

**The 8th ITG International Vacuum Electronics Workshop 2022
September 1 - 3, 2022; Physikzentrum, Bad Honnef, Germany**



Surface electron emission model of Ba dispenser cathodes, especially of Ba scandate cathodes

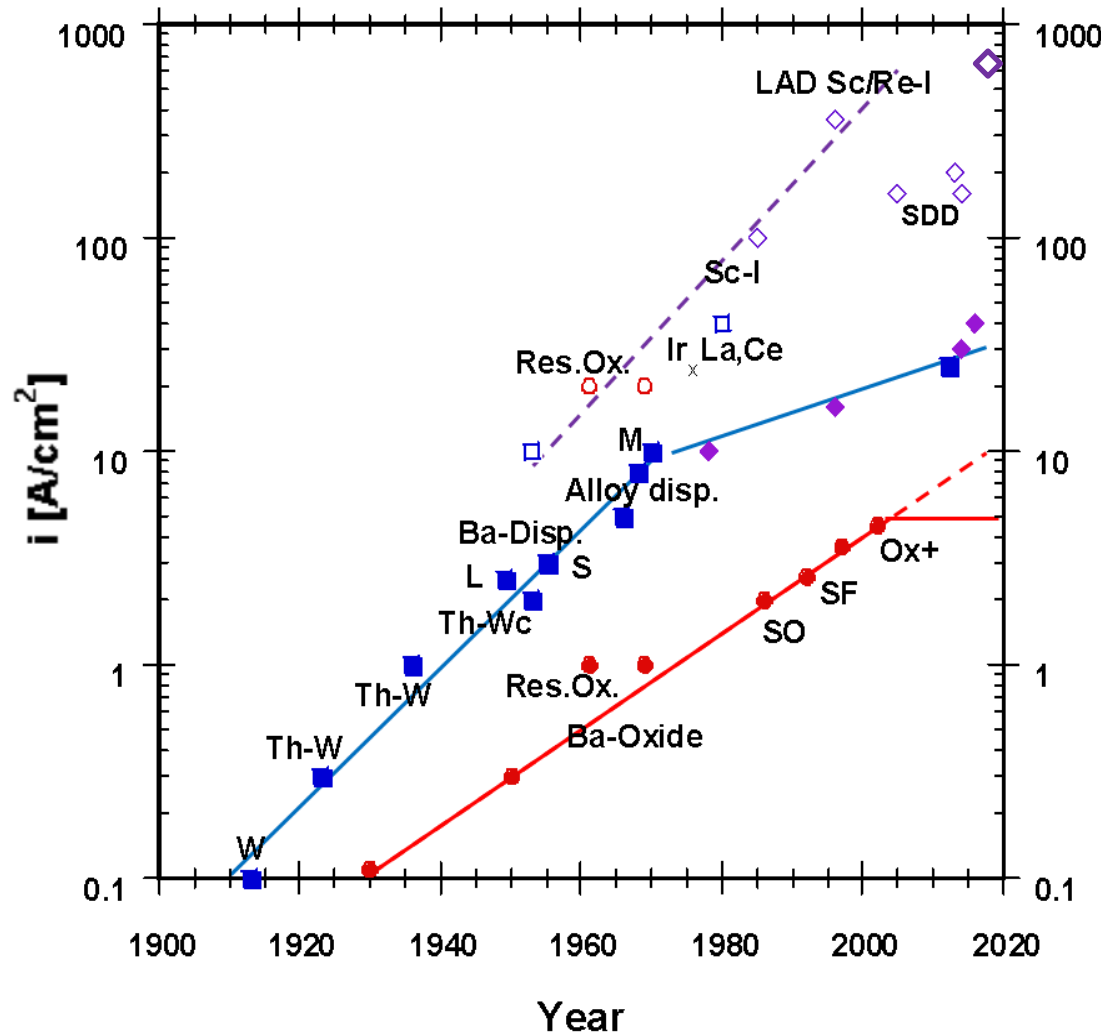
**Georg Gaertner, Consultant, Aachen, Germany
ITG-IVEW Session 1, 2.9.2022**

Surface electron emission model of Ba dispenser cathodes, especially of Ba Scandate cathodes

Outline:

- 1. High emission capability of Ba Scandate cathodes**
 - a) anomalous behavior in „saturation“**
 - b) comparison with I cathodes**
- 2. Evaluation of Richardson plots of I and Scandate cathodes**
- 3. Surface analysis results for I and Scandate cathodes**
- 4. The beamlet effect (J. Hasker)**
- 5. Model of Scandate cathode emission**
- 6. Conclusions and outlook**

1a) Progress in cathode performance over time



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Historical development of thermionic cathode emission capabilities - an update.

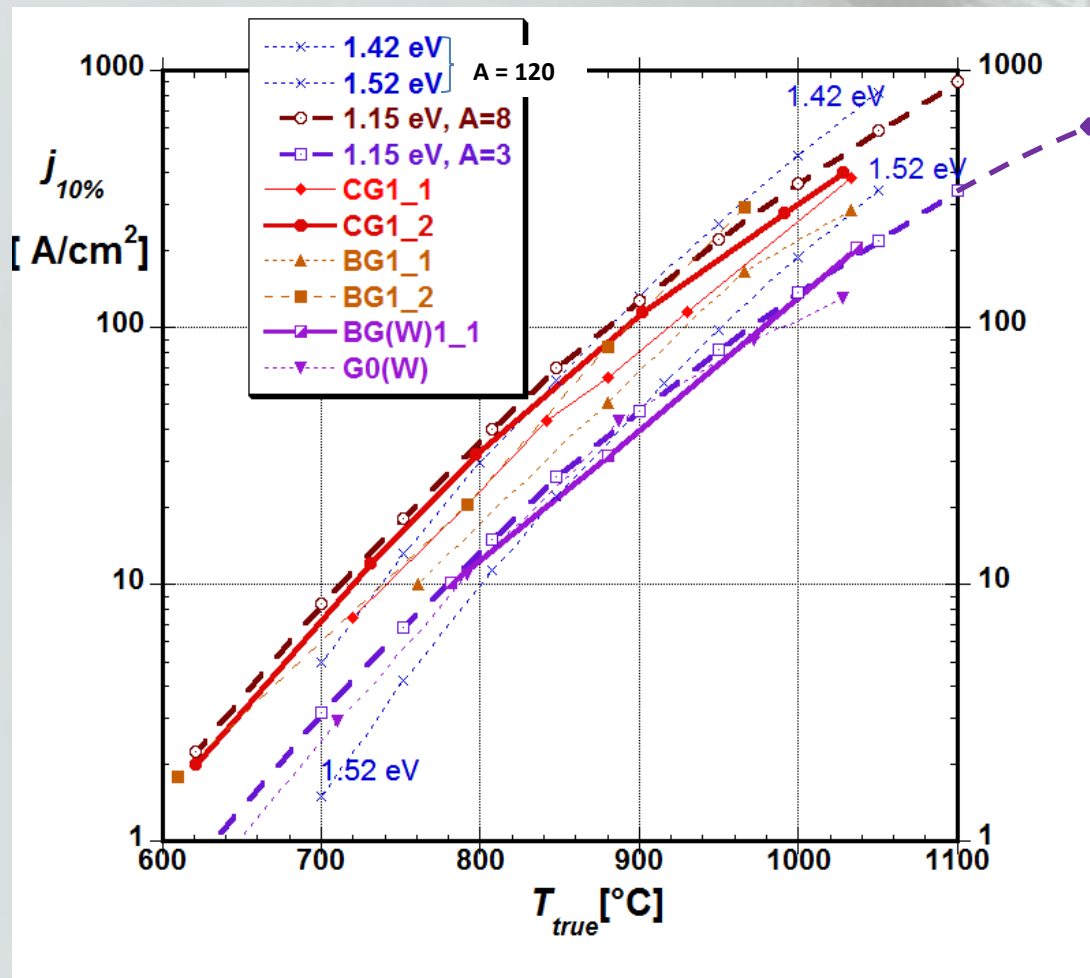
The lower red line : dc emission of oxide cathodes (red circles). The upper blue line: metal matrix based (including Ba dispenser) cathodes; Scandate cathodes: violet diamonds.

Open symbols + dashed line: pulsed emission data including Scandate cathodes.

It illustrates the high emission capability of Scandate cathodes

Current top result 670 A/cm² pulsed 20 Hz, 5 μ s, $T = 1100^\circ\text{C}_b$ of S. Yin 2020

1b) Saturation emission current density $j_s \approx j_{10\%}$ versus true temperature of LAD top-layer Scandate cathodes



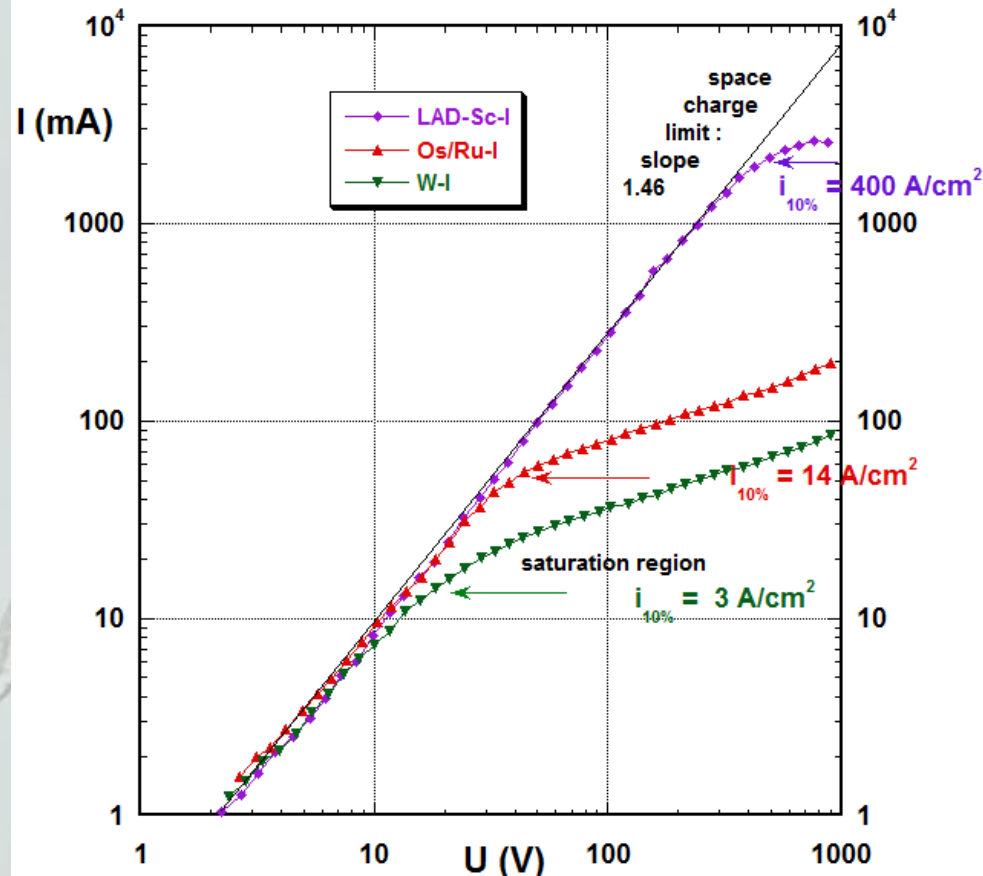
Red and orange curves: LAD with Re (on W)

Violet curves: LAD with W

The violet (check) data point outside the scale is the top result of Shengyi Yin from 2020, compatible with W and $A_R = 3$ at about 1190 °C true temperature

A general feature of all Ba-scandate cathodes prepared by LAD is the **work function** of about **1.15 eV** (also determined by FERP), whereas the Richardson constant A_R is varying between 3 and 8 A/(cm²K²). For comparison the saturated emission density j_s curves for work functions of 1,52 eV and 1,42 eV with $A = 120,4$ are also shown

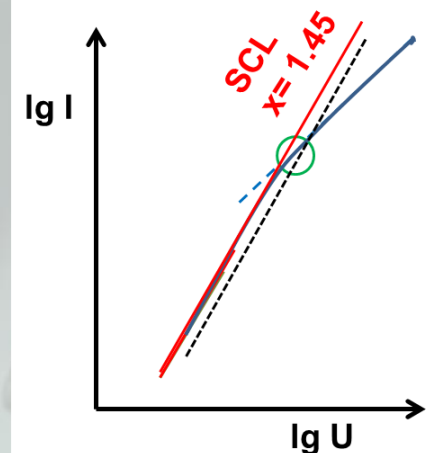
1c) I/U characteristics of Ba dispenser + Scandate cathodes



Philips 0,65 Watt I cathode unit as used in the tests

Current-voltage characteristics of:
 LAD top-layer Scandate cathode (LAD-'Sc'/Re-I),
 Os/Ru-I cathode and W-I cathode
 at 965°C Mo-brightness temperature (1030°C true temp.), as determined in the diode mode in a Philips 45AX electron gun

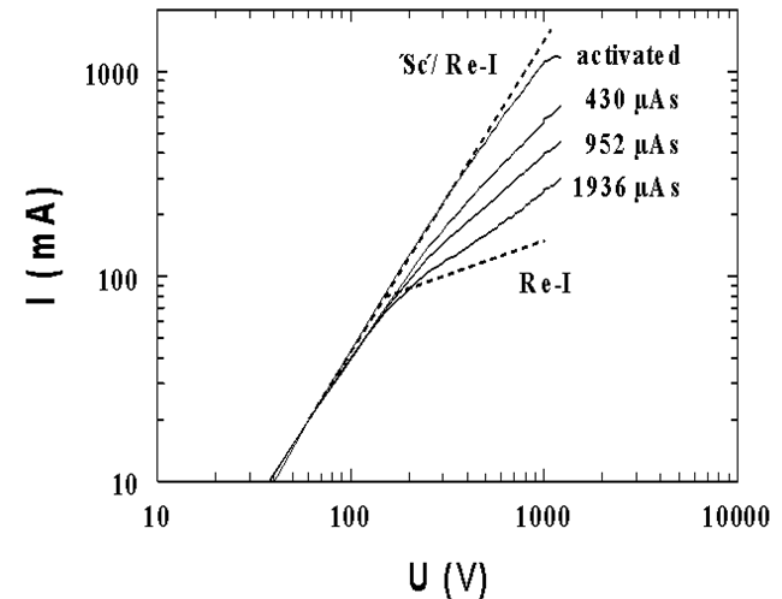
Determination of zero field emission current density $i_{10\%}$



1e) Ba Scandate cathode I/U characteristics:

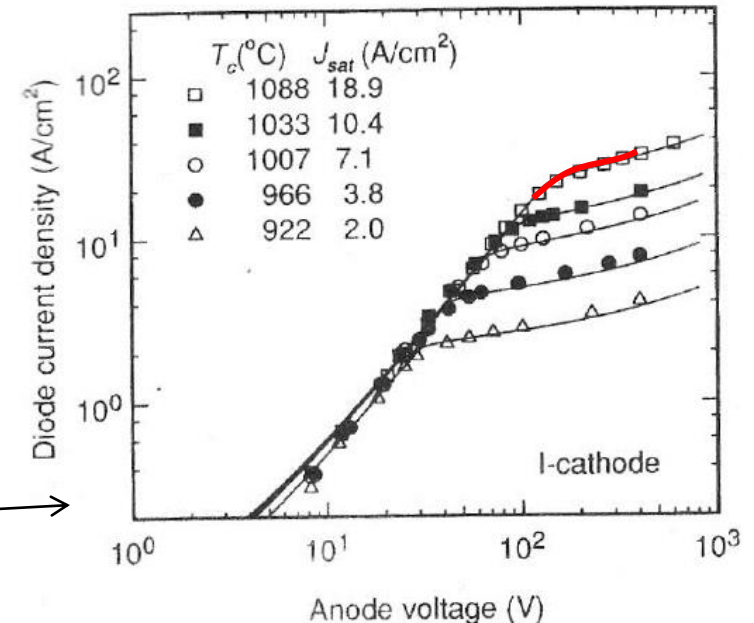
- **anomalous Schottky behavior** (stronger increase in “saturation”) similar to **oxide cathodes**: the work function $e\Phi_R$ is modified by field penetration into the semiconductor and by space charge of ionized donors. The image charge is smaller and dependent on ϵ
- “pseudo space charge behavior” of $\lg I - \lg U$ characteristics: deviation from SCL **power law** $I = g U^x$, $x \leq 1.4$ observed. The slope x is changing during activation, life and during ion bombardment (=IB).

Decrease of slope x with increasing IB



1f) Ba dispenser (I-) cathode I/U characteristics:

- theoretical description of **transition range between SCL and Schottky** saturation also for I cathodes missing, which is needed for j_0 (Schottky extrapolations not reliable)
- Transition region described by **Scott [4]** by **numerical calculations** and by **Hasker via an approximation [3]**, see Manenschijn [1]



2. Evaluation of Richardson plots of I and scandate cathodes

For saturated (zero field) thermionic emission current density as a function of true temperature T the **Richardson equation** holds:

$$j_s = A_{th} * T^2 \exp (- e\Phi / kT) \quad (1)$$

with j_s the saturated emission current density, $e\Phi$ the work function and $A_R = A_{Th} = 120.4 \text{ Acm}^{-2}\text{K}^{-2}$ the thermionic constant for uniform $e\Phi$, e.g. a pure metal.

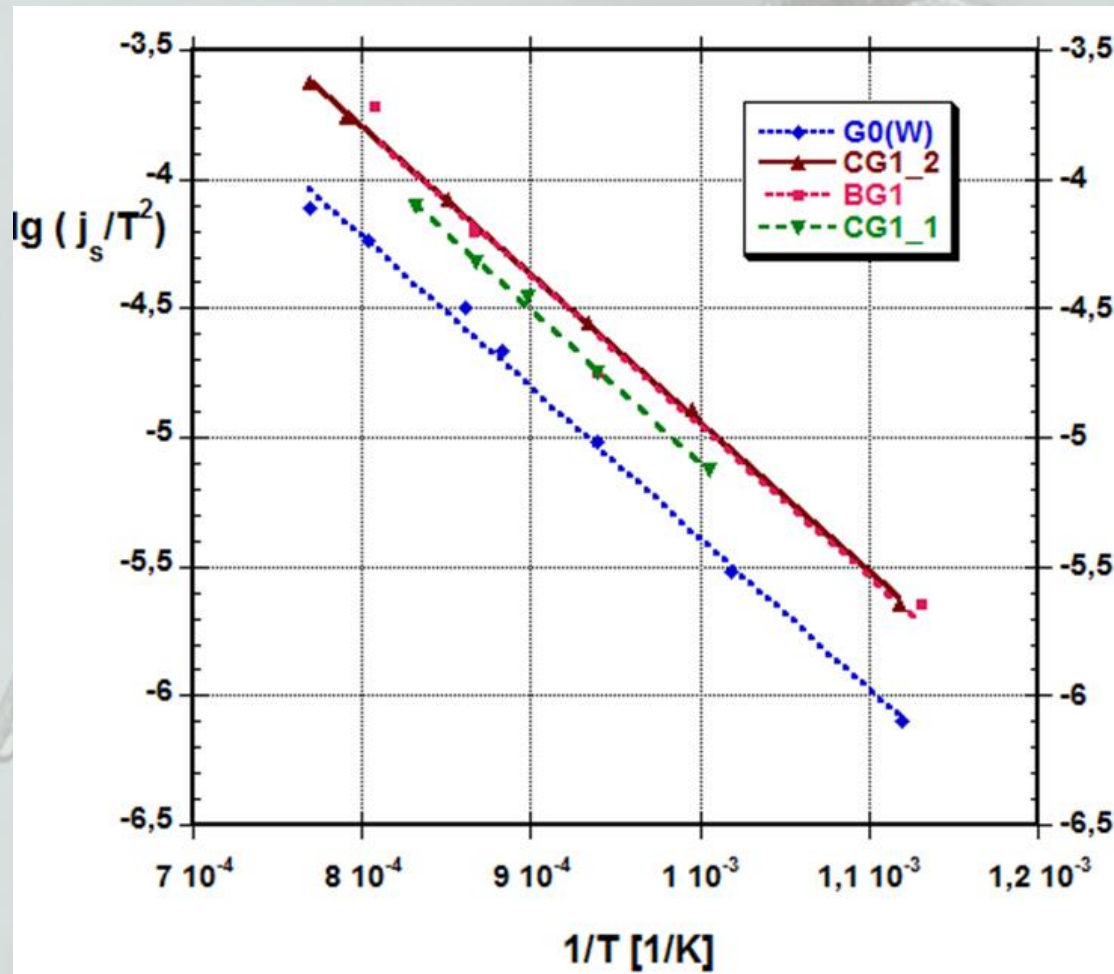
In its more heuristic practical form it is written as

$$j_s = A_R * T^2 \exp (- e\Phi_R / kT) \quad (2)$$

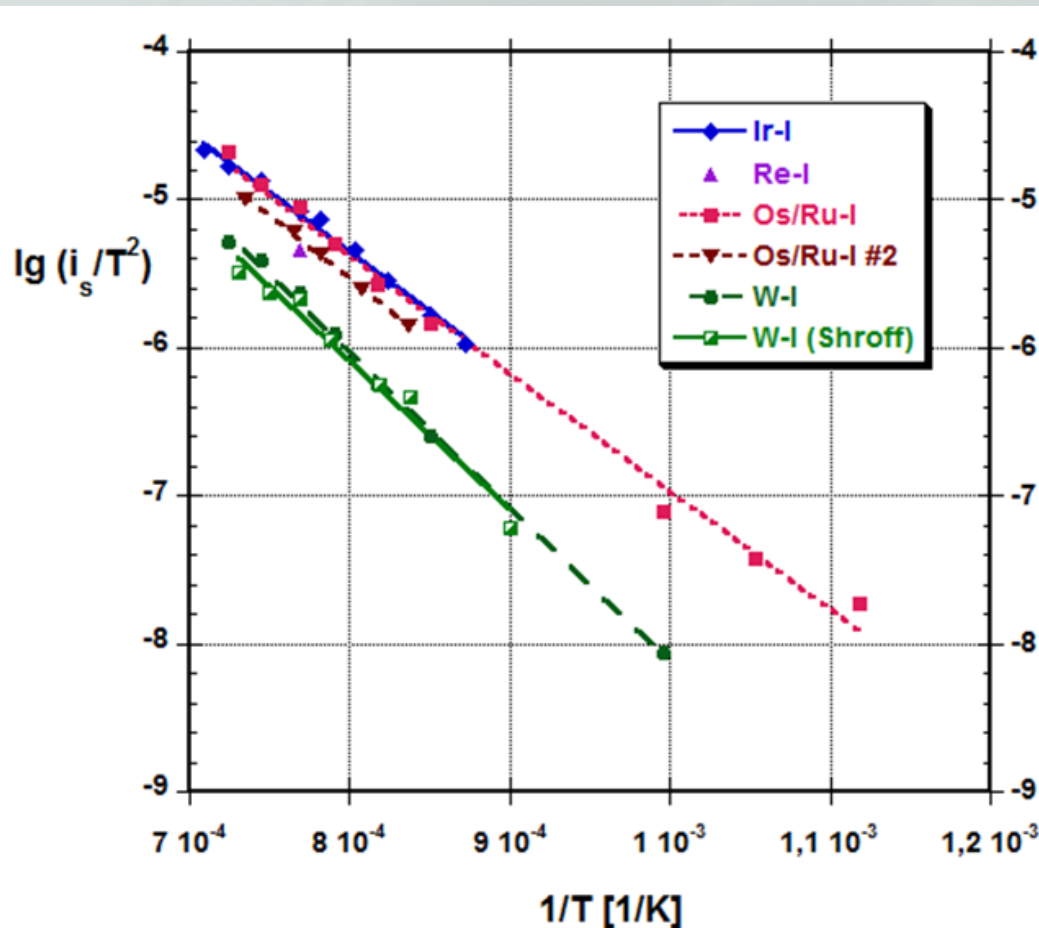
With A_R the fitted Richardson constant and $e\Phi_R$ the Richardson WF; They can be obtained from a Richardson plot of $\lg(j_s / T^2)$ versus $1/T$

$$\lg(j_s / T^2) = \lg A_R - e\Phi / (k * \ln 10) * 1/T \quad (3)$$

The Richardson constant A_R can also be expressed as a temperature dependence of work function $e\Phi = e\Phi_R + \alpha * T$, with the coefficient $\alpha = k * \ln (A_{th} / A_R)$. This of course only makes sense for uniform surfaces.



Richardson Plot of LAD top-layer scandate cathodes G_0 (without 411x layer, no Re), CG_{1_1} ($x=1/4$), CG_{1_2} ($x=1/4$), and BG_1 ($x=1/2$); x is the portion of Sc_2O_3 added to $4Ba.Ca.Al_2O_3$ target



Richardson plots of different Ba dispenser (I-) cathodes

From the measured dependence of j_s on true temperature T one can determine work function and Richardson constant from a so-called Richardson plot [1] of $\lg(j_s/T^2)$ versus $1/T$:

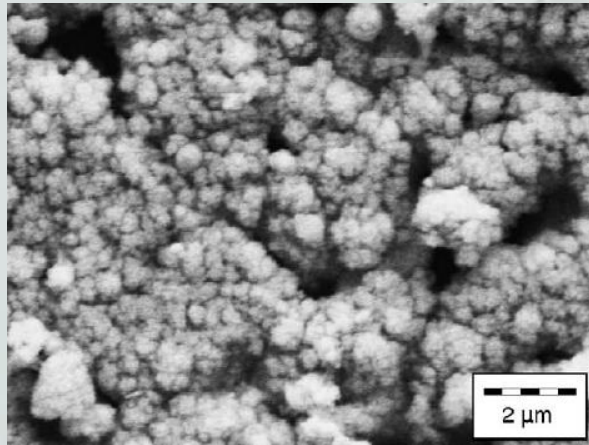
$$\lg(j_s/T^2) = \lg A_R - e\Phi/(k \cdot \ln 10) \cdot 1/T$$

Fitted results of Richardson plots for LAD Scandate cathodes and Ba dispenser cathodes

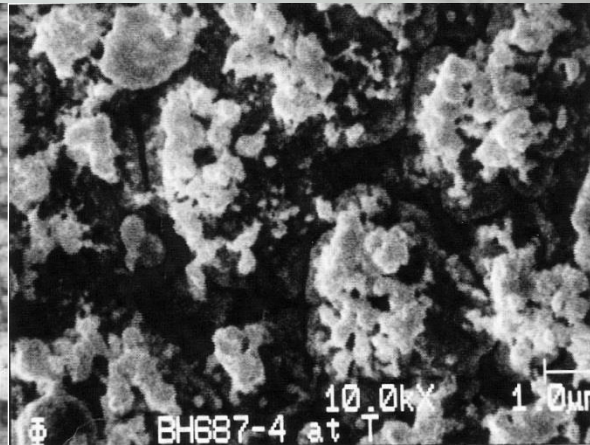
Ba dispenser cathode type	Reference	Richardson work function $e\Phi_R$ [eV]	Richardson constant A_R [A/(cm ² *K ²)]
LAD Scandate BG ₁ (‘Sc’/Re-I) LAD Scandate Sc/W-I	G. Gaertner et al., see [1]	1.15 1.16	7.0 2.8
Os/Ru-I (M)	G. Gaertner et al., [1]	1.58	10.5
Os/Ru-I (M), #2	A. Manenschijn et al., see [1]	1.71	22.1
Ir-I (M)	P. Geittner et al., [1]	1.60	13.0
W-I (S)	G. Gaertner et al., [1]	2.08	178
W-I (S)	A. Shroff et al., 1980*	2.01	102.6

In conclusion, **W-I cathodes are rather uniform with good coverage of Ba on (-O-) W** and A_R is in the order of A_{Th} . The Os or Ir coated cathodes seem to be less uniform, but with lower WF, whereas **Sc-I** are characterized by **low coverage correlated with Sc (+Ba) spots**

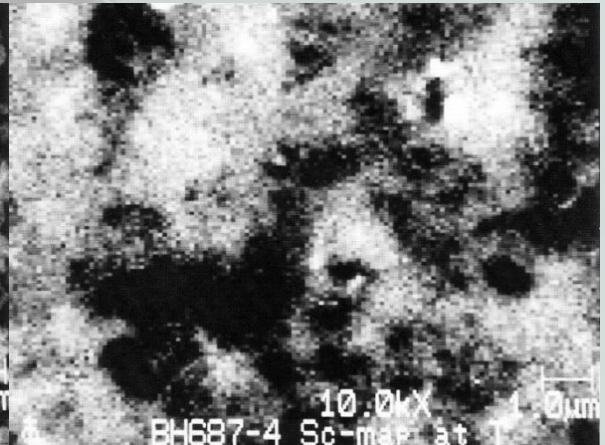
3) Structural and compositional characterization of LAD Scandate cathodes (on I bases)



a) after LAD



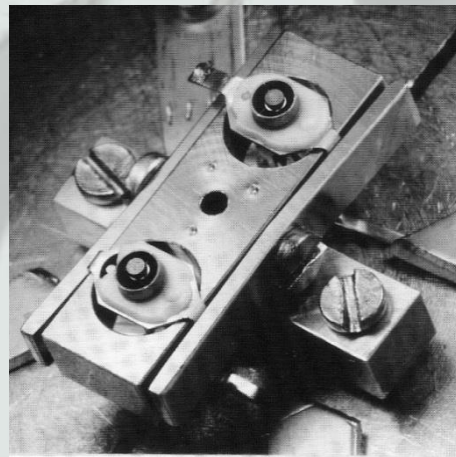
b) after activation



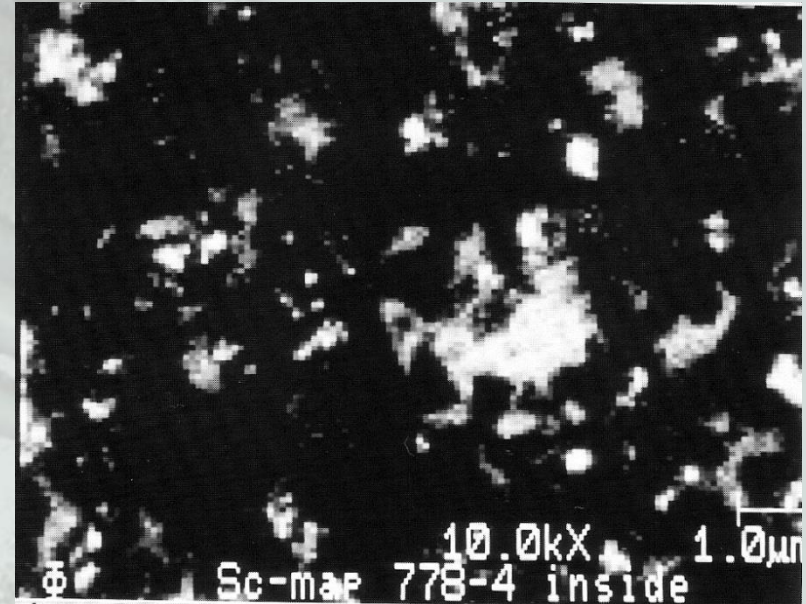
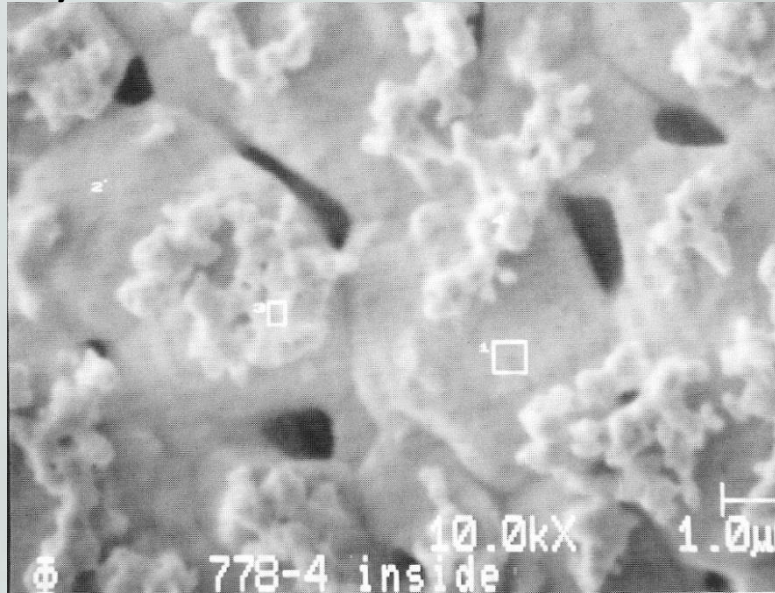
c) Auger Sc-mapping

LAD Ba-scandate cathode SEM micrographs (G. Gaertner et al.)

In order to overcome the Problems of transferring the cathodes from test chamber to the SEM /AES chamber, a mount with 2 I cathode units and heating connections was designed to do **AES at operating temperature**, avoiding contamination



3b) Structural and compositional characterization of LAD Scandate cathodes (on I bases)



SEM micrograph (above) of LAD top layer Scandate cathode in the central beam emitting region **after ion bombardment**; the cathode was re-activated for 30 min at 1060°C Mo-brightness and then kept at 965°C Mo-Br. during SAM.

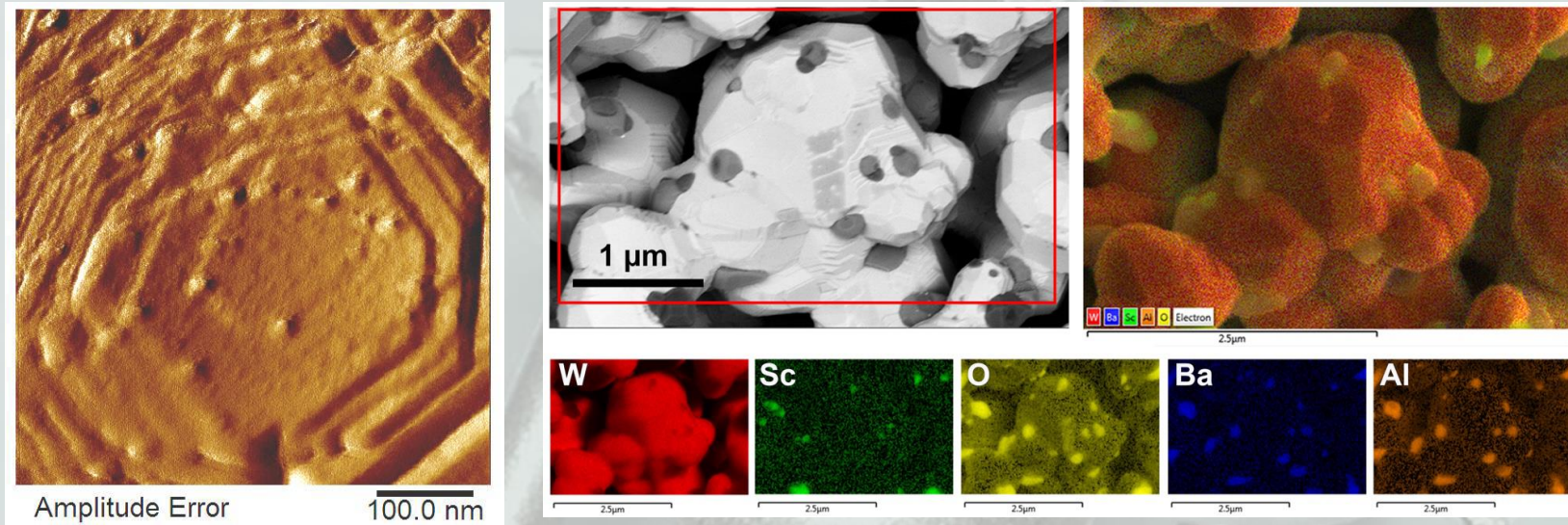
Top right: AES map of Sc

Bottom right: AES map of Ba

Typical: Islands of Sc and overall distribution of Ba.



3c. Further surface analysis results for Scandate cathodes



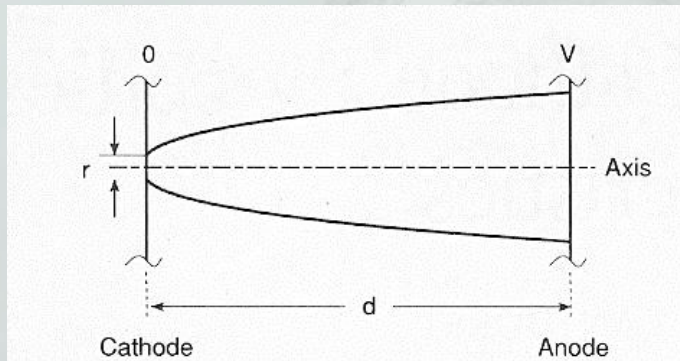
SDD Cathode (Y. Wang et al., J. Balk et al.); separate Sc containing **nanoparticles; somewhat in contradiction to Auger depth profiles**

From the Auger maps above one can derive some average values

- For LAD Scandate cath.: ($2 \cdot r_{sp}$) average diameter of the Sc patches; range 100 nm – 1 μ m (d_{sp}) average distance of the patch centers 0,5 - 2 μ m (average grain size 3 – 5 μ m)
- For SDD Scandate cathodes: ($2 \cdot r_{sp}$) average diameter of the Sc patches; 100 – 300 nm (d_{sp}) average distance of the patch centers 1 - 2 μ m (average grain size 1 – 3 μ m)

There is the paradox situation, that Scandate cathode show unprecedented high emission capability, but evidently only from small spots of a Ba, Sc and O compound, which could not be explained so far.

4. The beamlet effect (J. Hasker, 1985)



Application to scandate cathodes:
In the highly emitting patches beamlets originate. In German: „die verteilten Spots mit niedriger Austrittsarbeit emittieren ein Bündel lokalisierter Einzelstrahlen“.

Model: Planar diode with one circular patch having space-charge limited emission [4];
With current density in the beamlet j_b
and Child-Langmuir current density j_{CH}

$$\rightarrow j_b = (d_{ca}/r_{sp})^{1/2} * j_{CH} \quad (4)$$

(based on scaling laws for Poisson equation)

Example Ba diffusion from the pores: If λ is the diffusion length \rightarrow Current

$$I = \pi * \lambda^2 * j \quad (5)$$

See also J. Hasker, N. van Hijningen, Appl. Surf. Sci. 24, (1985), 318 – 329 [5]

The current density in the beamlet or micro beam at the spot-surface is much higher than expected from the Child-Langmuir space charge law!

E.g. for $d_{ca} = 250 \mu m$ and $r_{sp} = 100 nm$

$$j_b = 50 * j_{CH}$$

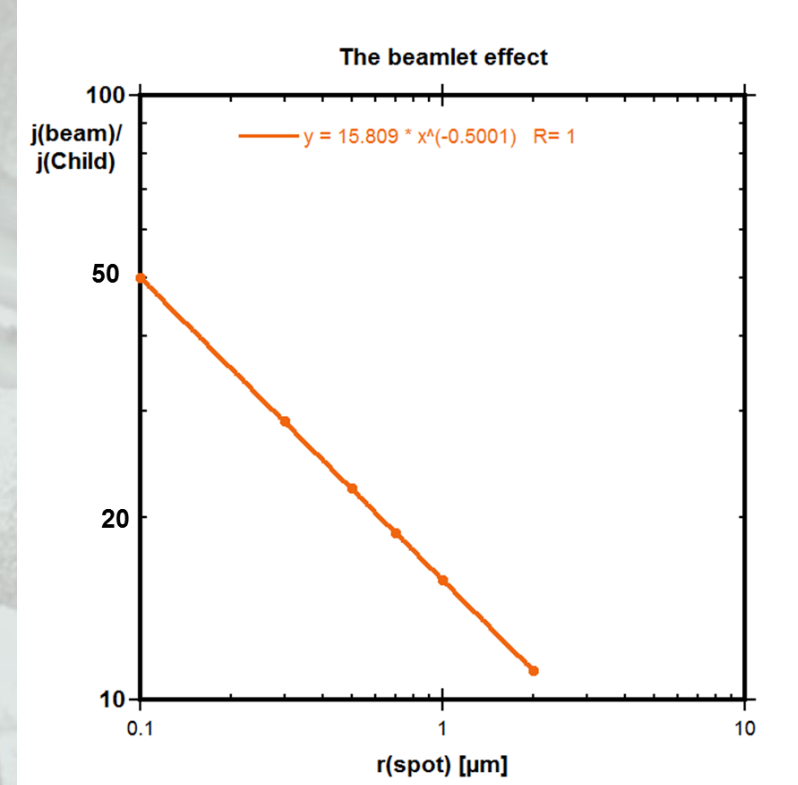
An experimental confirmation of the beamlet effect was obtained by Free and Gibson [6] for I cathodes by measuring the Ba diffusion length from AES Ba maps.

4b. The beamlet effect (J. Hasker) and scaling of Poisson equation

$$\frac{\partial^2 V}{\partial x^2} + \frac{\partial^2 V}{\partial y^2} + \frac{\partial^2 V}{\partial z^2} = - \frac{\rho}{\epsilon_0},$$

Eventually a solution of the **Poisson equation** in and around the patch is not available and hence Hasker derived equation (4) as an approximate result from the **scaling laws of Poisson equation**.

Scaling of Poisson equation means, that when multiplying all space dimensions with n , all current densities must be divided by n^2 . If all potentials are multiplied with a factor f_p , all current densities must be multiplied by a factor $f_p^{3/2}$, in order to obtain solutions again. From these conditions Hasker derived formula (4), see [5].

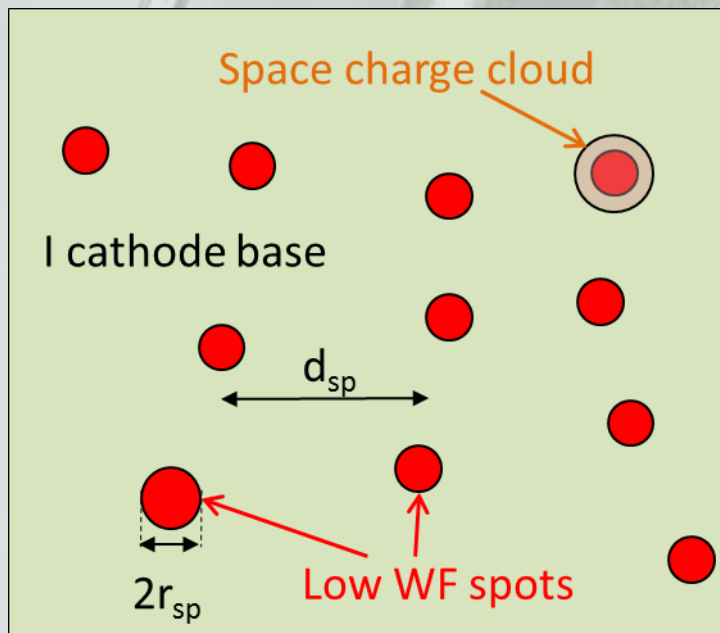


A qualitative explanation of the beamlet effect is, that for small spots space charge in front can expand into the surroundings and hence the space charge barrier is reduced. This also implies an extension of the (virtual) spot.

5. Model of Scandate cathode emission

This model has to solve **two problems**:

a) First the explanation of the low work function in the patches by a molecular model of the atomic arrangement at the surface, especially of Ba, Sc and O, similar to that given by Shengyi Yin in 2020, which eventually needs to be combined with a semiconductor theory.



Idealized map of local emitting spots

b) The second problem is the theoretical explanation of the **total emission** and I-U characteristics arising **from** localized distributed **highly emitting patches**.

The **quantification** is determined by several parameters (see beamlet effect), including the average distance (density) of the spots; but also the expansion of the space charge cloud (virtual cathode) can contribute.

5b) Model of Scandate cathode emission; some calculations

Of course in case b) **total emission** can be obtained as **superposition** of the emission **of all spots**.

The **model parameters** are the following (in brackets: used for calculation)

$2*r_{sp}$ = average diameter of the spots (0,3 μm)

d_{sp} = average distance of the spot centers (1,5 μm)

d_{ca} = cathode to anode distance (typically 250 μm)

Let us consider the case $j_{ch} = 100 \text{ A/cm}^2$, then $j_b = (250/0,15)^{1/2} * j_{ch} = 4082 \text{ A/cm}^2$

For a cathode with surface diameter of 1 mm (Philips 0,65 Watt unit) we obtain a total emitting area of $A_{cath} = 0,7854 \text{ mm}^2$ and from $d_{sp} = 1,5 \mu\text{m}$ about $N_{spot} = 394066$ emitting spots. From j_b we obtain $I_b = 2,89 * 10^{-6} \text{ A}$ per spot and $I = I_b^{sc} * N_{spot} = 1,14 \text{ A}$ (pulsed) emission and a total current density of **145 A/cm²**, which is a good confirmation of the model.

Looking at the spot area contribution including the space charge expansion one obtains an **area coverage of 3 %**, consistent with a **Richardson constant of $A_R = 3,7 \text{ Acm}^{-2}\text{K}^{-2}$** and a Scandate cathode on a tungsten base.

Of course we could have calculated one of the estimated parameters from the formula below, derived from (4) to obtain the exact starting value of j_{ch} :

$$I/A_{cath} = j_{ch} * (d_{ca}/r_{sp})^{1/2} * \pi * r_{sp}^2 * N_{spot} / A_{cath} \quad (6)$$

5d. Superposition model of highly emitting scandate cathode (semiconductor) patches, surrounded by W - I or Re - I cathode emission [1].

- explains deterioration with t_{op} and by IB, decreasing the size / number of patches by ablation [1].
power law $I = g U^x$ remains valid, but with x decreasing with increasing IB / t_{op}
- $e\Phi_R = 1.15 \text{ eV} \pm 0.05 \text{ eV}$ for all types of scandate cathodes also during deterioration, namely the WF of the highly emitting patches.
- **figure of merit** of improved scandate cathodes is **increase of Richardson constant**, i.e. increase of patch area or number

6. Conclusion:


The **superposition model of highly emitting patches together with the beamlet effect** explains the anomalous behavior of the I/U characteristics of Scandate cathodes in transition to saturation and also the degradation during life and during continuing ion bombardment. It is also consistent with the low work function of the patches and a Richardson constant in the order of $3 - 8 \text{ Acm}^{-2}\text{K}^{-2}$.

From this model one can derive 3 directions for further improvement of Scandate cathodes:

- I. Increase of the patch size**
- II. Increase of the patch density, i.e. the total number of patches on the cathode surface**
- III. Study and understand the surface structure and composition of the highly emitting patches**

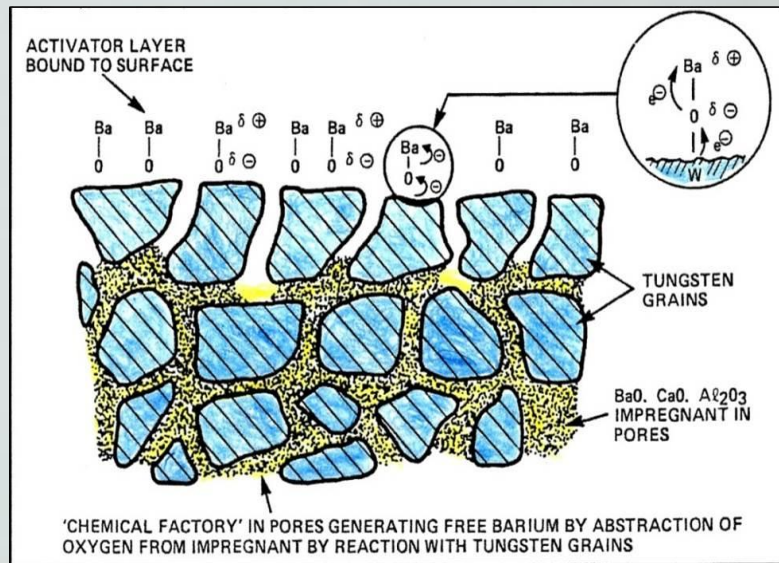
References:

- [1] G. Gaertner, Y. Wang, "State of the Art and Future Perspectives of Ba Scandate Dispenser Cathodes", Chapter 3 of **"Modern Developments in Vacuum Electron Sources"**, Springer 2020, 83 - 172.
- [2] G. Gärtner, "Hochemittierende Glühkathoden für Vakuum-Elektronenröhren - Historische Entwicklung, Stand der Technik und Ausblick", ViP Journal 34, (2022) 24 - 29
- [3] G. Gaertner, "Peculiarities of Ba scandate cathodes and constituents of a model", ITG-IVEW 2020, Book of abstracts, p. 26
- [4] J. Hasker, P. van Dorst, "Pitfalls in the evaluation of cathode properties from I-V characteristics", IEEE Trans. on ED 36, (1989) 201 – 208
- [5] J. Hasker, N. van Hijningen, "Cathode and scaling properties related to the shape of current- voltage characteristics", Appl. Surf. Sci. 24, (1985) 318f
- [6] B. A. Free, R. G. Gibson, "Dependence of surface coverage on pore geometry in dispenser cathodes", Appl. Surf. Sci. 24, (1985) 358 - 371



Thank you for your attention

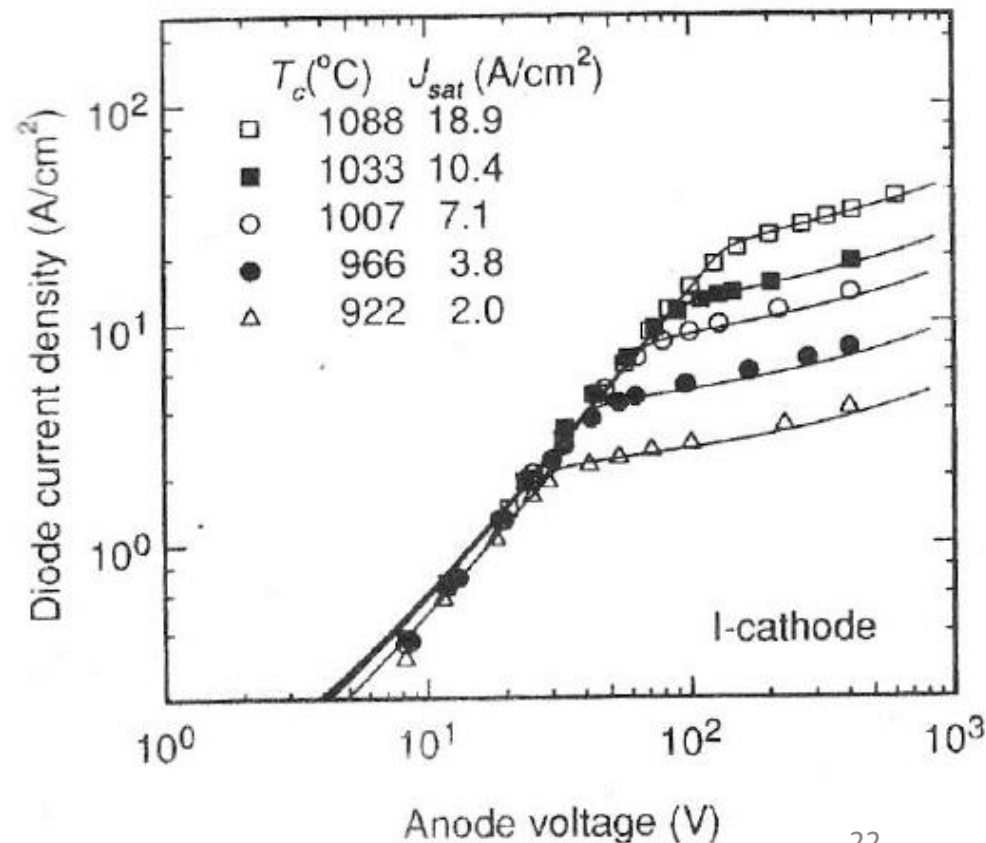
**Vielen Dank für Ihre
Aufmerksamkeit**



Cross sectional view of an impregnated cathode; free Ba is generated in the pores by a reaction of BaO of the impregnant with the tungsten walls

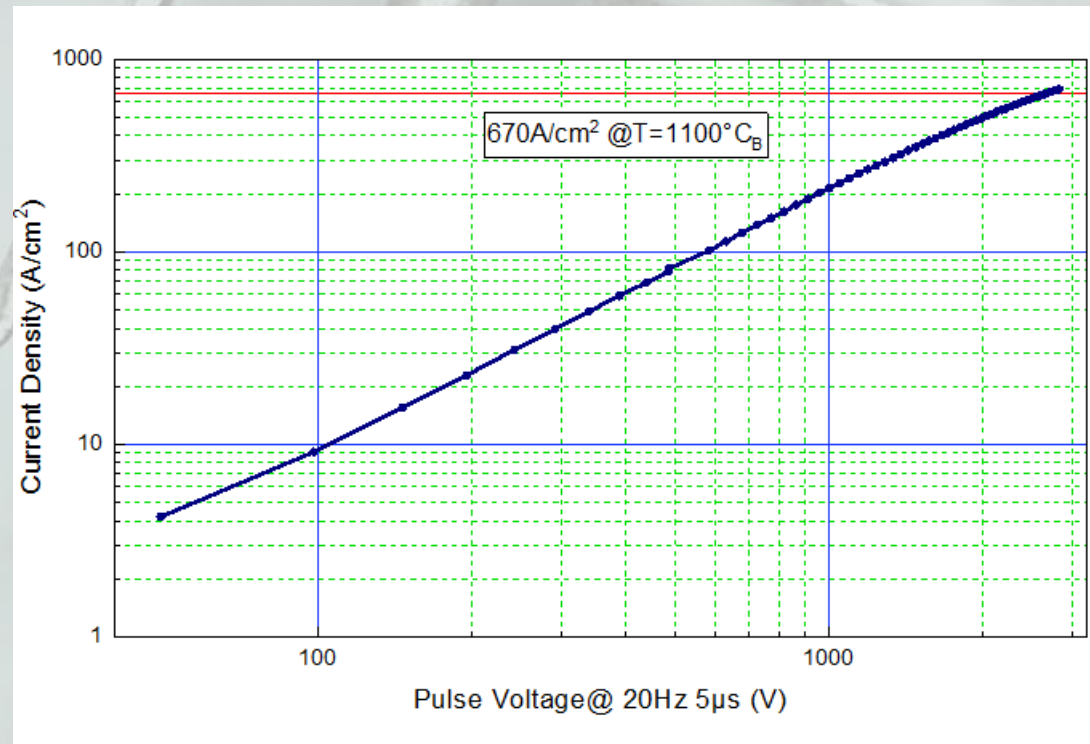
Ba dispenser or impregnated cathodes

The $\lg j/\lg U$ characteristics are for a Philips Os/Ru-I cathode at different true operating temperatures. A clear Schottky behavior in saturation is seen

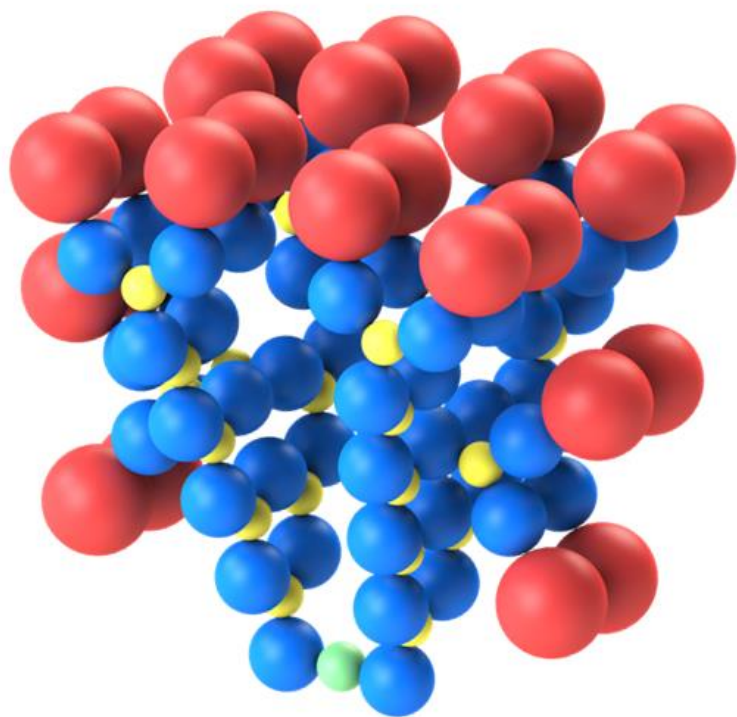


Current top result 670 A/cm² pulsed 20 Hz, 5 μs, T = 1100°C_b of Shengyi Yin* 2020 obtained with improved impregnated Scandate cathode, impregnant Ba₂ScAlO₅ +3at% SrO.

***Key Laboratory of High Power Microwave Sources and Technology, Chinese Academy of Sciences, Beijing 101407, China**



Binary Tree Model of Scandate cathode



Typical structure

- There are 16 Ba^+ ions on the outmost layer, and 8 Sc^{3+} ions on the second outmost layer in height direction;
- Other 8 Ba^+ ions and 4 Sc^{3+} ions are on the side branches;
- The total Ba:Sc molecular ratio is 24:12 or simplified **2 : 1**, which is perfectly consistent with that of our salt recipe.

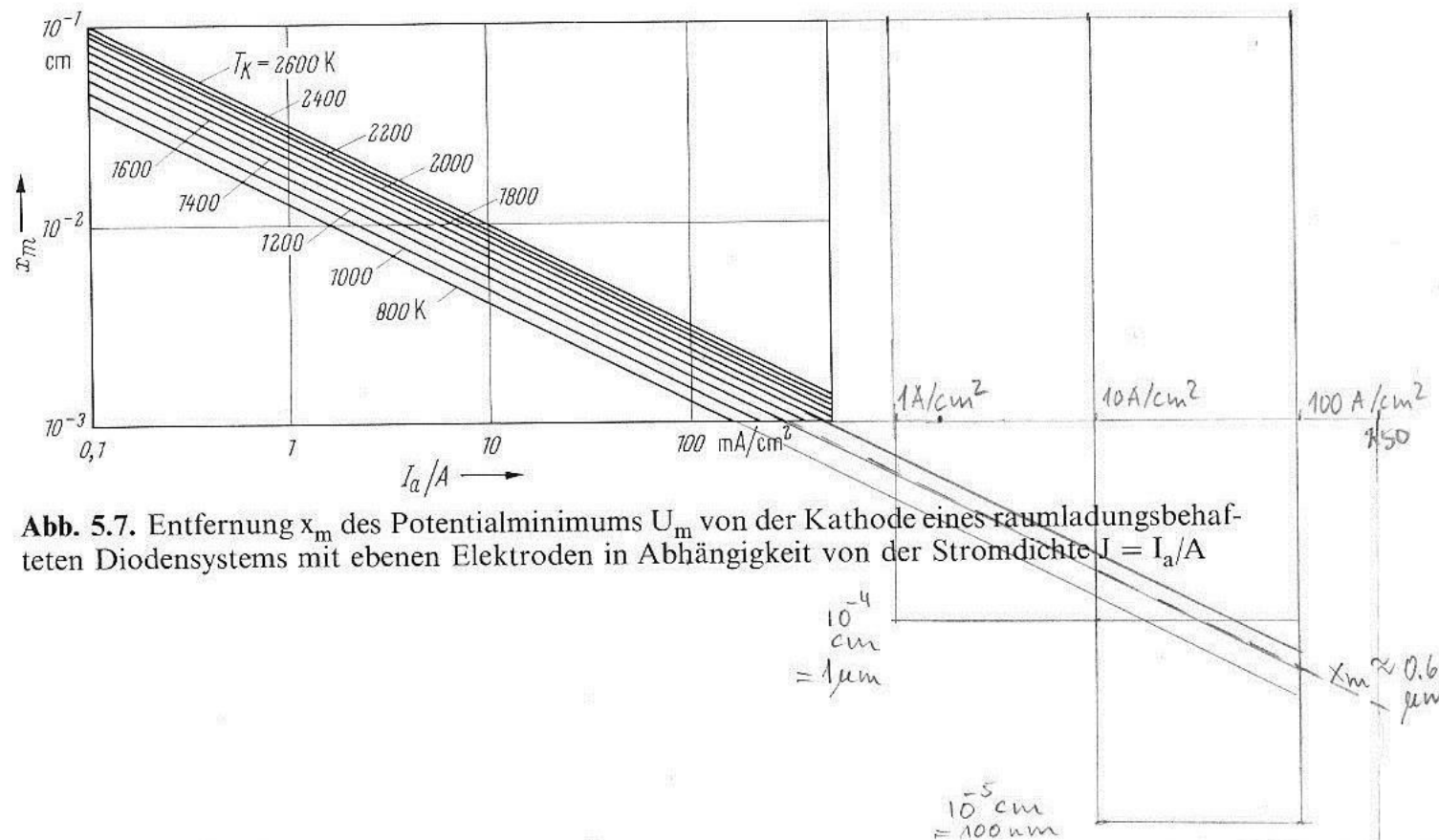


Abb. 5.7. Entfernung x_m des Potentialminimums U_m von der Kathode eines raumladungsbehafteten Diodensystems mit ebenen Elektroden in Abhängigkeit von der Stromdichte $I = I_a/A$

Fig. : According to **J. Eichmeier, Moderne Vakuumelektronik, Springer 1981, page 209**: The extended diagram shows the distance x_m of the space charge minimum from the cathode surface as a function of the current density; this also should be a measure for the space charge coupling length.

