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Recent advance in Thermionic cathode

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Outline

Research Background

Vacuum electronic devices have a wide range of applications.

Communication

Electronic countermeasure

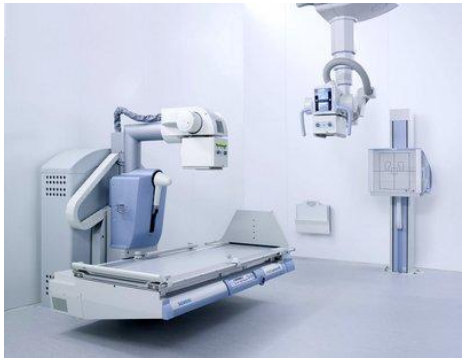
Missile

Vacuum electronic device

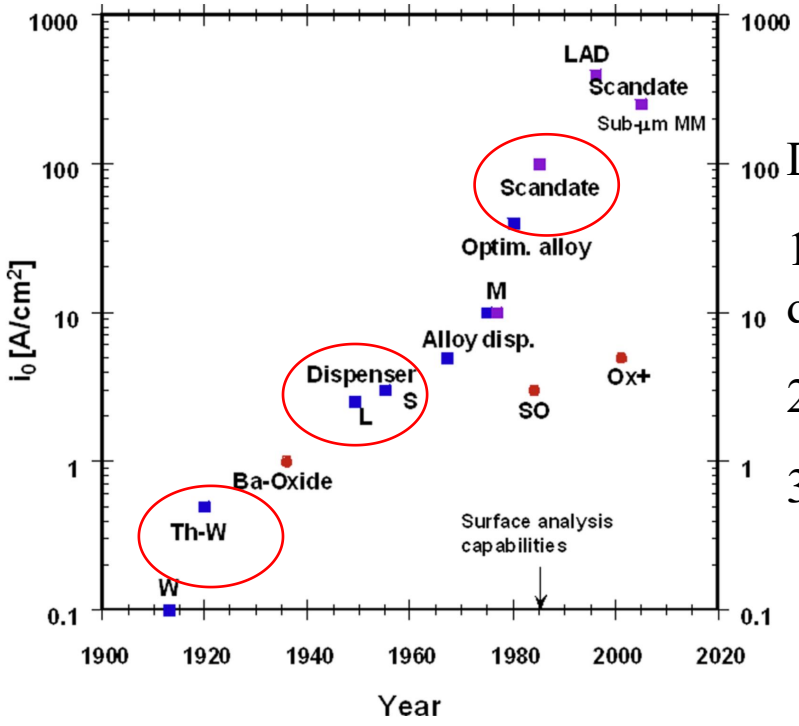
Radar

Microwave heating

Medical care



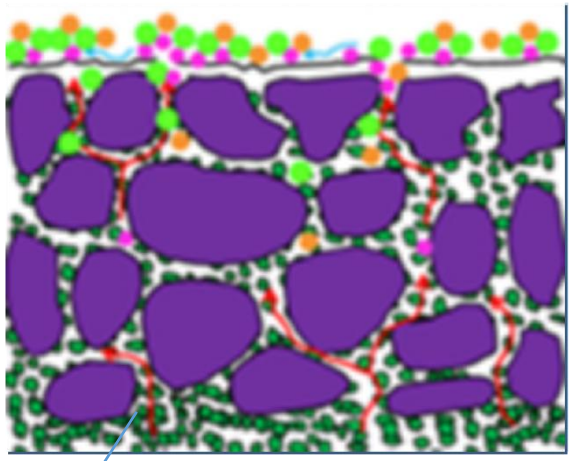
Research background



Themionic cathodes

Dispenser cathode

1. Ba-W dispensercathode
2. M type cathode
3. Scandate cathode



Porous tungsten

Aluminates

Filament cathode

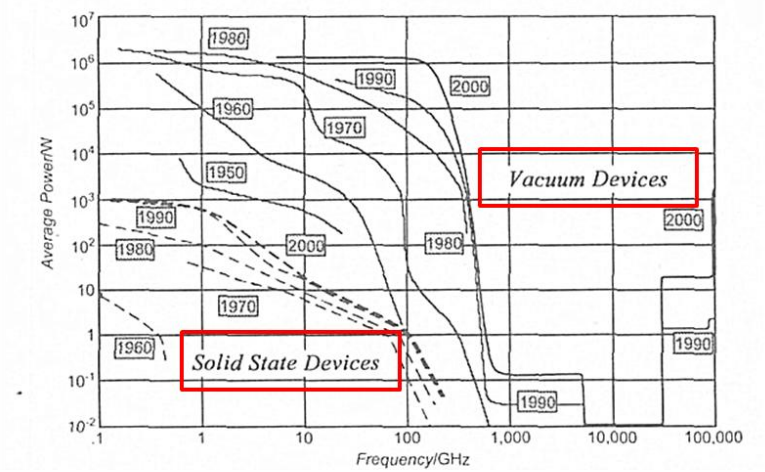
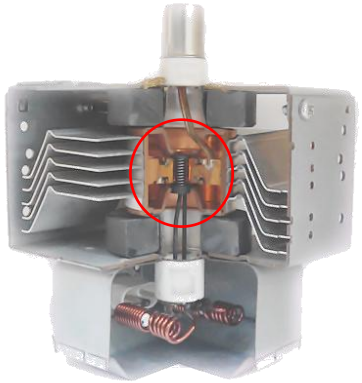


图1 微波真空电子器件和半导体器件单管功率输出

微波炉

磁控管



ThO_2-W cathode

Created in 1913 by Pintsch

High emission stability

1. **Radioactivity**
2. **Unrecycled**
3. **Poor machinability**
4. **Brittle**
5. **High cost**
6. **High working temperature**

Table Parameters of different refractory metals

Metal	Melting point(K)	Work function （eV）	A($A/cm^2 K^2$)	J=1A/cm^2 Temperature（K） and evaporation rate（ μ $g/cm^2.s$）
W	3650	4.52	75	2630 0.012
Mo	2890	4.24	51	2460 4
Ta	3300	4.19	55	2500 0.014
Nb	2770	4.01	30	2420 0.1
Re	3453	4.74	52	2780 0.5

RE→Th

Difficulties

RE_2O_3-Mo alloys

High Emission property

High concentration
RE_2O_3（wt.%）
Bulk cathode：30%
Filament cathode：3-5%

Good ductility

Low concentration
RE_2O_3
(wt.%)
0-0.6%

~1000% deformation

Sintered
Φ16mm

Sintered
Φ16mm

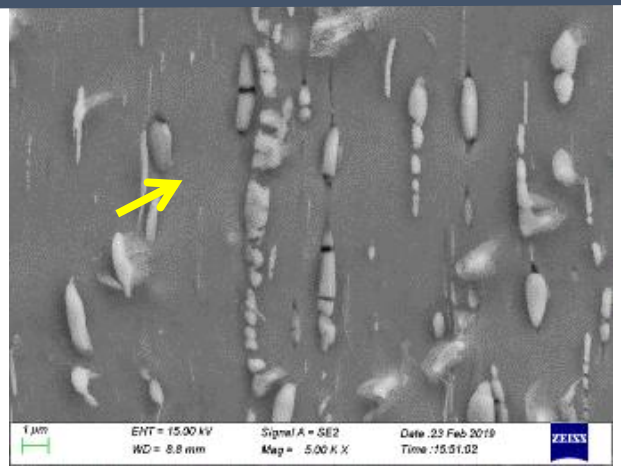
Wires：
Φ0.5mm

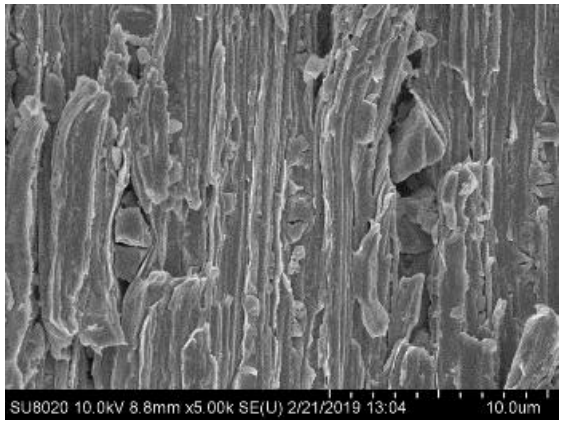
Spring：twist
Φ0.5mm

Weight percent/wt.%	STRAIN/% 25 ℃	STRESS/MPa 25 ℃
0	22.33	562.746
0.01(La_2O_3)	23.97	626.196
0.03(La_2O_3)	40.03	630.907
0.05(La_2O_3)	32.72	645.592
0.08(La_2O_3)	31.02	645.315
0.10(La_2O_3)	24.47	645.315
0.50(La_2O_3)	25.34	606.977
0.60(La_2O_3)	37.58	850
1.00(La_2O_3)	23.30	613.953
1.00(ZrO_2)	17	700
1.50(La_2O_3)	16.12	633.721
1.50(La_2O_3)	35	
1.50(ZrO_2)	18	760
2.00(La_2O_3)	9.61	619.767
2.80(ZrO_2+La_2O_3)	10	988

Competition between
Emission property & ductility

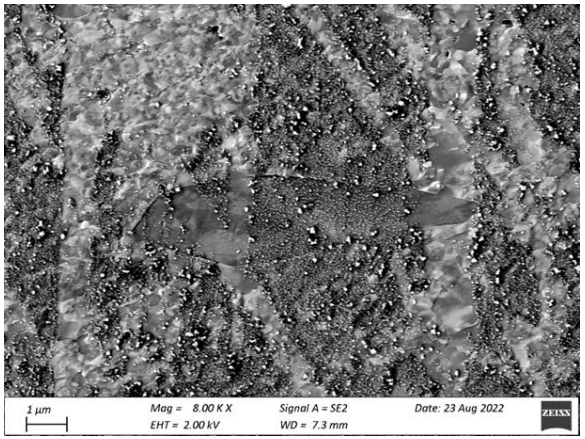
Deformation of La_2O_3-Mo alloys



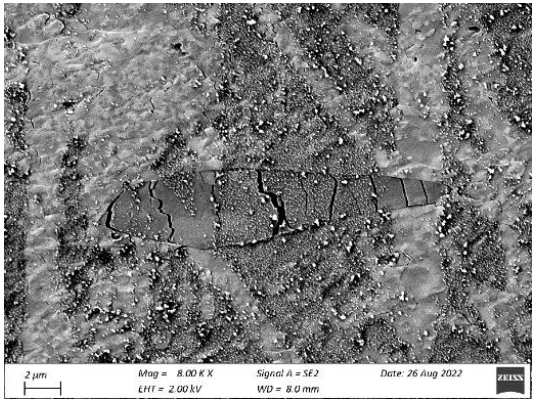


Poor ductility

Brittleness of ceramics: Ionic/Covalent bonding→High dislocation nucleation energy →cracks exist before deformation



strain=0%



strain=2%

How to improve the ductility of alloys

Metal:

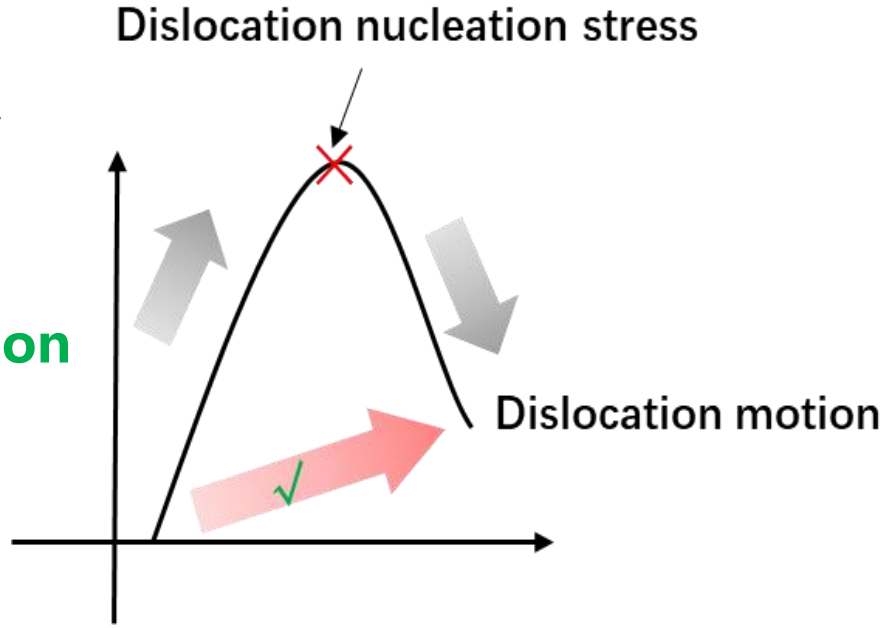
High ductility

Relatively easy for nucleation, multiplication and sliding of dislocations

metal-ceramic Interface

Dislocation corridor

Ceramics(Difficult for dislocation nucleation)



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Borrow dislocations from metal for dislocations propagation and multiplication

How to borrow dislocation for ceramics

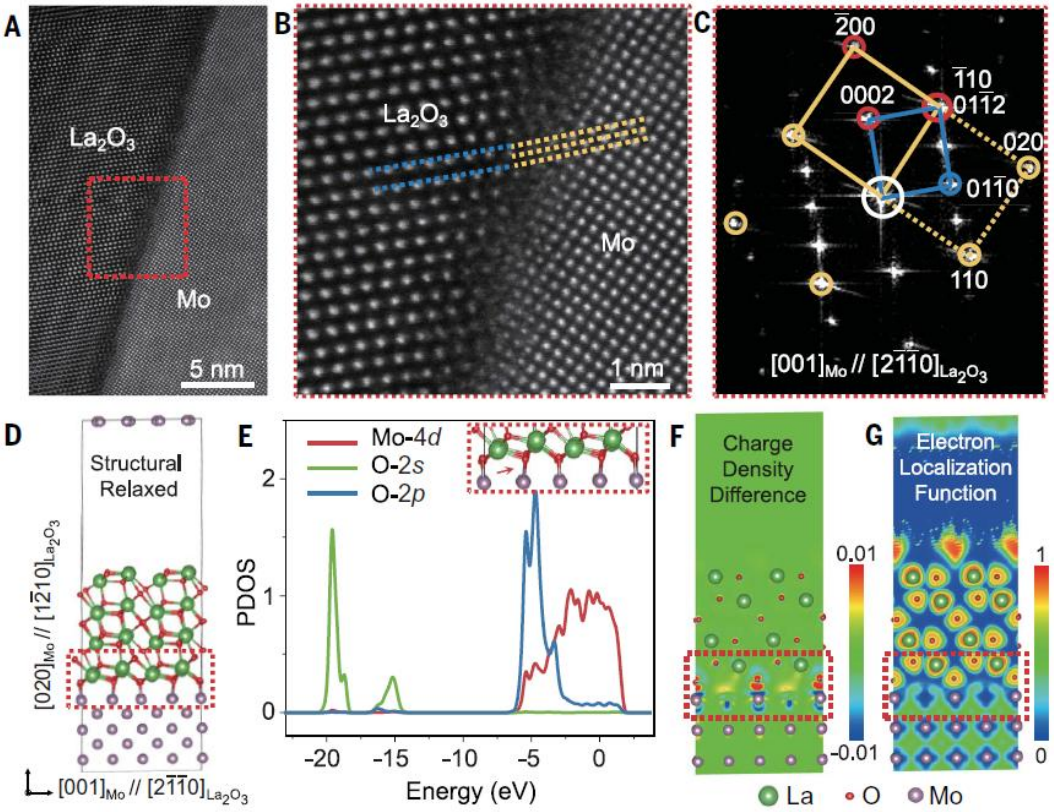
To build the orderly-bonded interface

Condition 1 **Condition 2**

Chemical bonds Ordered interface

⇩ ⇩

Strong No dislocation pile-up



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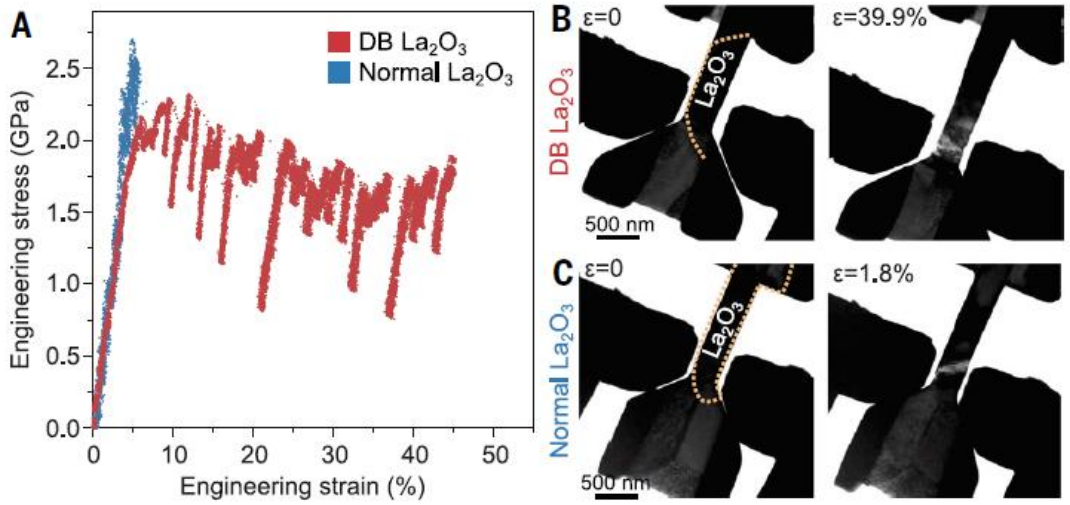
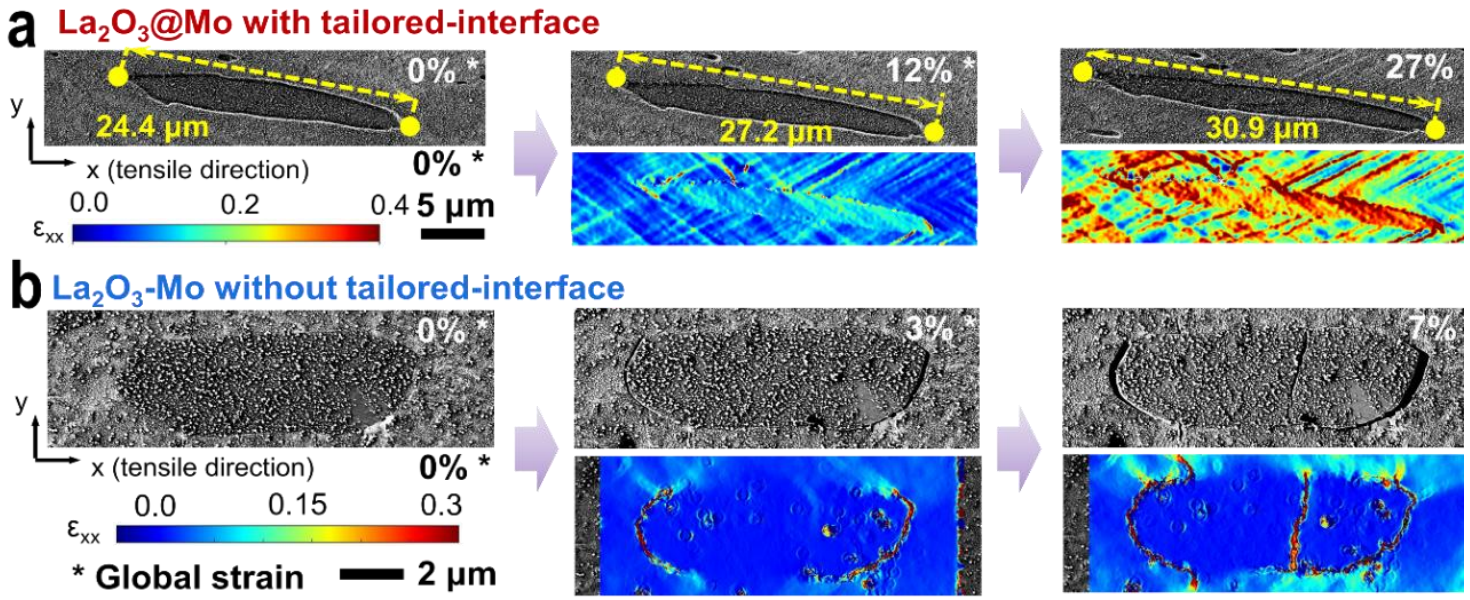
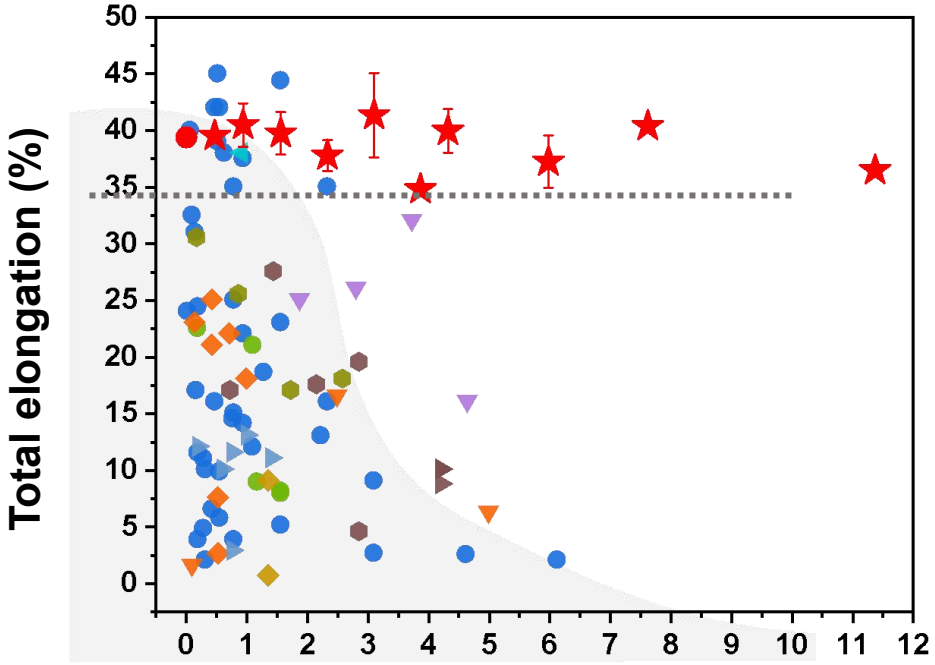


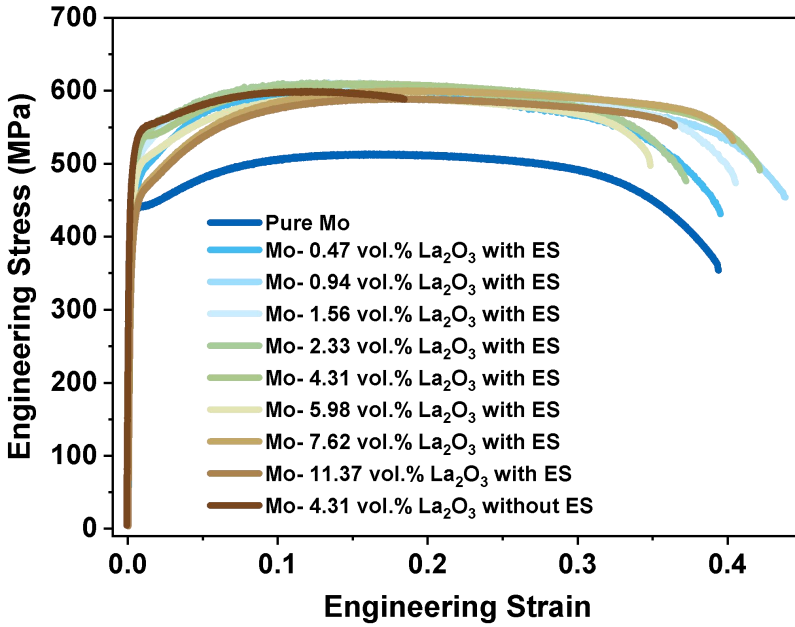
Engineering strain reach to 39.9%, Strength 2.3 Gpa



Tensile ductility of large ceramic particles can reach to 27.7%

The ductility of the Mo alloys with high concentration ceramic phase





The ductility of alloy keeps stable even with high concentration ceramic phase.

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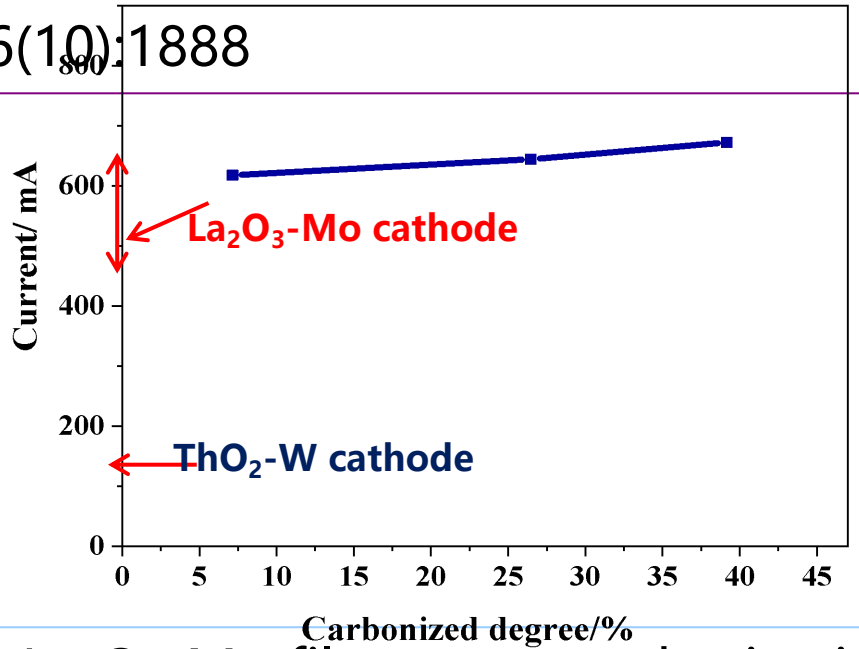
Poor emission stability of La_2O_3-Mo cathode

ThO_2-W

Ch.Buxbaum,Baden: Mo powder + 2%La_2O_3 powder→La_2O_3-Mo body→Plating(Pt)→Carburization(USP,4019081,1977)

Cathode is heated in the mixed gases of benzene and H_2

D M Goebel: Mo powder + 2%La_2O_3 →La_2O_3-Mo sintered body →Carburization，life 100h. Rev.Sci.Instrum,1985, 56(10):1888

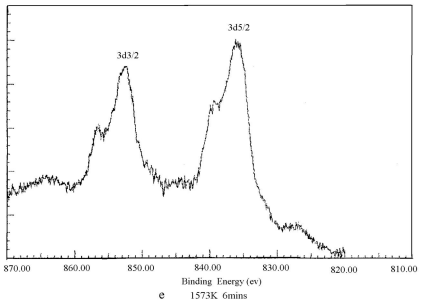


La_2O_3-Mo filament → carburization，life: 90-100h

La_2O_3-Mo cathode
Short lifetime

Poor emission stability?

High temperature evaporation of La $3Mo_2C+La_2O_3=6Mo+2La+3CO$



Carburization temperature 1700°C

Low temperature carburization?

$2Mo+C=Mo_2C$

Kinetics

1350-1450°C

First stage:

$$\frac{\Delta W}{A} = 3.6668\times10^{5}\exp\left(-\frac{2.4075\times10^{5}}{RT}\right)t$$

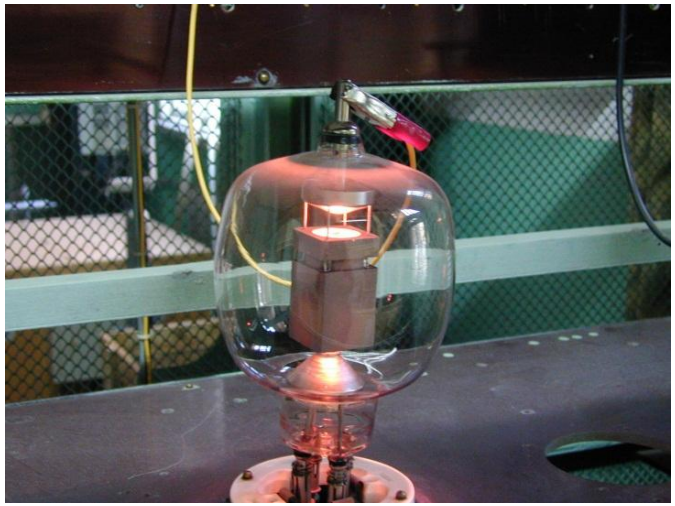
Second stage:

$$\left(\frac{\Delta W}{A}\right)^{2} + 2.0517\times10^{5}\exp\left(-\frac{1.9455\times10^{5}}{RT}\right)\left(\frac{\Delta W}{A}\right) = 4.58678\times10^{10}\exp\left(-\frac{4.27273\times10^{5}}{RT}\right)t$$

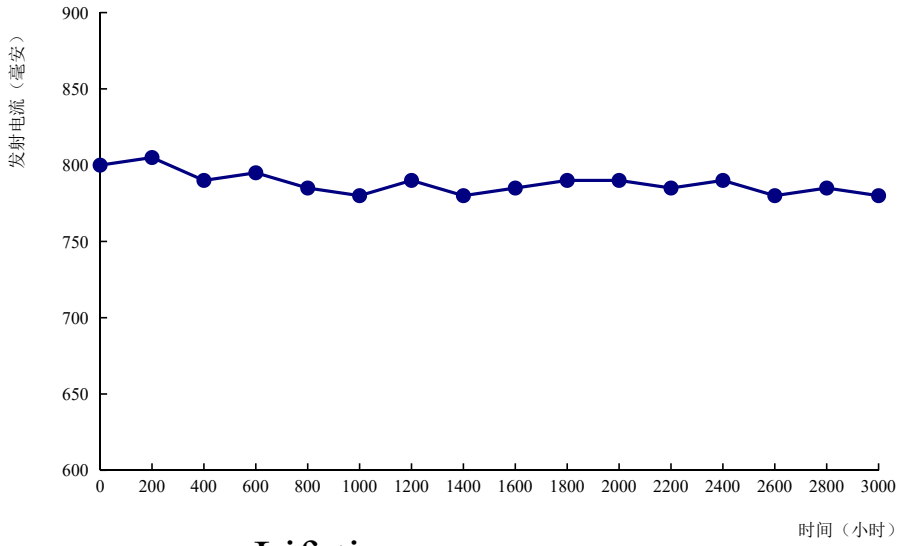
Third stage:

$$\left(\frac{\Delta W}{A}\right)^{2} = 5.2782\times10^{10}\exp\left(-\frac{4.3114\times10^{5}}{RT}\right)t$$

Carburization at lower temperature



FU6051 electron tube



Lifetime

Lifetime: 100h→3000h

(technical standard 1000h)

Ba-W cathode

(Ba,Ca)aluminates

Type of aluminates： 532,612 and 411

Reaction process of the aluminates

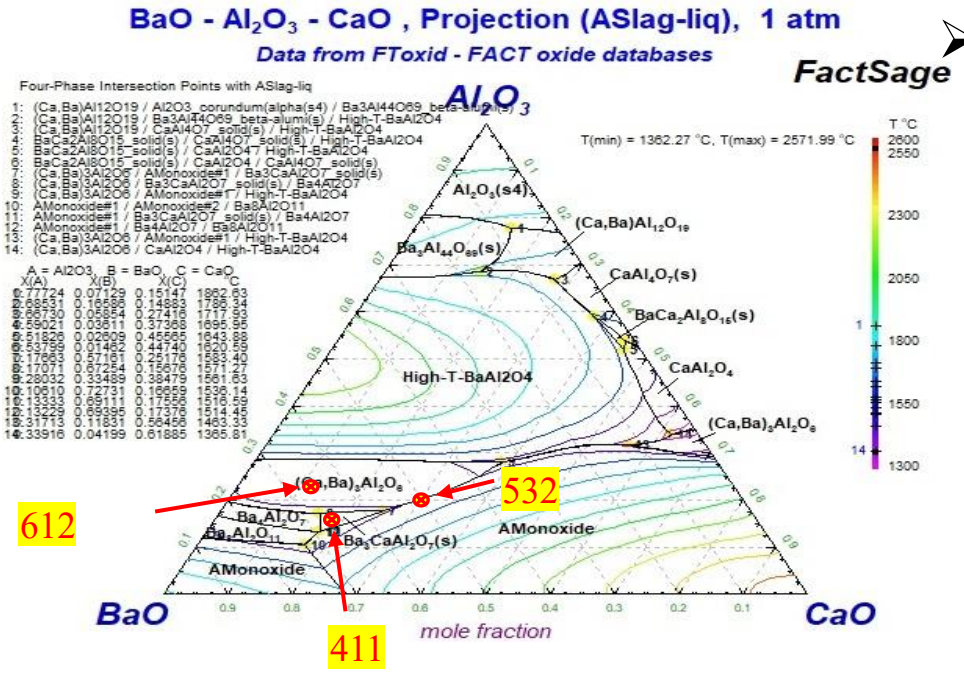


Fig. Ternary phase diagram of Al_2O_3-BaO-CaO

- Theoretical phases

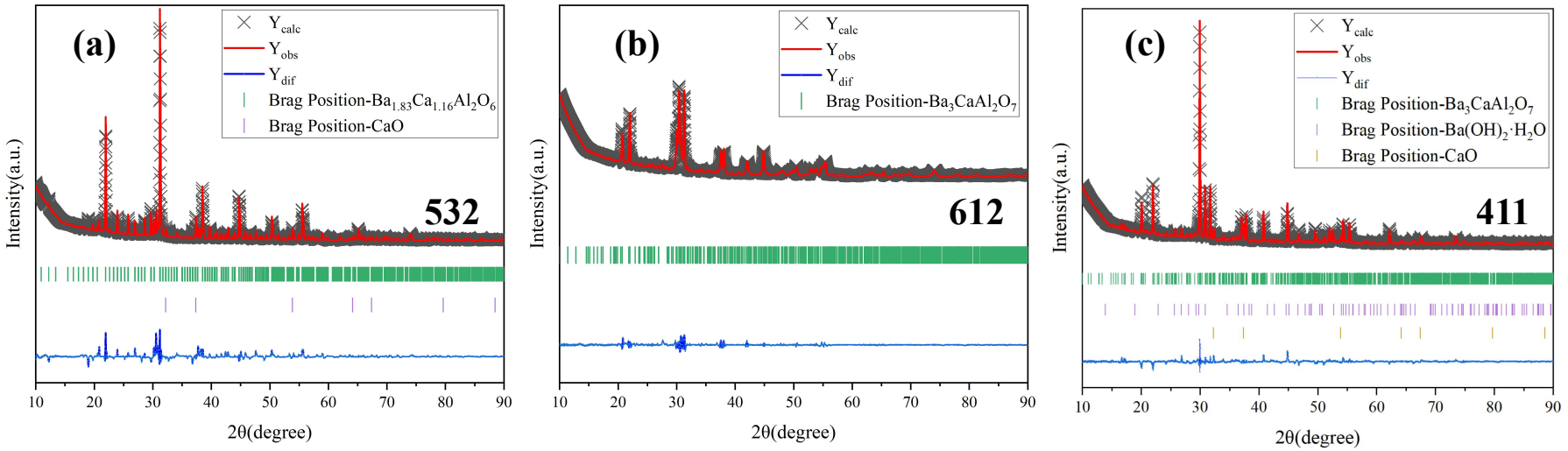
  532： CaO, $(Ca, Ba)_3Al_2O_6$

  612： $(Ca, Ba)_3Al_2O_6$

  411： $Ba_3CaAl_2O_7$

- Possile active phases
- $Ba_8Al_2O_{11}$, $Ba_7Al_2O_{10}$, $Ba_5CaAl_4O_{12}$,$Ba_4Al_2O_7$,
- $Ba_3CaAl_2O_7$,$(Ba, Ca)_3Al_2O_6$

Active phase and crystal stucuture

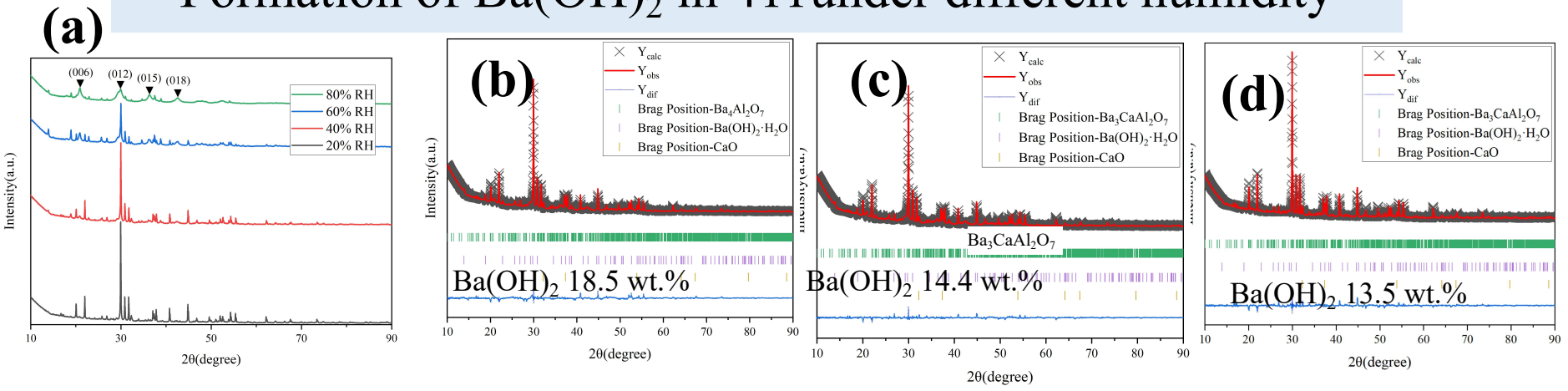


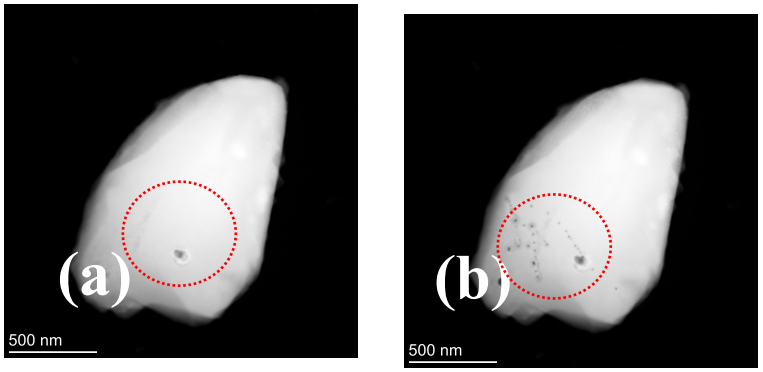
Sample	532	612	411
Data source	ICSD(246227)	PDF(48-0569)	ICSD(150769)
Phase	$Ba_{1.83}Ca_{1.17}Al_2O_6$	$Ba_3CaAl_2O_7$	$Ba_3CaAl_2O_7$(ss)
Mass(wt.%)	88.8(5)	100	81.7(5)
Space group	*$P2_13$*	*Cmca*	*Cmca*
a(Å)	16.1857(9)	11.1360	8.0886(6)
b(Å)	16.1857(8)	27.6413	8.0866(4)
c(Å)	16.1857(7)	11.6731	26.5488(10)
V(Å³)	4240.28(15)	3601.88952	1735.91(17)
α, β, γ(deg.)	90, 90, 90	90, 90, 90	90, 90, 90
Z	24	16	16
R_{wp}(%)	13.89	5.33	7.09

- 532:main phae $Ba_{1.83}Ca_{1.17}Al_2O_6$(ss), a small amount CaO
- 612: $Ba_3CaAl_2O_7$
- 411: $Ba_3CaAl_2O_7$(ss), a small amunt of CaO and **$Ba(OH)_2$**

Cystal structure of active phase

Formation of $Ba(OH)_2$ in 411under different humidity



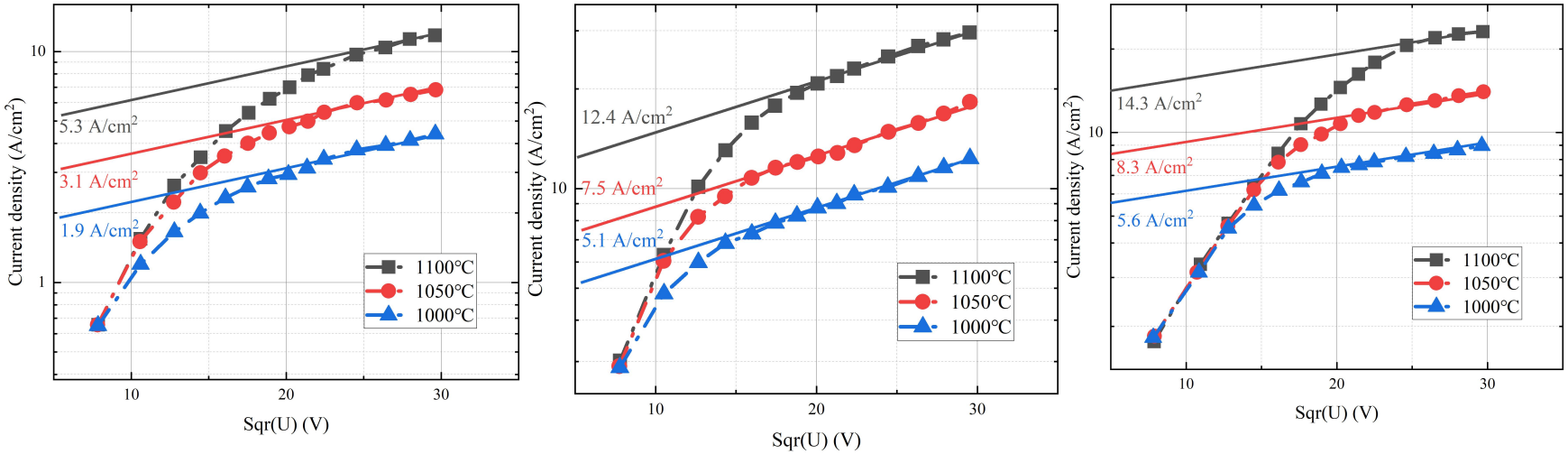


- 20-60% RH， Main phase $Ba_3CaAl_2O_7$ (ss)
- 80% RH ， Main phase is hydrotalcite

PXRD and Rietveld PXRD (b) 60% RH-411; (c) 40% RH-411; (d) 20% RH-411; TEM (e) before (f) after electron bombardment

Compared with 532 and 612, 411 is not stable and easy to absorb water to form $Ba(OH)_2$

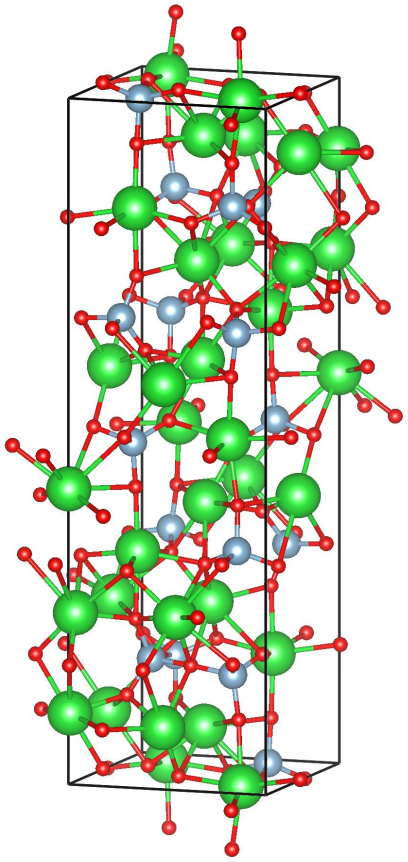
Emission property of the Ba-W cathode impregnated with different aluminates



(a) 532 (b) 612 (c)411

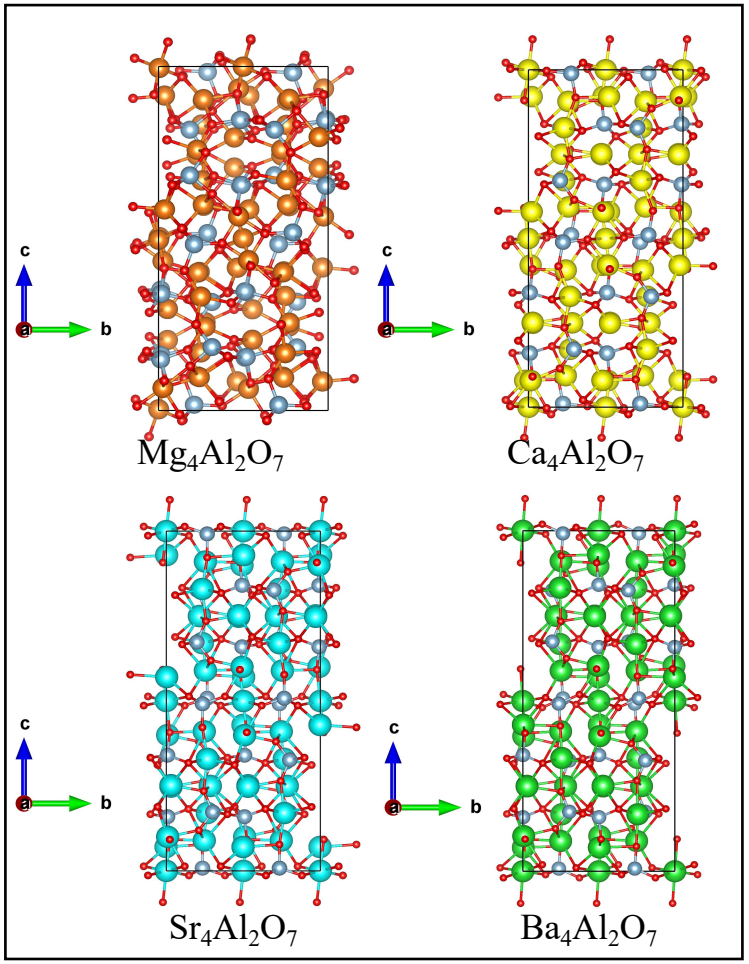
- Electron emission property： 411 > 612 > 532
- The contribution of different main phases:$Ba_3CaAl_2O_7$(ss) > $Ba_3CaAl_2O_7$ > $Ba_{1.83}Ca_{1.17}Al_2O_6$(ss)

Cystal structure of $Ba_3CaAl_2O_7$(ss)--$M_4Al_2O_7$



➢ Substitution

Mg,Ca,Sr,Ba-->$Mg_4Al_2O_7$, $Ca_4Al_2O_7$,$Sr_4Al_2O_7$ and $Ba_4Al_2O_7$

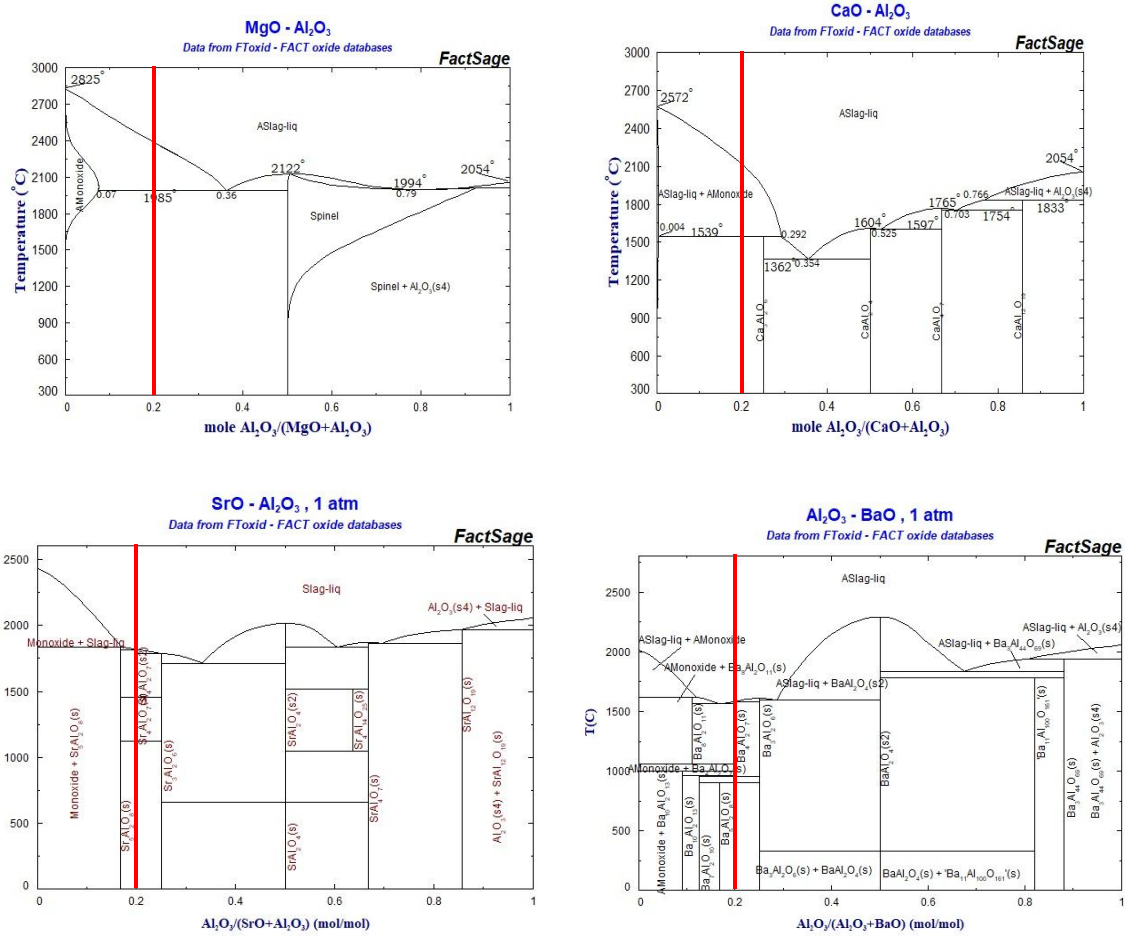


ICSD-No. 150769

$M_4Al_2O_7$

Fig.(a) Schematic daigram of $Ba_3CaAl_2O_7$(ss) initial $M_4Al_2O_7$

$M_4Al_2O_7$ phase



- $4MO \cdot Al_2O_3$ theoretical phase
- $MgO\text{-}Al_2O_3$: $MgO, MgAl_2O_4$

  $CaO\text{-}Al_2O_3$: $CaO, Ca_3Al_2O_6$

  $SrO\text{-}Al_2O_3$: $Sr_4Al_2O_7$

  $BaO\text{-}Al_2O_3$: $Ba_4Al_2O_7$
- $4MO \cdot Al_2O_3$ experimental results

  $MgO\text{-}Al_2O_3$: $MgO, MgAl_2O_4$

  $CaO\text{-}Al_2O_3$: $CaAl_2O_4, Ca_3Al_2O_6$

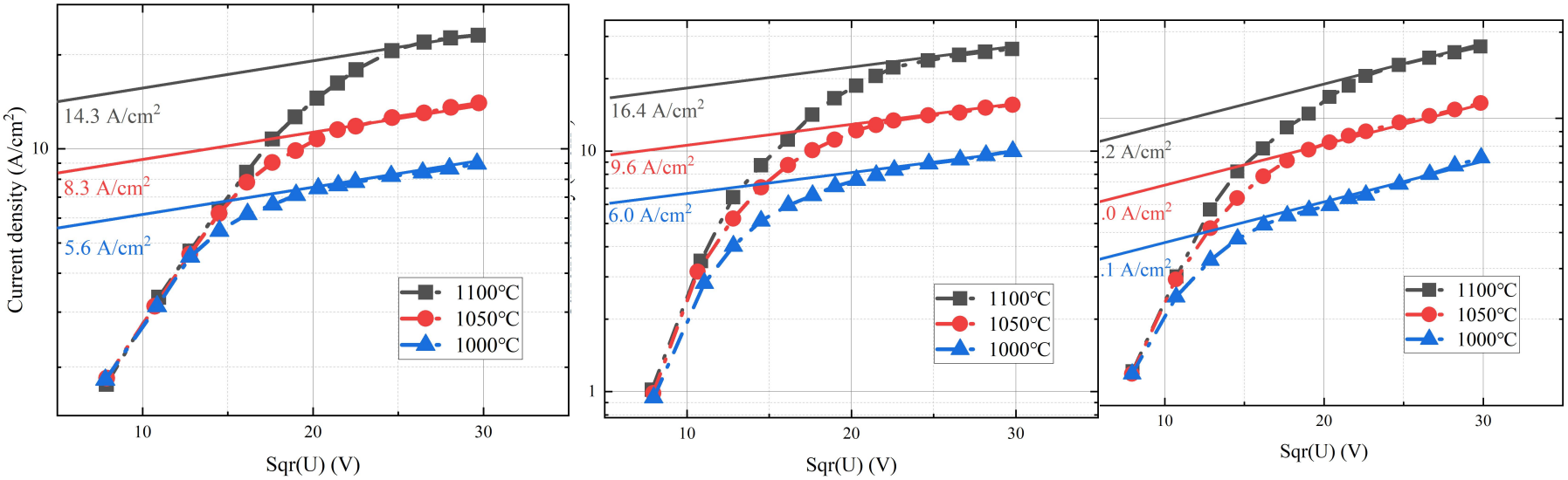
  $SrO\text{-}Al_2O_3$: $Sr_4Al_2O_7$

  $BaO\text{-}Al_2O_3$: $Ba_4Al_2O_7$

Fig. $MO \cdot Al_2O_3$ phase diagram

Sr and Ba exist as $M_4Al_2O_7$

Electron emission property of the cathodes impreganated with Sr addition amount



(a) 0Sr (b) 0.6Sr (c) 1Sr

- Emission property： 0.6Sr-W > 0Sr-W > 1Sr-W
- Contribution of the main phase in cathode electron emission：

$(Ba, Sr, Ca)_4Al_2O_7 > Ba_3CaAl_2O_7(ss) > (Ba, Sr)_4Al_2O_7$

Appropriate amount of Sr addition in the alluminates could increase the emission property of the cathode

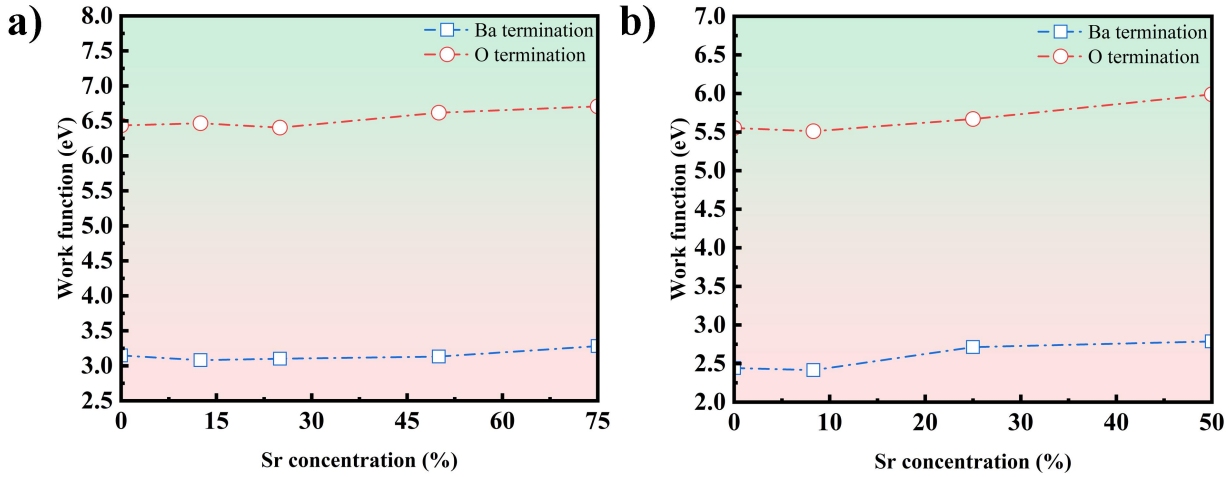
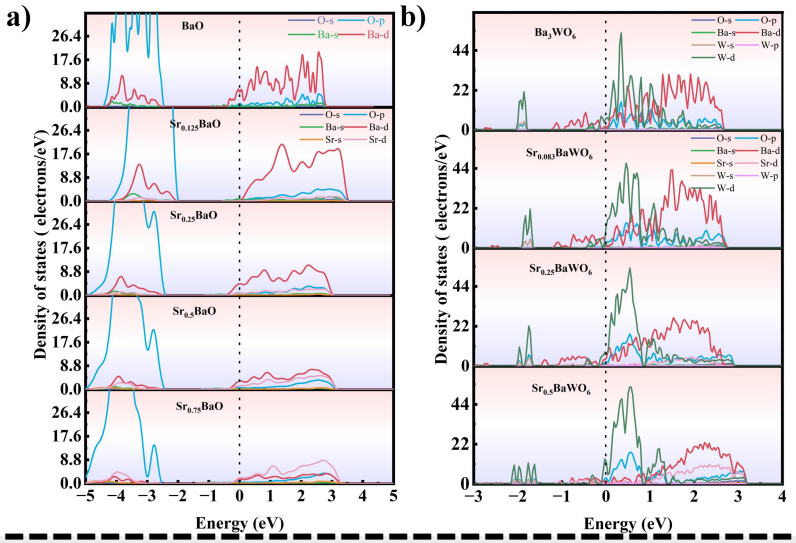
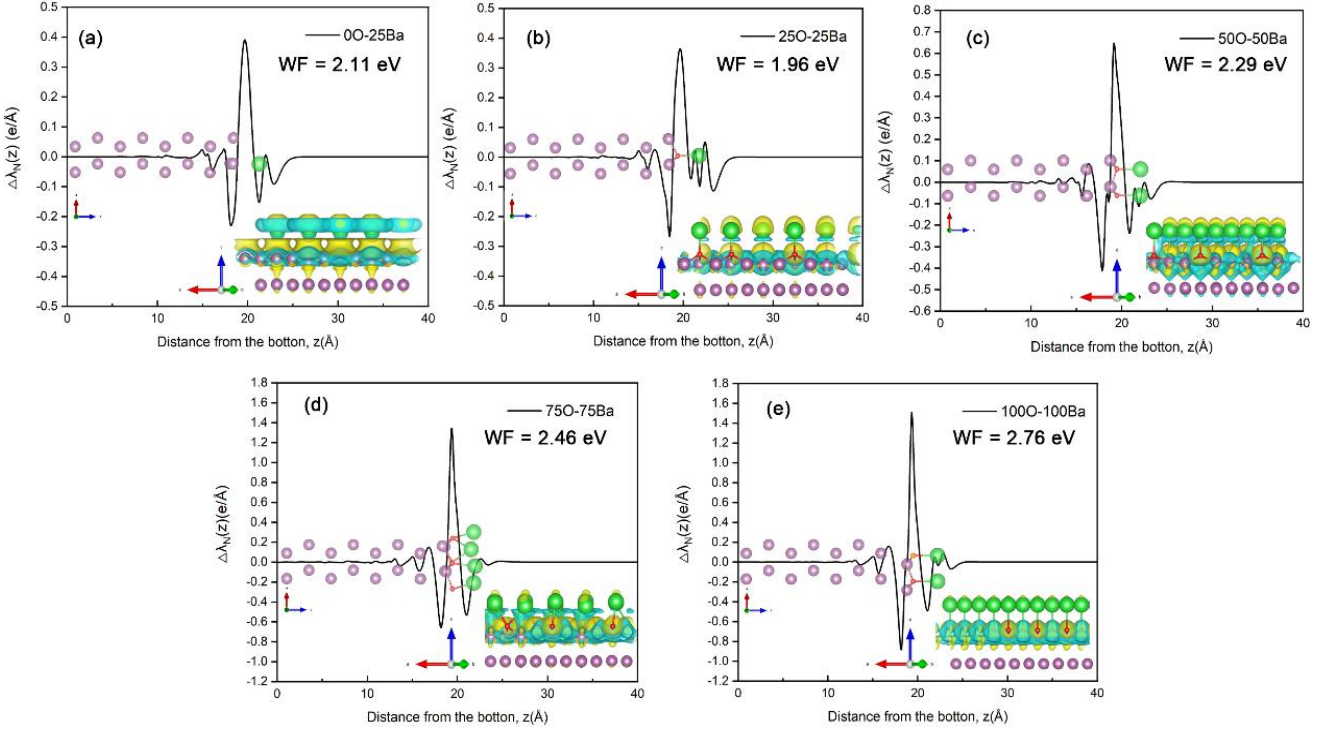


Fig. Work function of the addtiion of Sr in (a) BaO and (b) Ba_3WO_6



- **The addition of 12.5%Sr could reduce the work function of BaO**
- **8.3%Sr, The work function of Ba in Ba_3WO_6 is the lowest 2.415 eV**



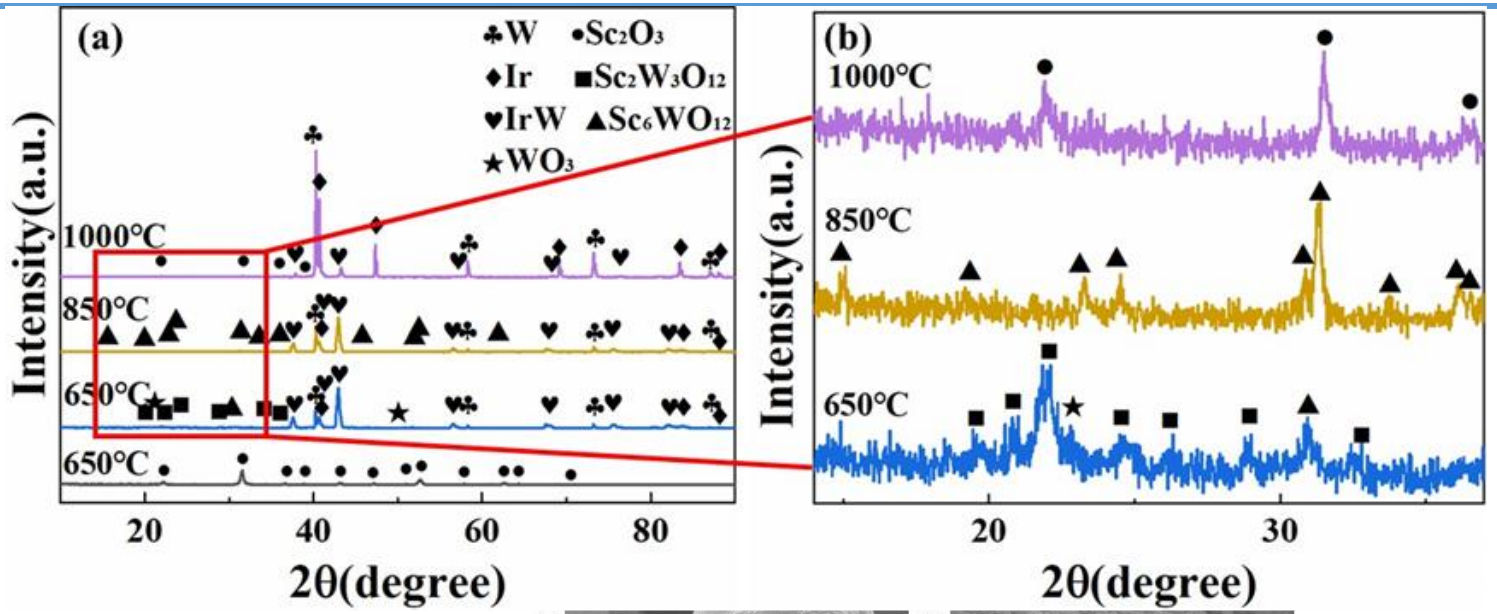
The curve of charge density difference ΔλN(z) for the adsorption of Ba_x-O_y on the Sc (002) surface with coverage of (a) 0%O-25%Ba, (b) 25%O-25%Ba, (c) 50%O-50%Ba, (d) 75%O-75%Ba and (e)100%O-100%Ba

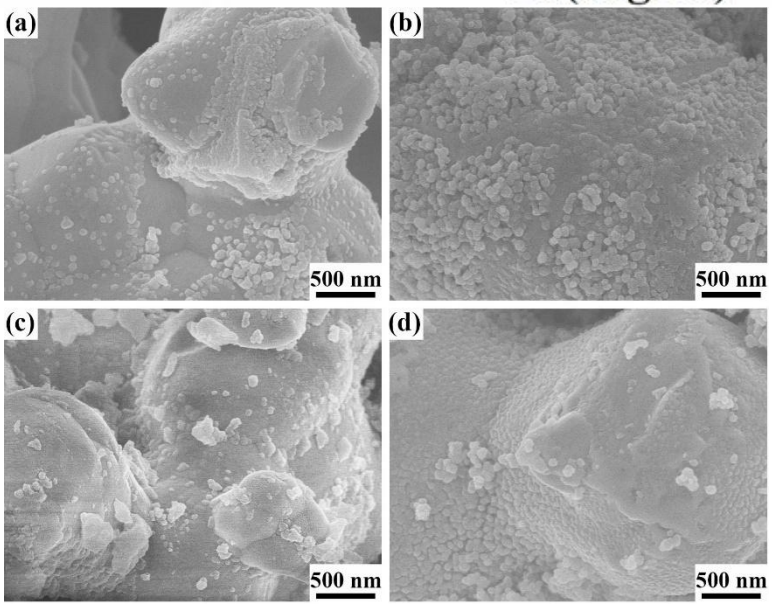
Table 1 The dipole moment $\Delta\mu_N$ of Ba_x-O_y absorbed on Sc (002)

Sc (002)-Ba_xO_y	Work Function φ(eV)	Dipole moment changes $\Delta\mu_N$ (D)
0%O -25%Ba	2.11	0.309
25%O -25%Ba	1.96	0.351
50%O -50%Ba	2.29	0.289
75%O -75%Ba	2.46	0.215
100%O - 100%Ba	2.76	0.196

Ir-W matrix scandate cathode

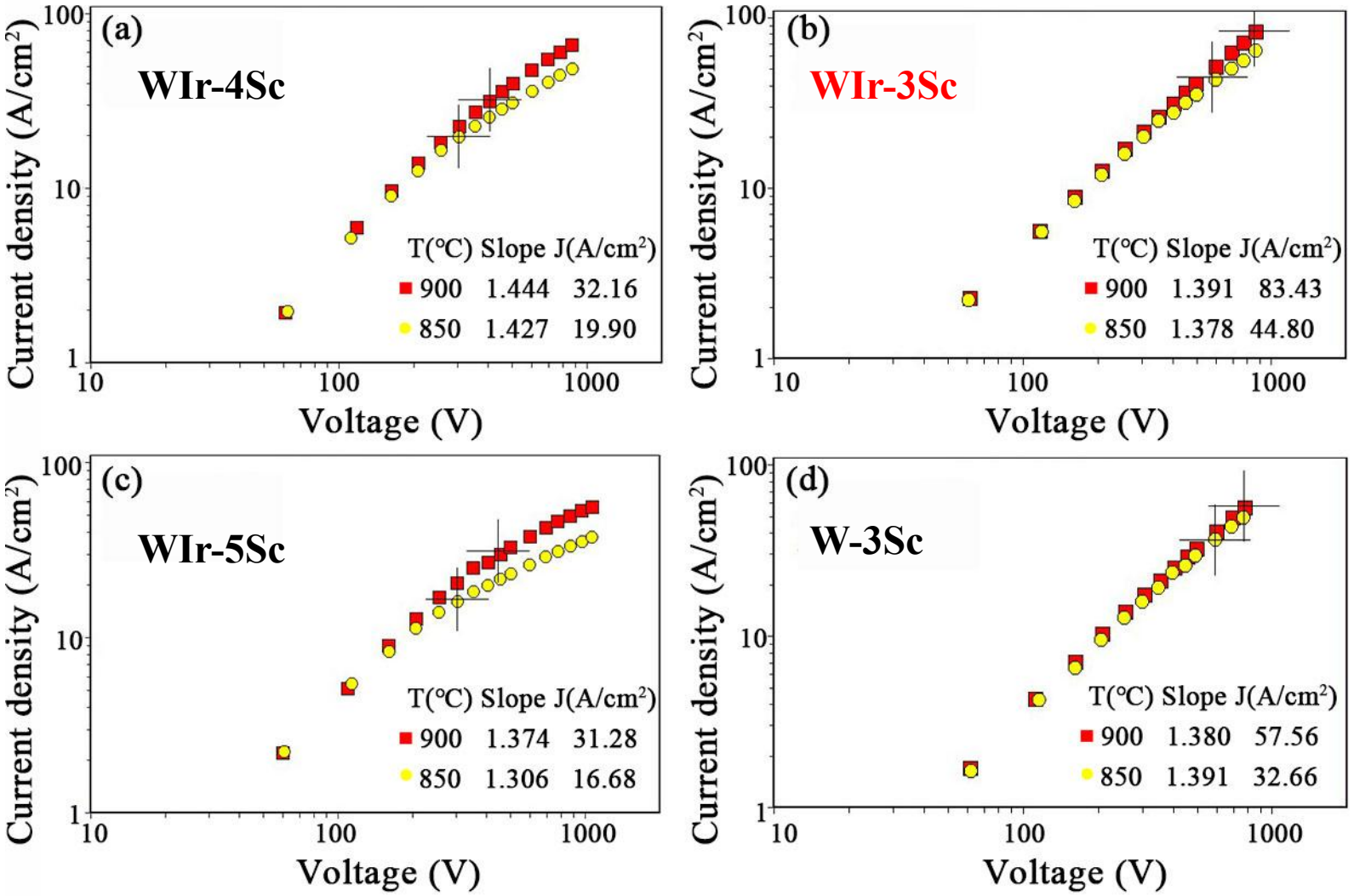
W-Ir scandate cathode



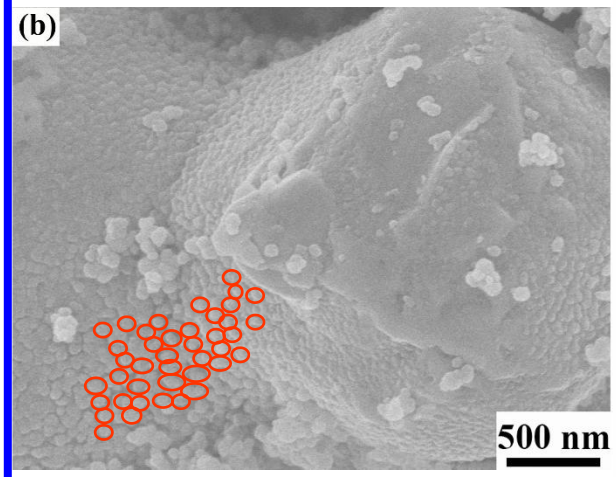


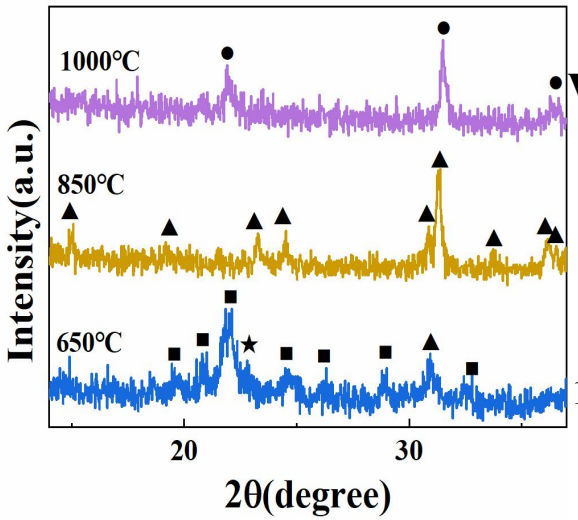
Sc_2O_3 nanoparticles grown on Ir and W powder surfaces synthesized by one step reduction

W-Ir scandate cathode



The electron emission current density could reach 83.43A/cm^2





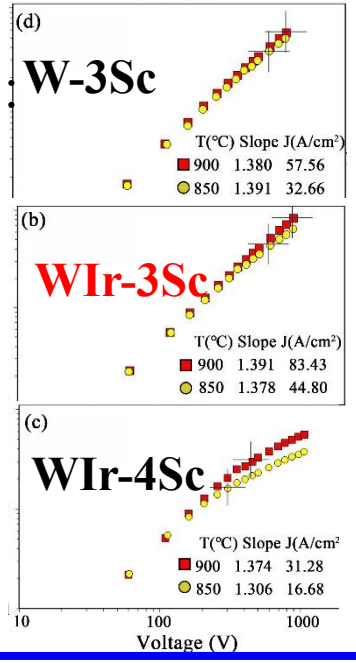
◆ Phase transformation:

W+Sc(NO$_3$)$_3$

650 ℃ → $Sc_2W_3O_{12}$

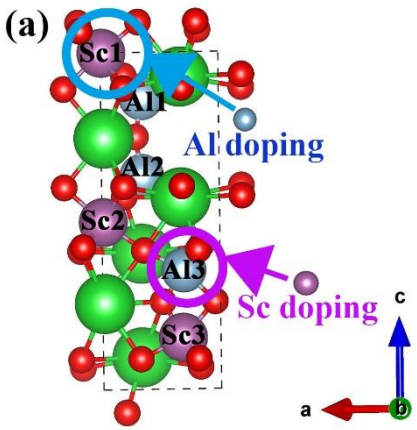
850 ℃ → Sc_6WO_{12}

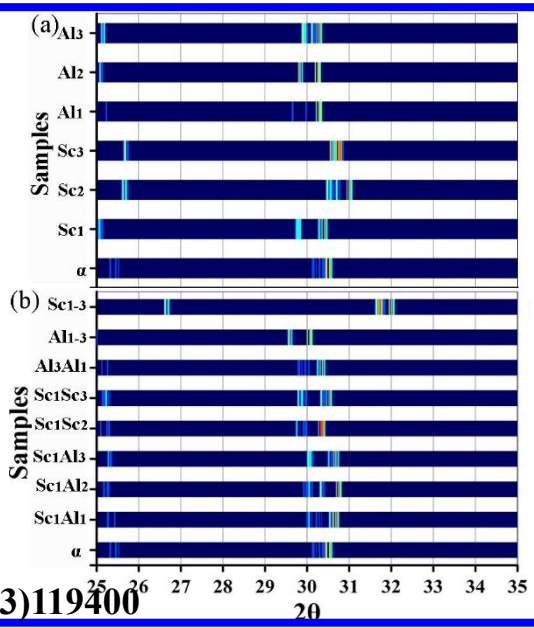
1000 ℃ → Sc_2O_3+W



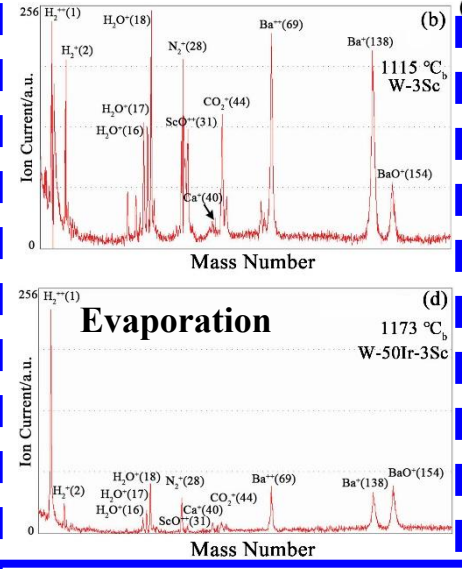
Powders grown on Ir and W surfaces of Sc_2O_3 nanoparticles synthesized in-situ in one step

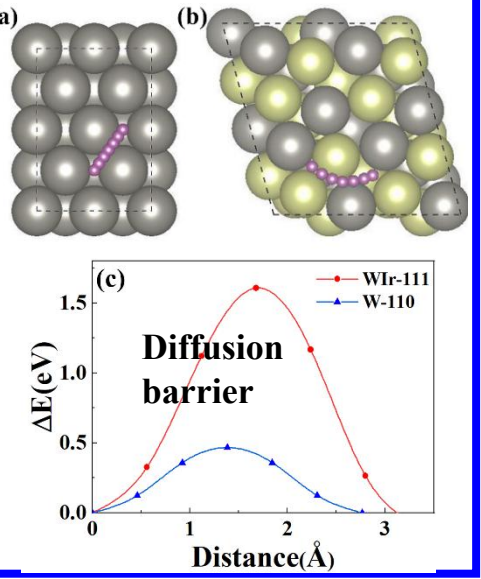
◆ Sc: active phase





◆ Ir stabilizes active substance





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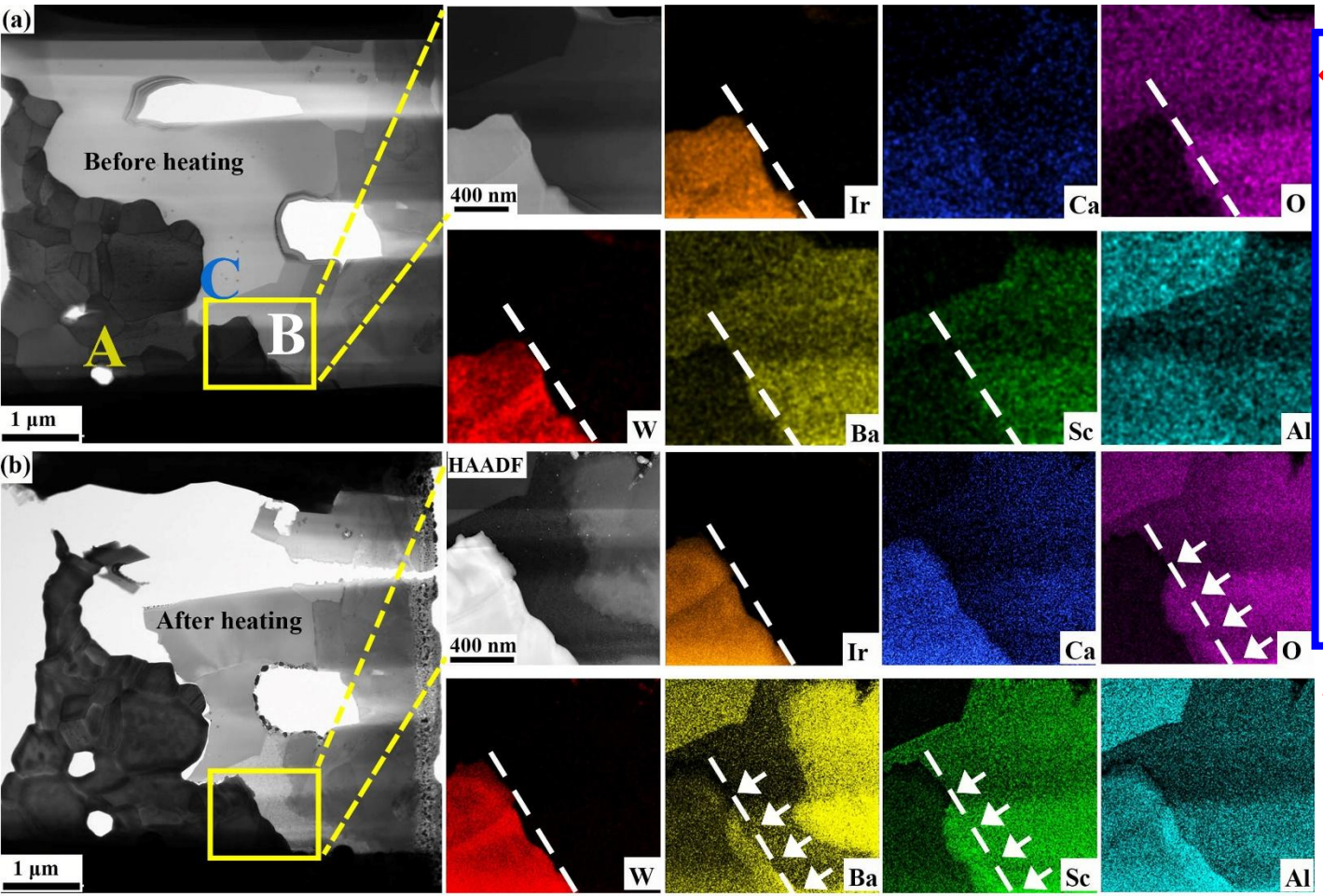
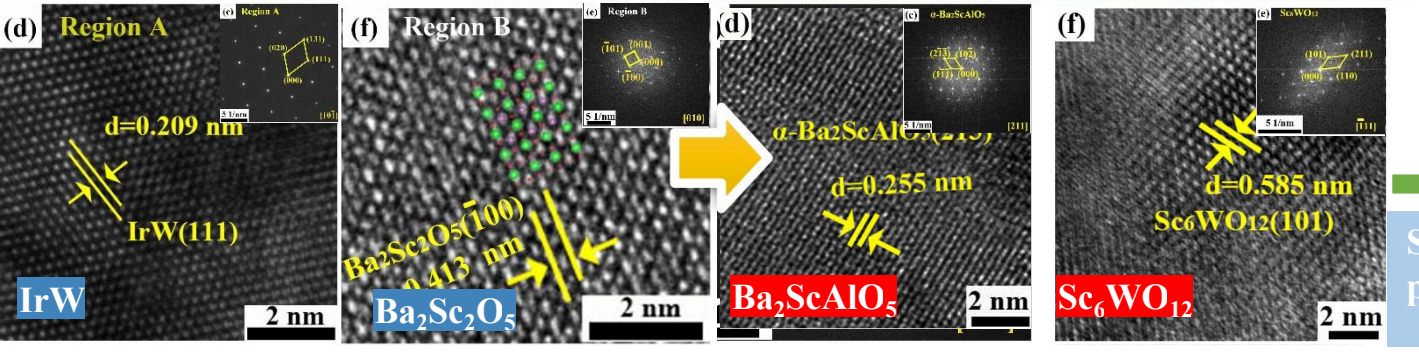


◆ **Deduction**：

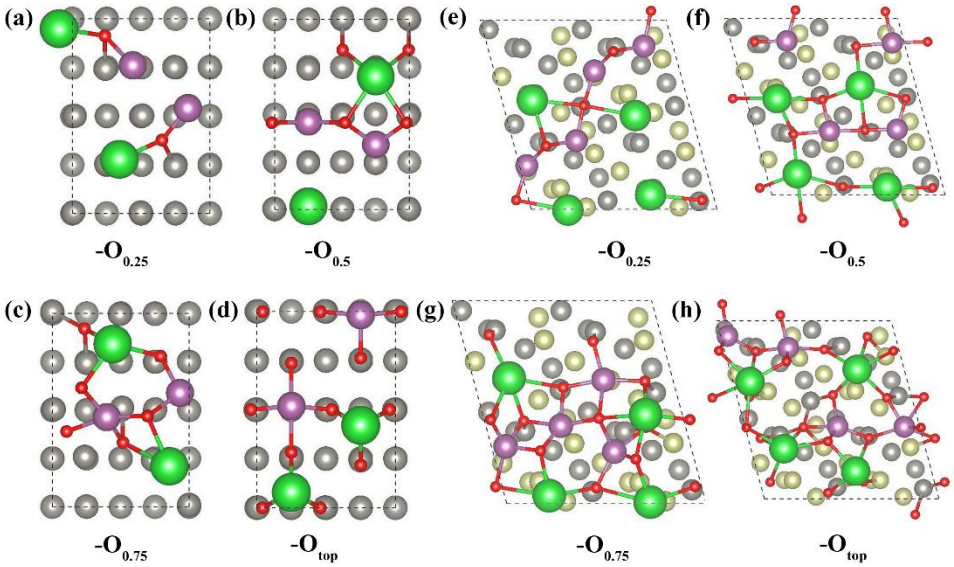
$2BaO+Al_2O_3+Ba_2Sc_2O_5 \rightarrow 2\alpha\text{-}Ba_2ScAlO_5$ (1)

$6Ba_2ScAlO_5+W \rightarrow Sc_6WO_{12}+3Ba_3Al_2O_6+3Ba$ (2)



◆ **Design**

- Emission mechanism
- Characte-rization
- Ir、Sc roles
- Performance
- Synthesized powder



$Ba_{0.25}$-$Sc_{0.25}O_x$ adsorpted on W and IrW

Table 3 Adsorption energy and formation energy

Crystal index	W (1 1 0)		IrW (1 1 1)	
	E_{ads} /eV	ΔE_F /eV	E_{ads} /eV	ΔE_F /eV
-$Ba_{0.25}$-$Sc_{0.25}$-$O_{0.25}$	-4.89	-2.00	-5.88	-12.22
-$Ba_{0.25}$-$Sc_{0.25}$-$O_{0.5}$	-6.30	-8.73	-5.63	-19.16
-$Ba_{0.25}$-$Sc_{0.25}$-$O_{0.75}$	-6.97	-13.21	-8.58	-28.74
-$Ba_{0.25}$-$Sc_{0.25}$-O_{top}	-7.63	-12.26	-8.41	-29.47

◆**Adsorption energy**： IrW-$Ba_{0.25}$-$Sc_{0.25}$-$O_{0.75}$ < W-$Ba_{0.25}$-$Sc_{0.25}$-O_{top}

◆Transferred electrons of Sc(1.88 *e*) and Ba(1.45 *e*) in IrW-$Ba_{0.25}$-$Sc_{0.25}$-$O_{0.75}$ is higher than Sc(1.44 *e*) and Ba(1.2 *e*) in W-$Ba_{0.25}$-$Sc_{0.25}$-O_{top}, and Sc plays a major role

◆Ir makes the interaction between the active substance and the matrix stronger, stabilizes the active substance, and improves the emission performance of the cathode.

Conclusion

1. $(Ba, Sr, Ca)_4Al_2O_7$ could improve the emission property of the impregnated cathode with the addition of apporirate amount of Sr in the Ba,Ca aluminates.
2. The calculation shows that properly increasing the concentration of oxygen on the Sc surface can reduce the work function.
3. Ir-W scandate cathode could provide the emission current density of 83A/cm^2 at 900^{o}Cb.
4. More transferred electrons of Sc(1.88 *e*) and Ba(1.45 *e*) in the cathode could benefit for cathode performance. Ir stabilizes the active substance on the cathode surface.

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……

Thanks for your kind attention