the standard deviation where  $N = 3$ .



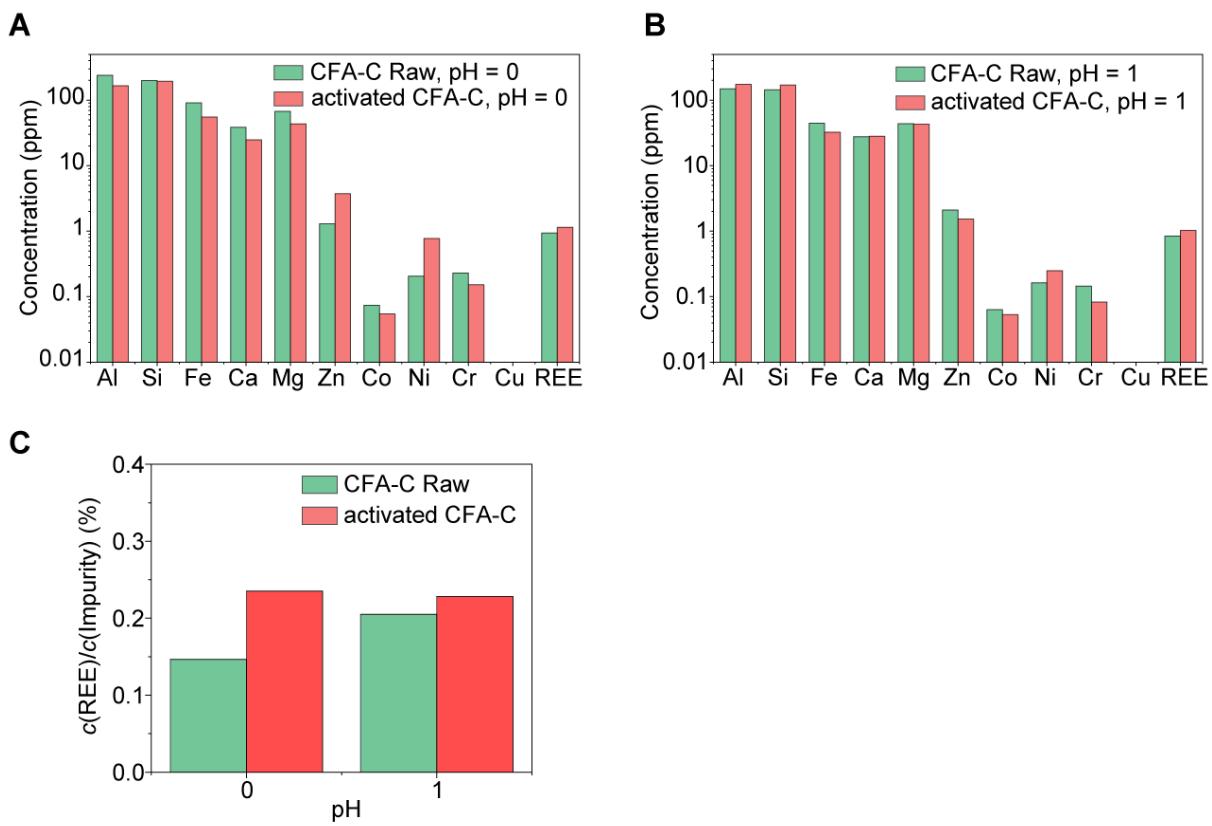
**图S17。连续FJH反应器的概念样机设计。**  
**Fig. S17. Conceptual prototype design of the continuous FJH reactor.**

连续生产过程包括四个步骤。  
 The continuous production process consists of four steps. First, the mixture of CFA/CB feedstock is loaded onto the chamber on the conveyor belt. Secondly, the sample is compressed to a specific resistance. Thirdly, the sample undergoes the FJH reaction. Lastly, the activated CFA product is collected.



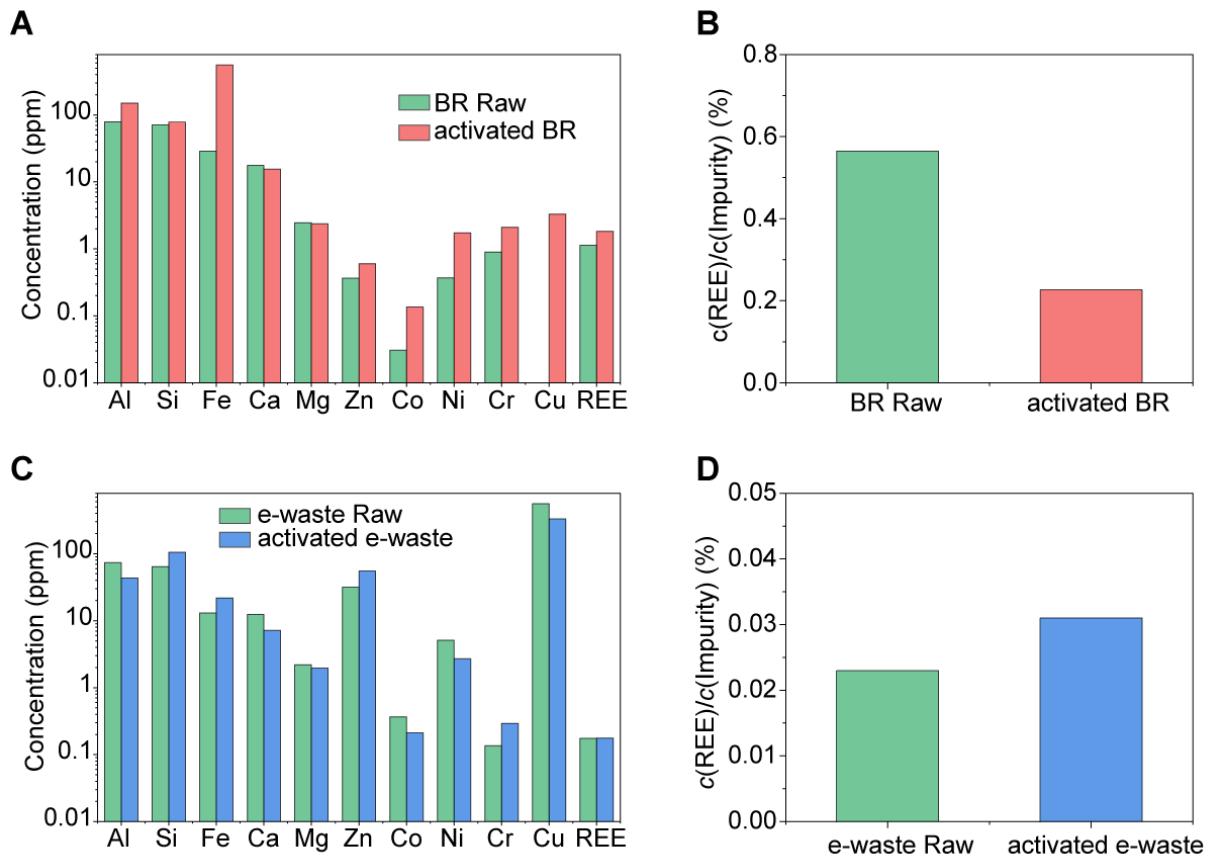
图S18。来自CFA-F的含稀土元素渗滤液中的杂质。

**Fig. S18. Impurities in the REE-containing leachate from CFA-F.** (A) Concentrations of metal impurities and REE in the leachate from CFA-F raw materials and activated CFA-F (1 M HCl, 85 °C). (B) Concentrations of metal impurities and REE in the leachate from CFA-F raw materials and activated CFA-F (0.1 M HCl, 85 °C). (C) Concentration of metal impurities and REE in the leachate from CFA-F raw materials and activated CFA-F (0.01 M HCl, 85 °C). (D) The ratio of REE content and metal impurities content in the leachate from CFA-F raw materials and activated CFA-F in leachant with different pH.



图S19。来自CFA-C的含稀土元素渗滤液中的杂质。  
**Fig. S19. Impurities in the REE-containing leachate from CFA-C.** (A) Concentrations of metal impurities and REE in the leachate from CFA-C raw materials and activated CFA-C (1 M HCl, 85 °C). (B) Concentrations of metal impurities and REE in the leachate from CFA-F raw materials and activated CFA-C (0.1 M HCl, 85 °C). (C) 研究了不同pH值的CFA-C原料渗滤液和活化CFA-C渗滤液中稀土元素含量和杂质含量的比值。

(A) 金属浓度  
(C) 研究了不同pH值的CFA-C原料渗滤液和活化CFA-C渗滤液中稀土元素含量和杂质含量的比值。



圖S20。BR和電子垃圾中含有稀土元素的滲濾液中的杂质。  
**Fig. S20. Impurities in the REE-containing leachate from BR and e-waste.** (A) Concentrations of metal impurities and REE in the leachate from BR raw materials and activated BR ( $0.5 \text{ M HNO}_3$ ,  $85^\circ\text{C}$ ). (B) The ratio of REE content and impurities content in the leachate from BR raw materials and activated BR. (C) Concentrations of metal impurities and REE in the leachate from e-waste raw materials and activated e-waste ( $1 \text{ M HCl}$ ,  $85^\circ\text{C}$ ). (D) The ratio of REE content and impurities content in the leachate from e-waste raw materials and activated e-waste.

表S1。二次废物活化的FJH参数。

**Table S1. The FJH parameters for activation of secondary wastes.**

Precursors	Mass 比率 Ratio	大 量 Mass (mg)	反 对 Resistance (Ω)	电 压 Voltage (V)	时 间 Time (s)	FJH后的质量 Mass after FJH (mg)
CFA-F:CB	2:1	200	1.0	50	1	190
CFA-F:CB	2:1	200	1.0	80	1	138
CFA-F:CB	2:1	200	1.0	100	1	115
CFA-F:CB	2:1	200	1.0	120	1	157
CFA-F:CB	2:1	200	1.0	150	1	88
CFA-C:CB	2:1	200	1.0	120	1	182
BR:CB	2:1	200	1.2	50	1	190
BR:CB	2:1	200	1.2	80	1	174
BR:CB	2:1	200	1.2	100	1	162
BR:CB	2:1	200	1.2	120	1	159
BR:CB	2:1	200	1.2	150	1	152
PCB:CB	2:1	200	1.0	50	1	162
PCB:CB	2:1	200	2.0	100	1	185
PCB:CB	2:1	200	2.0	120	1	98

表S2。代表性稀土磷酸盐的热分解温度。

**Table S2. Thermal decomposition temperature of representative REE phosphates.**

材料反应 Material	Reaction	H( kJ mol <sup>-1</sup> ) $\Delta H$ (kJ mol <sup>-1</sup> )	S( J mol <sup>-1</sup> K <sup>-1</sup> ) $\Delta S$ (J mol <sup>-1</sup> K <sup>-1</sup> )	Temp (°C)
LaPO <sub>4</sub>	LaPO <sub>4</sub> = 1/2La <sub>2</sub> O <sub>3</sub> + PO <sub>2</sub> + 1/4O <sub>2</sub>	817.65	260.45	2866
CePO <sub>4</sub>	CePO <sub>4</sub> = 1/2Ce <sub>2</sub> O <sub>3</sub> + PO <sub>2</sub> + 1/4O <sub>2</sub>	785.8	256.3	2793
PrPO <sub>4</sub>	PrPO <sub>4</sub> = 1/2Pr <sub>2</sub> O <sub>3</sub> + PO <sub>2</sub> + 1/4O <sub>2</sub>	798.8	269.8	2688
NdPO <sub>4</sub>	NdPO <sub>4</sub> = 1/2Nd <sub>2</sub> O <sub>3</sub> + PO <sub>2</sub> + 1/4O <sub>2</sub>	780.85	262.1	2706
SmPO <sub>4</sub>	SmPO <sub>4</sub> = 1/2Sm <sub>2</sub> O <sub>3</sub> + PO <sub>2</sub> + 1/4O <sub>2</sub>	774.3	256.5	2746
EuPO <sub>4</sub>	EuPO <sub>4</sub> = 1/2Eu <sub>2</sub> O <sub>3</sub> + PO <sub>2</sub> + 1/4O <sub>2</sub>	765.0	266.7	2595
GdPO <sub>4</sub>	GdPO <sub>4</sub> = 1/2Gd <sub>2</sub> O <sub>3</sub> + PO <sub>2</sub> + 1/4O <sub>2</sub>	766.4	253.9	2745

注：REEPO<sub>4</sub>=1/2REE<sub>2</sub>O<sub>3</sub>+PO<sub>2</sub>+1/4O<sub>2</sub>的分解反应用于所有稀土磷酸盐(30)。

Note: The decomposition reaction, REEPO<sub>4</sub> = 1/2REE<sub>2</sub>O<sub>3</sub> + PO<sub>2</sub> + 1/4O<sub>2</sub>, is used for all REE phosphates (30). The standard molar enthalpies and standard molar entropies of REE phosphates (30) are from the literature (62), and these constants of REE oxides, PO<sub>2</sub>, and O<sub>2</sub> are from the CRC Handbook of Physics and Chemistry (63).

表S3。25 °C下稀土金属、氧化物和磷酸盐溶解反应的吉布斯自由能变化和溶解度积常数( $K_{sp}$ )。  
**Table S3. Gibbs free energy change and solubility product constants ( $K_{sp}$ ) of REE metals, oxides, and phosphates dissolution reactions at 25 °C.**

材料反应 Materials	Reaction	$\Delta G$	$\log_{10}K_{sp}$
Sc	$Sc + 3H^+ = Sc^{3+} + 3/2H_2$	-614.2 kJ mol <sup>-1</sup>	107.61
Sc <sub>2</sub> O <sub>3</sub>	$1/2Sc_2O_3 + 3H^+ = Sc^{3+} + 3/2H_2O$	-32.61 kJ mol <sup>-1</sup>	5.71
ScPO <sub>4</sub>	$ScPO_4 = Sc^{3+} + PO_4^{3-}$	---	-26.96
Y	$Y + 3H^+ = Y^{3+} + 3/2H_2$	-693.8 kJ mol <sup>-1</sup>	121.56
Y <sub>2</sub> O <sub>3</sub>	$1/2Y_2O_3 + 3H^+ = Y^{3+} + 3/2H_2O$	-141.19 kJ mol <sup>-1</sup>	24.74
YPO <sub>4</sub>	$YPO_4 = Y^{3+} + PO_4^{3-}$	---	-25.02
La	$La + 3H^+ = La^{3+} + 3/2H_2$	-683.7 kJ mol <sup>-1</sup>	119.79
La <sub>2</sub> O <sub>3</sub>	$1/2La_2O_3 + 3H^+ = La^{3+} + 3/2H_2O$	-186.49 kJ mol <sup>-1</sup>	32.67
LaPO <sub>4</sub>	$LaPO_4 = La^{3+} + PO_4^{3-}$	---	-25.7
Ce	$Ce + 3H^+ = Ce^{3+} + 3/2H_2$	-672 kJ mol <sup>-1</sup>	117.74
Ce <sub>2</sub> O <sub>3</sub>	$1/2Ce_2O_3 + 3H^+ = Ce^{3+} + 3/2H_2O$	-174.59 kJ mol <sup>-1</sup>	30.59
CePO <sub>4</sub>	$CePO_4 = Ce^{3+} + PO_4^{3-}$	---	-26.2
Pr	$Pr + 3H^+ = Pr^{3+} + 3/2H_2$	-679.1 kJ mol <sup>-1</sup>	118.98
Pr <sub>2</sub> O <sub>3</sub>	$1/2Pr_2O_3 + 3H^+ = Pr^{3+} + 3/2H_2O$	-174.89 kJ mol <sup>-1</sup>	30.64
PrPO <sub>4</sub>	$PrPO_4 = Pr^{3+} + PO_4^{3-}$	---	-26.4
Nd	$Nd + 3H^+ = Nd^{3+} + 3/2H_2$	-671.6 kJ mol <sup>-1</sup>	117.67
Nd <sub>2</sub> O <sub>3</sub>	$1/2Nd_2O_3 + 3H^+ = Nd^{3+} + 3/2H_2O$	-166.89 kJ mol <sup>-1</sup>	29.24
NdPO <sub>4</sub>	$NdPO_4 = Nd^{3+} + PO_4^{3-}$	---	-26.2
Sm	$Sm + 3H^+ = Sm^{3+} + 3/2H_2$	-666.6 kJ mol <sup>-1</sup>	116.79
Sm <sub>2</sub> O <sub>3</sub>	$1/2Sm_2O_3 + 3H^+ = Sm^{3+} + 3/2H_2O$	-154.99 kJ mol <sup>-1</sup>	27.16
SmPO <sub>4</sub>	$SmPO_4 = Sm^{3+} + PO_4^{3-}$	---	-26.1
Eu	$Eu + 3H^+ = Eu^{3+} + 3/2H_2$	-574.1 kJ mol <sup>-1</sup>	100.58
Eu <sub>2</sub> O <sub>3</sub>	$1/2Eu_2O_3 + 3H^+ = Eu^{3+} + 3/2H_2O$	-151.39 kJ mol <sup>-1</sup>	26.52
EuPO <sub>4</sub>	$EuPO_4 = Eu^{3+} + PO_4^{3-}$	---	-25.9
Gd	$Gd + 3H^+ = Gd^{3+} + 3/2H_2$	-661 kJ mol <sup>-1</sup>	115.81
Gd <sub>2</sub> O <sub>3</sub>	$1/2Gd_2O_3 + 3H^+ = Gd^{3+} + 3/2H_2O$	-134.79 kJ mol <sup>-1</sup>	23.62
GdPO <sub>4</sub>	$GdPO_4 = Gd^{3+} + PO_4^{3-}$	---	-25.6
Tb	$Tb + 3H^+ = Tb^{3+} + 3/2H_2$	-651.9 kJ mol <sup>-1</sup>	114.21
Tb <sub>2</sub> O <sub>3</sub>	$1/2Tb_2O_3 + 3H^+ = Tb^{3+} + 3/2H_2O$	-119.19 kJ mol <sup>-1</sup>	20.88
TbPO <sub>4</sub>	$TbPO_4 = Tb^{3+} + PO_4^{3-}$	---	-25.3
Dy	$Dy + 3H^+ = Dy^{3+} + 3/2H_2$	-665 kJ mol <sup>-1</sup>	116.51
Dy <sub>2</sub> O <sub>3</sub>	$1/2Dy_2O_3 + 3H^+ = Dy^{3+} + 3/2H_2O$	-134.94 kJ mol <sup>-1</sup>	23.64
DyPO <sub>4</sub>	$DyPO_4 = Dy^{3+} + PO_4^{3-}$	---	-25.1
Ho	$Ho + 3H^+ = Ho^{3+} + 3/2H_2$	-673.7 kJ mol <sup>-1</sup>	118.03
Ho <sub>2</sub> O <sub>3</sub>	$1/2Ho_2O_3 + 3H^+ = Ho^{3+} + 3/2H_2O$	-133.84 kJ mol <sup>-1</sup>	23.45
HoPO <sub>4</sub>	$HoPO_4 = Ho^{3+} + PO_4^{3-}$	---	-25.0
Er	$Er + 3H^+ = Er^{3+} + 3/2H_2$	-669.1 kJ mol <sup>-1</sup>	117.23
Er <sub>2</sub> O <sub>3</sub>	$1/2Er_2O_3 + 3H^+ = Er^{3+} + 3/2H_2O$	-120.44 kJ mol <sup>-1</sup>	21.10
ErPO <sub>4</sub>	$ErPO_4 = Er^{3+} + PO_4^{3-}$	---	-25.1
Tm	$Tm + 3H^+ = Tm^{3+} + 3/2H_2$	-662 kJ mol <sup>-1</sup>	115.98
Tm <sub>2</sub> O <sub>3</sub>	$1/2Tm_2O_3 + 3H^+ = Tm^{3+} + 3/2H_2O$	-120.44 kJ mol <sup>-1</sup>	21.10
TmPO <sub>4</sub>	$TmPO_4 = Tm^{3+} + PO_4^{3-}$	---	-25.0

连续表  
Continuing table

材料反应 Materials	Reaction	$\Delta G$	$\log_{10}K_{sp}$
Yb	$Yb + 3H^+ = Yb^{3+} + 3/2H_2$	-644 kJ mol <sup>-1</sup>	112.83
Yb <sub>2</sub> O <sub>3</sub>	$1/2Yb_2O_3 + 3H^+ = Yb^{3+} + 3/2H_2O$	-136.34 kJ mol <sup>-1</sup>	23.89
YbPO <sub>4</sub>	$YbPO_4 = Yb^{3+} + PO_4^{3-}$	---	-24.8
Lu	$Lu + 3H^+ = Lu^{3+} + 3/2H_2$	-628 kJ mol <sup>-1</sup>	110.03
Lu <sub>2</sub> O <sub>3</sub>	$1/2Lu_2O_3 + 3H^+ = Lu^{3+} + 3/2H_2O$	-89.19 kJ mol <sup>-1</sup>	15.63
LuPO <sub>4</sub>	$LuPO_4 = Lu^{3+} + PO_4^{3-}$	---	-24.7

注：对于稀土金属， $\log_{10}K_{sp}$ 值是根据CRC物理和化学手册(63)中的形成常数自由能计算得出的。  
**Note:** For REE metals, the  $\log_{10}K_{sp}$  values are calculated from the free energy of formation constants in the CRC Handbook of Physics and Chemistry (63). For REE oxides, the  $\log_{10}K_{sp}$  values are calculated from the free energy of formation constants reported in the NBS tables of thermodynamic properties (64). There is no free energy of formation for Pr<sub>2</sub>O<sub>3</sub>, Gd<sub>2</sub>O<sub>3</sub>, and Tb<sub>2</sub>O<sub>3</sub> in NBS table; for these oxides, the  $\log_{10}K_{sp}$  values are calculated based on the enthalpy of formation and standard entropy for element from NBS table (64), and standard entropy of the oxides from a literature (65). For REE phosphates, the  $\log_{10}K_{sp}$  values are reported in literatures for ScPO<sub>4</sub> (66) and other REE phosphates (67). Generally, the REE phosphates are hard to dissolve and have the  $\log_{10}K_{sp}$  values in the range of -24 to -27. These values are significantly smaller than those of REE metals and REE oxides. As a result, REE metals and REE oxides are much more extractable by acid than the REE phosphate counterparts.

表S4。La和Y的XPS峰拟合。  
Table S4. The XPS peak fitting of La and Y.

样品 Samples	要素 Element	位置(eV), Position (eV), 峰 peak	化学状态、化学键 Chemical state, Chemical bond
La <sub>2</sub> O <sub>3</sub>	La	834.8, La 3d <sub>5/2</sub>	+3, La-O
		838.2, 卫星 838.2, satellite	+3, La-O
FJH La之后的La <sub>2</sub> O <sub>3</sub> La <sub>2</sub> O <sub>3</sub> after FJH	La	834.8, La 3d <sub>5/2</sub>	+3, La-O
		836.0, La 3d <sub>5/2</sub>	0, La-La
		838.1, 卫星 838.1, satellite	+3, La-O
		839.6, 卫星 839.6, satellite	0, La-La
Y <sub>2</sub> O <sub>3</sub>	Y	157.4, Y 3d <sub>5/2</sub>	+3, Y-O
		159.4, Y 3d <sub>3/2</sub>	+3, Y-O
FJH Y之后的Y <sub>2</sub> O <sub>3</sub> Y <sub>2</sub> O <sub>3</sub> after FJH	Y	156.4, Y 3d <sub>5/2</sub>	0, Y-Y
		157.5, Y 3d <sub>5/2</sub>	+3, Y-O
		158.5, Y 3d <sub>3/2</sub>	0, Y-Y
		159.6, Y 3d <sub>3/2</sub>	+3, Y-O