

Cs to Ba Element Transmutation in the Presence of Hydride

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Abstract: This work reported evidences of Cs to Ba transmutation in the presence of hydride compounds. Experiments identified that the concentration and isotope ratio were changed after reaction of Cs salt with hydride at room temperature. In addition, we confirmed that these phenomena are closely related to Ru catalytic role. Those results imply that some of barium isotope in nature might originate from an unknown low-energy nuclear reaction between Cs and hydride under very mild conditions.

Key words: Cs to Ba element transmutation; hydride; isotope concentration and ratio; ICP-MS

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The origin of the elements in the universe is one of the most basic scientific questions. According to our current knowledge, the universe originated from the Big Bang, which created some light elements, such as hydrogen, helium, and a very small amount of lithium within very short time of this incredible process^[1-3]. After then, more than 80 elements are generated *via* a series of complicated nuclear reactions under very harsh conditions^[3]. Radioactive decay or natural nuclear reaction might also lead to formation of some elements or their isotopes, such as U to Pd^[4-6]. It is known that the ⁴⁰Ar and small amount of radio nuclides ¹⁴C are formed by constantly bombing under the high energy cosmic rays in the Earth's atmosphere^[4-6]. Of course, we can produce some of element else from target element by high energy neutron and proton bombing typically in accelerators or Tokamak^[7-10]. However, there are some mysterious questions related to the element abundance in our universe, for example, current understanding can not account for measured excess helium element abundance in our universe^[11]. Scientists found that there were excess ratios of ³He and ⁴He in submarine hydrothermal water^[12]. The investigation of energy balance of Jupiter also seems indicate the possibility of low energy fusion reaction there^[13].

It is well known that one element or its isotope can

be converted into another element or isotope by nuclear transmutation^[14]. In 1919, Rutherford reported the first artificial transmutation from nitrogen into oxygen by alpha particle bombing^[15]. In 1932, John Cockcroft and Ernest Walton fulfilled an artificial nuclear reaction by ⁷Li bombing with accelerated protons to split the Li nucleus into two alpha particles^[6]. In 1938, Otto Hahn et al. discovered artificial heavy element transmutation, ¹⁹⁷Au + n → ¹⁹⁸Au, finally → ²⁰⁴Ti and ²⁰⁴Pb^[9]. In fact, scientists found that transmutation can be induced in “mild” reactive systems, like in metabolism processes of vegetal and animal organisms^[16], in which some elemental atoms are transmuted into the different ones while discharging energy slowly. Kervran *et al.* investigated potassium and calcium content variation during the growth of 840 seeds and 403 sprouts in biological systems, and found potassium may transmute into calcium during the process of seeds growing, ³⁹K + ¹p = ⁴⁰Ca + E^[17]. Some other elemental transmutations were also reported, such as sodium to magnesium (²³₁₁Na + n → ²⁴₁₁Na * → ²⁴₁₂Mg + e⁻ + ν_e *) and manganese to iron (⁵⁵₂₅Mn + n → ⁵⁶₂₅Mn * → ⁵⁶₂₆Fe + e⁻ + ν_e *)^[17]. Those investigations imply that the elemental transmutation is essential to maintain a balance of certain elements in the biological bodies. Later on, much more reports of element transmutation by chemical or physi-

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cal was appeared, for instance, the so-called cold fusion experiments which was done by Fleischmann and Pons, in which they found that trace amount of He could be produced by electrochemical method under normal conditions while excess heat was generated^[18].

We previously reported that deuterium and helium were generated from protons by low energy nuclear reaction under light irradiation^[19–23] and the transmutation of potassium element to calcium during photochemical reaction of hydrogen evolution (HER) in the presence of hydride (negative hydrogen ions— H^- , which is offered by negative hydrogen compounds such as NaBH_4 , LiBH_4 and NH_3BH_3 ^[22]). Potassium was chosen as the target transmutation element because of the essential roles of potassium and calcium in biological bodies.

In this work, we found interesting results about the concentration variation of Ba isotopes in the presence of hydride and CsCl. Experimental results showed that the significant concentration and ratio variation of ^{130}Ba , ^{132}Ba , ^{134}Ba , ^{135}Ba , ^{136}Ba , ^{137}Ba and ^{138}Ba isotopes after very short time reaction. Especially, the changes of ^{130}Ba and ^{132}Ba concentration are remarkable compared with other isotopes. These results are extremely meaningful for understanding the elements evolution on the Earth, as well as providing new way to looking into the low energy nuclear reaction.

1 Experimental details

All chemicals were purchased and used without

further purification, NaBH_4 (Shanghai Guangming Chemical Reagent Co., Ltd, AR, $\geq 97\%$), CsCl (Kemiao Chemical Co., Ltd., AR, $\geq 99.5\%$), RuCl_3 (Shanghai Guangming Chemical Reagent Co., Ltd, AR, $\geq 37.3\%$), and BaCl_2 (Xilong Chemical Works, AR, $\geq 99.5\%$). Ultrapure water: $18.2\text{ M}\Omega \cdot \text{cm}^{-1}$ at $25\text{ }^\circ\text{C}$ (Milli-Q water, Millipore Mili-Q reference ultrapure water purification system, USA) was used in this study. The polypropylene (PP) volumetric flask was used in this work since it is chemical stable both in the acidic and alkaline environment. The involvement of other elements can be eliminated after very careful checking.

Prior to the reaction, all PP volumetric flasks (100 mL) were recalibrated and washed several times with high pure Mill-Q water, and then they were dried at room temperature. The typical isotope trasmutation (Cs-Ba) experiment was done as follows. 50.0 mL of ultrapure water, 1.0 mL of CsCl solution ($C_{\text{Cs}} = 200\text{ mg} \cdot \text{kg}^{-1}$) and 2.0 mL RuCl_3 solution ($20\text{ mg} \cdot \text{mL}^{-1}$) were added in a 100 mL PP volumetric flask. After 5 min ultrasound treatment, different content NaBH_4 was added in the mixture solution, and the reaction was operated for days until no bubbles were generated under the room temperature. After that, the mixed solution was volumed to 100 mL with Mill-Q high purity water. For the above reaction system, it was labeled as $\text{NaBH}_4 + \text{CsCl} + \text{RuCl}_3$. The experimental results are shown in Fig.1.

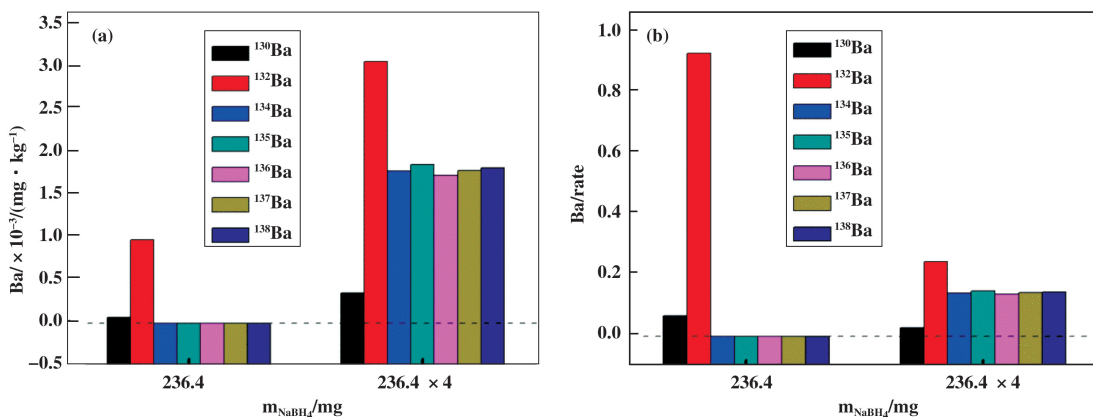


Fig.1 (a) Ba isotope concentration variation in $\text{NaBH}_4 + \text{CsCl} + \text{RuCl}_3$ mixture with different NaBH_4 content 236.4 and 236.4×4 mg; (b) the corresponding isotope ratio

The element and isotope concentration and ratio variation before and after reaction were measured by inductive coupled plasma mass spectrometry (ICP-MS, iCAP Q, Thermo, Waltham, USA) technique. The apparatus limits of detection for Ba and Cs isotope are 0.40 and 0.05 ppt respectively. The samples for ICP-MS measurement were not further diluted prior to analysis. The every sample was detected at least three times and the average value was presented.

2 Results and discussion

ICP-MS measurement results are shown in Fig.1a and Table 1. The reaction mixture contains 1 mL CsCl ($C_{Cs} = 200 \text{ mg} \cdot \text{kg}^{-1}$) and 2.0 mL RuCl_3 ($20 \text{ mg} \cdot \text{mL}^{-1}$) solution in 100 mL PP volumetric flask. By changing the amount of NaBH_4 that are 236.4 and $236.4 \times 4 \text{ mg}$, respectively, we found that the Ba isotope concentration changed with the increase of negative hydrogen compound loading (NaBH_4). It should be noted that there is no additional barium source in these initial reaction mixtures. Meanwhile, the Ba isotope ratio between ^{130}Ba , ^{132}Ba , ^{134}Ba , ^{135}Ba , ^{136}Ba , ^{137}Ba and

Table 1 Ba isotope concentration variation in the mixture of $\text{NaBH}_4 + \text{CsCl} + \text{RuCl}_3$ with different NaBH_4 loading

Isotope $\times 10^{-3} /$ ($\text{mg} \cdot \text{kg}^{-1}$)	m_{NaBH_4} (236.4 mg)	m_{NaBH_4} (236.4 $\times 4$ mg)
^{130}Ba	0.07	0.36
^{132}Ba	0.98	3.08
^{134}Ba	<0.01	1.79
^{135}Ba	<0.01	1.87
^{136}Ba	<0.01	1.74
^{137}Ba	<0.01	1.80
^{138}Ba	<0.01	1.83

^{138}Ba also changed after the reaction, as shown in Fig. 1b. In particular, the amount of isotope ^{130}Ba and ^{132}Ba decrease remarkably with the increase of negative hydrogen compound loading.

To avoid misunderstanding of the experimental results, we double checked the initial Ba and Cs isotope-concentration in NaBH_4 , RuCl_3 , CsCl and BaCl_2 solutions by ICP-MS. As results shown in Fig.2a, the Ba

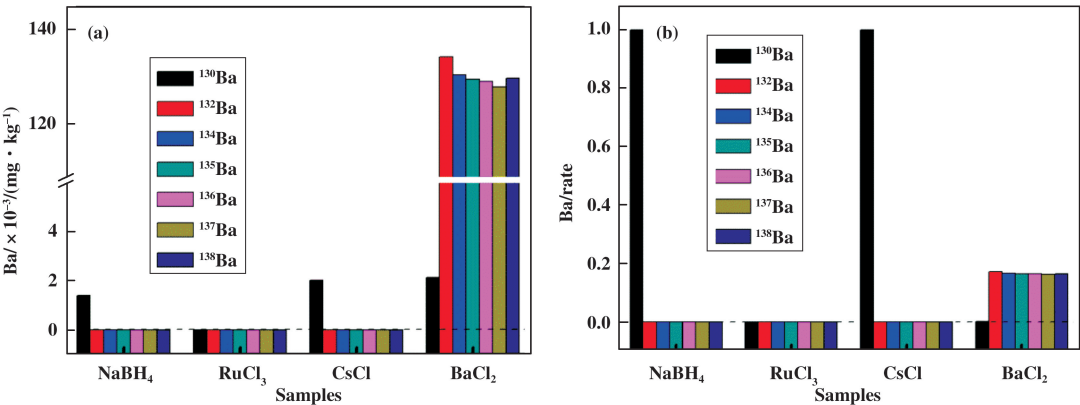


Fig.2 The Ba isotope concentration in used reagents
(a) Ba isotope concentrations in the NaBH_4 , RuCl_3 , CsCl and BaCl_2 solution respectively;
(b) the corresponding isotope ratio in the NaBH_4 , RuCl_3 , CsCl and BaCl_2 solution respectively

isotope concentrations of ^{132}Ba , ^{134}Ba , ^{135}Ba , ^{136}Ba , ^{137}Ba and ^{138}Ba in NaBH_4 and CsCl solution are lower than the instrument's limit of detection. Only a trace amount ^{130}Ba was found in NaBH_4 ($1.41 \times 10^{-3} \text{ mg} \cdot \text{kg}^{-1}$) and CsCl ($2.03 \times 10^{-3} \text{ mg} \cdot \text{kg}^{-1}$) solution respectively. The concentrations of the Ba isotopes in RuCl_3 aqueous solution are also very low, as shown in

Table 2. This means that the changing of Ba isotope concentration can only be ascribed to elemental transmutation reaction in the $\text{NaBH}_4 + \text{CsCl} + \text{RuCl}_3$ mixture. The distribution of isotope ratio in BaCl_2 aqueous solution ($C_{\text{Ba}} = 132.0 \times 10^{-3} \text{ mg} \cdot \text{kg}^{-1}$) was also detected, and detected ^{130}Ba concentration in BaCl_2 is significantly lower than other isotope concentrations.

Table 2 Ba isotope concentrations in NaBH₄, RuCl₃, CsCl₃ and BaCl₂ solution

Isotope×10 ⁻³ / (mg · kg ⁻¹)	NaBH ₄ (236.4 mg)	RuCl ₃ (40 mg)	CsCl (C _{Cs} = 20×10 ⁻³ mg · kg ⁻¹)	BaCl ₂ (C _{Ba} = 132×10 ⁻³ mg · kg ⁻¹)
¹³⁰ Ba	1.41	<0.01	2.03	2.15
¹³² Ba	<0.01	<0.01	<0.01	134.17
¹³⁴ Ba	<0.01	<0.01	<0.01	130.38
¹³⁵ Ba	<0.01	<0.01	<0.01	129.4
¹³⁶ Ba	<0.01	<0.01	<0.01	128.98
¹³⁷ Ba	<0.01	<0.01	<0.01	127.76
¹³⁸ Ba	<0.01	<0.01	<0.01	129.71

Fig. 3a shows the Ba isotope concentrations in NaBH₄+CsCl+RuCl₃ and NaBH₄+CsCl mixtures with different CsCl concentrations. We found that the Ba i-

sotope concentrations, such as ¹³⁰Ba and ¹³²Ba, are obvious different in two reaction mixtures (with or without RuCl₃) as shown in Table 3. The results indicates that

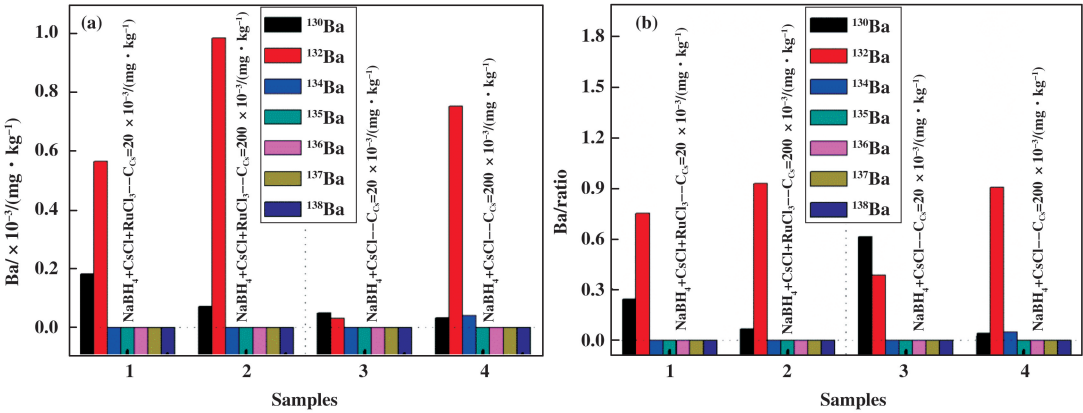


Fig.3 (a) Ba isotope concentrations in NaBH₄+CsCl+RuCl₃ and NaBH₄+CsCl mixtures with different Cs content (C_{Cs} = 20×10⁻³ mg · kg⁻¹) (1#, 2#) and (C_{Cs} = 200×10⁻³ mg · kg⁻¹) (3#, 4#); (b) the corresponding isotope ratio in their respective mixtures

Table 3 Ba isotope concentrations in NaBH₄+CsCl+RuCl₃ and NaBH₄+CsCl mixture

Isotope (×10 ⁻³ mg · kg ⁻¹)	1	2	3	4
¹³⁰ Ba	0.183	0.072	0.051	0.034
¹³² Ba	0.565	0.984	0.032	0.752
¹³⁴ Ba	<0.01	<0.01	<0.01	0.041
¹³⁵ Ba	<0.01	<0.01	<0.01	<0.01
¹³⁶ Ba	<0.01	<0.01	<0.01	<0.01
¹³⁷ Ba	<0.01	<0.01	<0.01	<0.01
¹³⁸ Ba	<0.01	<0.01	<0.01	<0.01

the Ru has a catalytic effect on the Ba isotope concentration change. Moreover, increasing the initial CsCl concentration, the contents of Ba isotopes also change significantly in the NaBH₄ + CsCl + RuCl₃ or NaBH₄ + CsCl mixture. In short, introduction of catalyst Ru and Cs are decisive for ¹³⁰Ba and ¹³²Ba isotope conversion. By comparison the Ba isotope ratio in Fig.3b, it is confirmed that the isotope concentration distribution of Ba is closely related to Ru and Cs addition.

Does the variation of Ba isotope concentration and ratio result from interaction of Ba itself and hydride? We conducted a reaction of hydride with Ba in the ab-

sence of Cs. In $\text{NaBH}_4 + \text{BaCl}_2 + \text{RuCl}_3$ mixture, the Ba isotope concentration almost maintained similar under different hydride loading, according to results shown in Fig. 4 and Table 4. That confirmed that the concentration variation was not due to of the reaction between Ba isotopes with negative hydrogen (NaBH_4), instead, from reaction between Cs and hydride, i. e., ${}^m\text{Cs} + \text{H}^- \rightarrow {}^n\text{Ba}$.

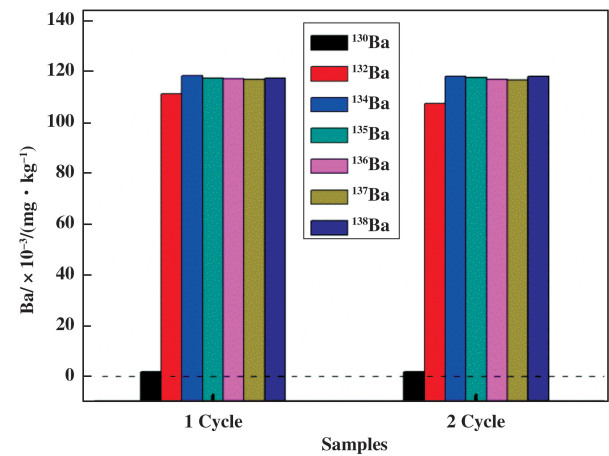


Fig.4 Ba isotope concentrations in $\text{NaBH}_4 + \text{BaCl}_2 + \text{RuCl}_3$ mixtures detected by ICP-MS incyclic experiments at 25 °C, (I) 1 cycle, $[\text{NaBH}_4]$: 236.4 mg; (II) addition of another portion of NaBH_4 (236.4 mg). Initial reactants; $m_{\text{RuCl}_3} = 40 \text{ mg}$ and $C_{\text{Ba}} = 132 \times 10^{-3} \text{ mg} \cdot \text{kg}^{-1}$

Table 4 Ba isotope concentrations in $\text{NaBH}_4 + \text{BaCl}_2 + \text{RuCl}_3$ mixtures detected by ICP-MS, (I) 1 cycle, $[\text{NaBH}_4]$: 236.4 mg; (II) addition of fresh NaBH_4 (236.4 mg)

Isotope ($\times 10^{-3} \text{ mg} \cdot \text{kg}^{-1}$)	1 Cycle	2 Cycle
¹³⁰ Ba	1.91	1.93
¹³² Ba	111.23	107.53
¹³⁴ Ba	118.48	118.3
¹³⁵ Ba	117.63	117.81
¹³⁶ Ba	117.38	117.1
¹³⁷ Ba	117.02	116.75
¹³⁸ Ba	117.4	118.28

3 Conclusion

This work reported evidences of Cs to Ba transmutation in the presence of hydride compounds. Experi-

ments identified that the concentration and isotope ratio were changed after reaction of Cs salt with hydride at room temperature. In addition, we confirm that these phenomena are closely related to Ru catalyst role. Those results imply that some of barium isotope in nature might originate from an unknown low-energy nuclear reaction between Cs and hydride under very mild conditions.

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References:

[1] Burbidge E M, Burbidge G R, Fowler W A, *et al.* Synthesis of the elements in stars [J]. *Rev Mod Phys*, 1957, **29**(4): 547.

[2] Fowler W A. Experimental and theoretical nuclear astrophysics: The quest for the origin of the elements [J]. *Mod Phys*, 1984, **56**(2): 149.

[3] Woosley S E, Heger A, Weaver T A. The evolution and explosion of massive stars [J]. *Mod Phys*, 2002, **74**(4): 1015.

[4] Badash L. Radium, radioactivity, and the popularity of scientific discovery [J]. *Proc Am Philos Soc*, 1978, **122**(3): 145–154.

[5] Howorth M. Pioneer research on the atom: Rutherford and soddy in a glorious chapter of science; the life story of Frederick Soddy[M]. *New World Publications*, 1958.

[6] Trenn T J. The self-splitting atom: the history of the Rutherford-Soddy collaboration [M]. *London: Taylor & Francis*, 1977, **42**: 58-60; 111–117.

[7] Cockcroft J, Walton E. Artificial Production of Fast Protons (Reprinted from Nature, February 13, 1932) [J]. *Nature*, 1969, **224**: 463.

[8] Henderson M. The disintegration of lithium by protons of high energy [J]. *Phys Rev*, 1933, **43**(2): 98–102.

[9] Paneth F A, Günyher P L. Chemical detection of artificial transmutation of elements [J]. *Nature*, 1933, **131**: 652–653.

[10] Bush R P. Recovery of platinum group metals from high level radio active waste [J]. *Platinum Metals Rev*, 1991, **35**: 202–208.

[11] Yoshida N, Oh S P, Kitayama T, *et al.* Early cosmological $\text{H}_2/\text{}^3\text{He}$ regions and their impact on second-generation star formation[J]. *Astrophys J*, 2007, **663**: 687–

- 707.
- [12] Craig H, Clarke W B, Beg M A. Excess ^3He in deep water on the east pacific rise [J]. *Earth Planet Sci Lett*, 1975, **26**: 125–132.
- [13] Hanel R A, Conrath B J, Herath L W, *et al.* Albedo, internal heat, and energy balance of jupiter: preliminary results of the voyager infrared investigation[J]. *J Geophys Res*, 1981, **86**(A10): 8705–8712.
- [14] Schaeffer O A, Zähringer J. Solar flare helium in satellite materials [J]. *Phys Rev Lett*, 1962, **8**: 389.
- [15] Badash L. Radium, radioactivity and the popularity of scientific discovery [J]. *Proc Am Philos Soc*, 1978, **122**: 145–154.
- [16] Kervran C L. Biological transmutations[M]. (revised and edited by H. Rosenauer and E. Rosenauer, Crosby Lockwood, London 1972, reprinted by Beekman, New York, 1980, reprinted 1998).
- [17] Biberian J P. Biological transmutations [J]. *Curr Sci*, 2015, **108**: 633–635.
- [18] Fleischmann M, Pons S. Electrochemically induced nuclear fusion of deuterium [J]. *J Electroanal Chem Interf Electrochem*, 1989, **261**: 301–308.
- [19] Lu Gong-xuan(吕功煊), Tian Bin (田彬). Formation of deuterium and helium during photocatalytic hydrogen generation from water catalyzed by Pt-graphene sensitized with Br-dye under visible light irradiation (溴染料敏化负载 Pt 石墨烯催化可见光制氢、氘和氦) [J]. *J Mol Catal (China)*(分子催化), 2017, **31**(2): 101–104.
- [20] Lu Gong-xuan (吕功煊), Zhen Wen-long (甄文龙). Formation of deuterium and helium during photocatalytic hydrogen generation from water catalyzed by Pt-Graphene sensitized with Br-dye under visible light irradiation (半导体 CdS 悬浮体系中可见光催化产氢同时生成氘-3 和氦-4) [J]. *J Mol Catal (China)*(分子催化), 2017, **31**(4): 299–304.
- [21] Lu Gong-xuan(吕功煊), Zhang Wen-yan(张文妍). Photocatalytic hydrogen evolution and induced transmutation of potassium to calcium via low-energy nuclear reaction (LENR) driven by visible light (可见光驱动的光催化产氢同时诱导低能核反应嬗变钾为钙) [J]. *J Mol Catal (China)*(分子催化), 2017, **31**(5): 401–410.
- [22] Lu Gong-xuan(吕功煊), Zhang Xu-qiang (张旭强). The detection of K-Ca transmutation in the mixture of K and hydride chemicals (关于在钾与负氢混合物中钾钙嬗变的检测) [J]. *J Mol Catal (China)*(分子催化), 2019, **33**(1): 1–9.

负氢体系中 Cs 到 Ba 的嬗变转化

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摘要: 我们发现在负氢化合物存在条件下元素 Cs 可以发生嬗变转变为 Ba. 实验证实在室温条件下 Cs 与负氢发生反应不仅导致 Ba 同位素浓度的变化, 而且会诱导发生 Ba 同位素比例的变化. 这个反应的发生与 Ru 的催化作用有关. 结果表明自然界中 Ba 的同位素有可能依照我们还未知的 Cs 与负氢在温和条件下的低能核反应产生.

关键词: Cs 嬗变为 Ba; 负氢; 同位素浓度和比例; ICP-MS