"Revisiting the Early BARC Tritium Results"

(1989-1996)

ICCF 18

23rd July 2013

University of Missouri

Scope of BARC Studies

- Target Materials studied : Pd, Ti & Ni
- Hydrogenous isotopes used : D & H
- Loading Techniques : Electrolysis , Gas, plasma , (even ion beam loading!)
- Objective of Program : Establish Nuclear origin of Phenomenon primarily through
- Measurement of Neutron & Tritium yield

The Beginning

- 24th March 1989 : Four line news item in Indian Newspapers on F & P press conf.
- Specifically mentioned neutrons observed.
- Involved in fission reactor expts at BARC
- Excited that neutrons produced during a simple electrolysis experiment!
- As Head of Neutron Physics Divn I could quickly mobilize neutron detectors.
- By a remarkable coincidence had worlds biggest cold fusion cell all set to go!

"Milton Roy" Pd-D_2O Electrolytic Cell

"Pure" H_2 generator - separated from O_2

- Procured from M_R co. of Ireland and adapted to generate "pure" D_2 gas for our ongoing "Plasma Focus" (hot fusion) expts
- Pd-Ag alloy tubular cathodes - 16 nos;
- (Jed Rothwell talk yesterday)
  Total cathode area 300 cm^2;
- Inner/outer Ni pipes serve as anode.
- Cathode tubes distributed in annular space between Anode pipes.
- Max current capacity ~100 amps.

Milton Roy Electrolytic Cell

Pd-Ag alloy tubular cathode

Ni pipes Anode

NaOH/NaOD Electrolyte

H_2 (or D_2) gas escapes through bottom separated from Oxygen

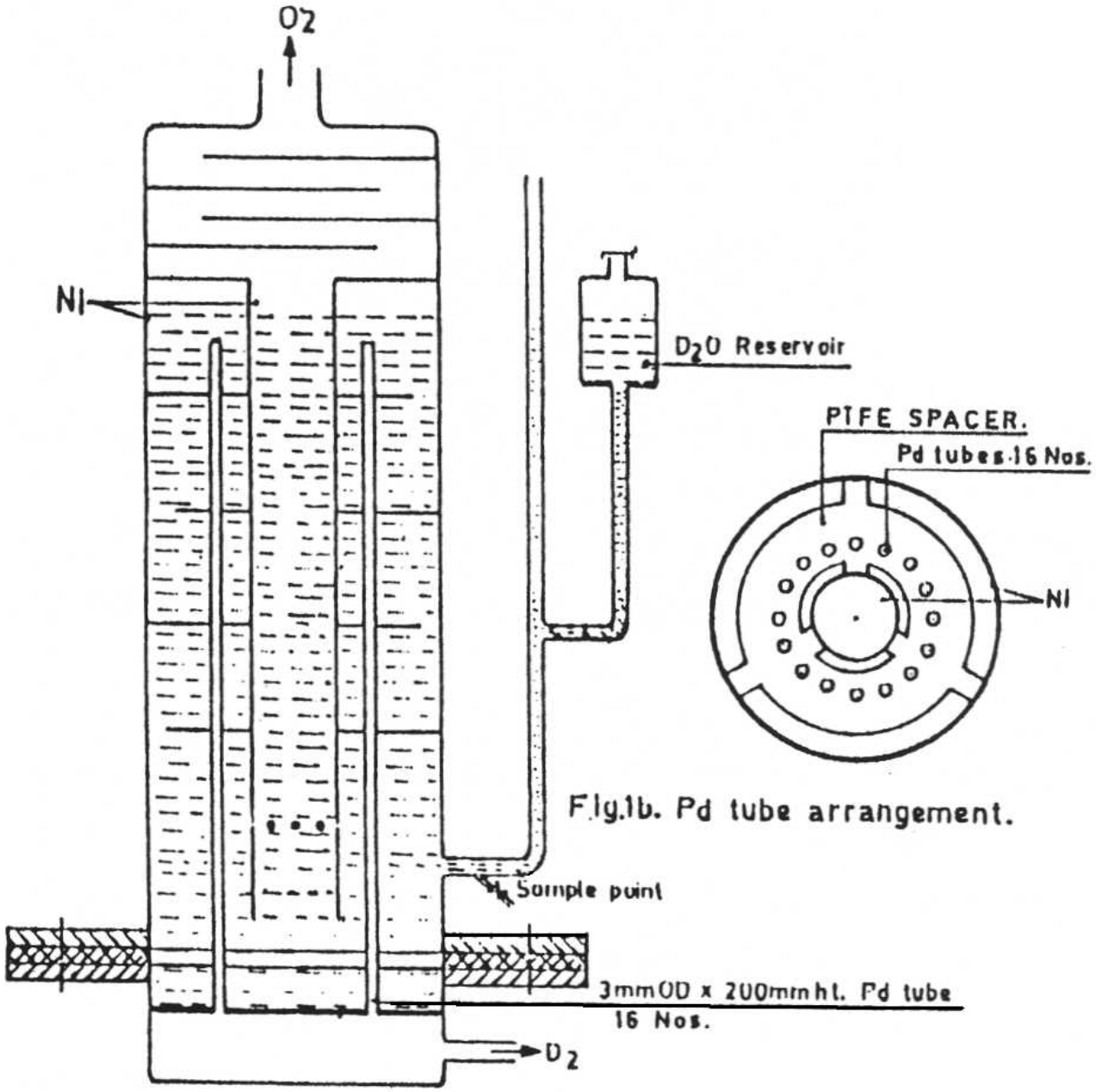
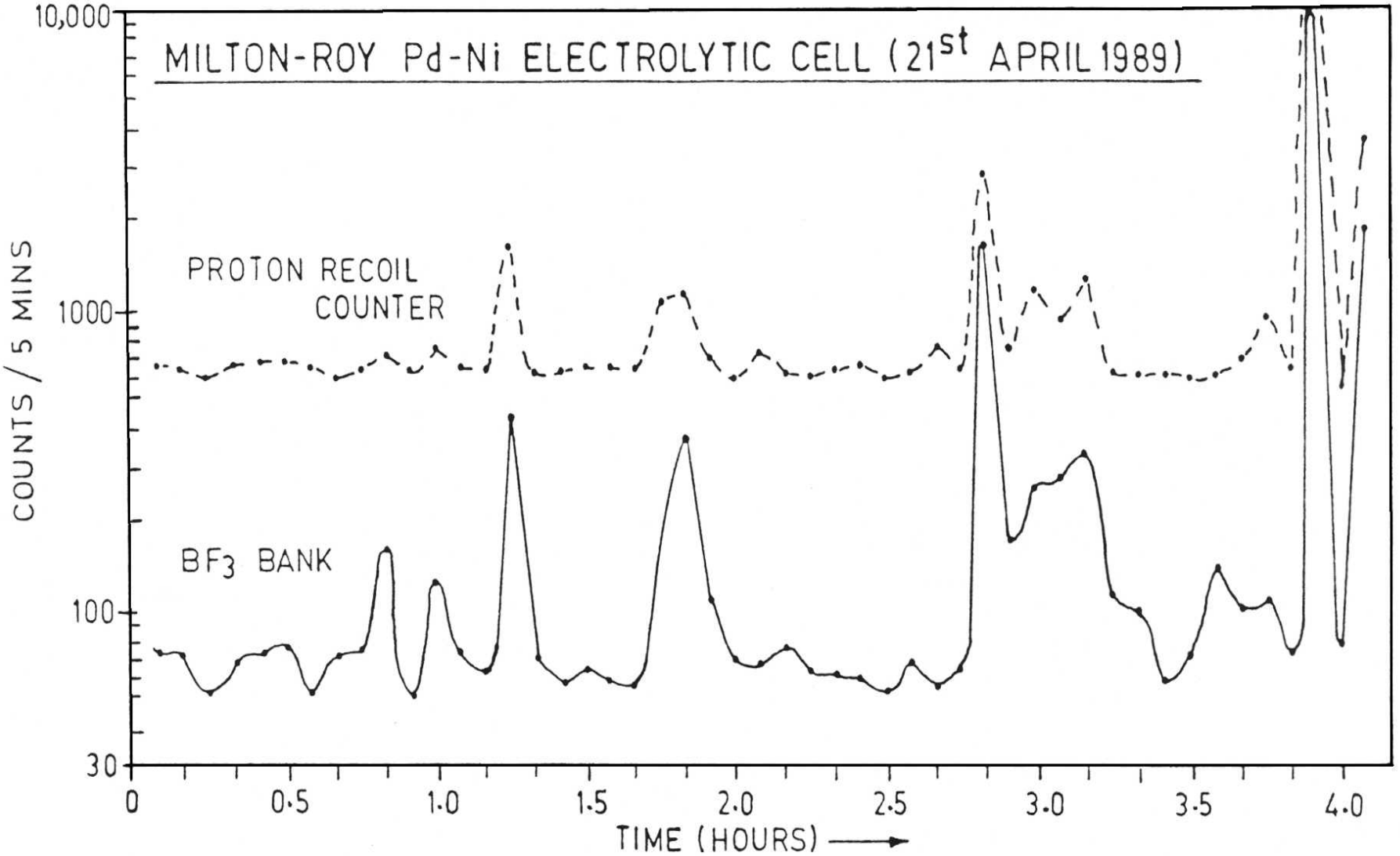


Fig.1b. Pd tube arrangement.

Neutron Detectors Used

- NE 102A : Proton recoil type plastic scintilator detector, sensitive mainly to fast neutrons & gamma rays.
- BF_3 type slow neutron counters (45 cm long) embedded in moderator block.
- He^3 type slow neutron counters embedded in paraffin block;banks of 3 counters each
- In the latter two, fast neutrons slow down in the moderator block before being detected by the or BF_3 or He^3 detectors
- Resultant statistical time spread exploited!

First Neutron Signal on 21st April 1989
(Presented at Conf. in Karlsruhe July 1989)



Tritium Measurements

- Tritium ($T_{1/2}$ ~12 years) emits very low energy betas (<18 Kev) .. cannot be measured on line; only post run time integrated yield possible.
- Tritium in electrolytic solution at end of run measured by std. liquid scintillation spectrometer by experts in field from Health Phys Division ;
- Chemiluminiscence & quenching effects avoided ; Vials with cocktail + solution kept in dark for several hrs for Chemiluminiscence cooling;
- Due to low β energy, K^{42} depleted vials used;
- Pre-electrolysis control samples & Stds. counted between post run samples using same system;

Results from Milton Roy Cell - Run 1

- Showed massive amount of 1.5 µCi/ml (or 55.5 kBq/ml) at end of run;
- 20,000 times initial heavy water content!
- Total T atoms produced 8.10^{15} atoms *
- Total neutron yield in run was : 4.10^{7}
- Hence (n/T) ratio → 5.10^{-7}
- First hint of "branching ratio anomaly" !
- ----------
- (*Actually T estimate conservative as tritium carried away by gas stream disregarded.)

BARC Electrolysis Experiments (1989-90)

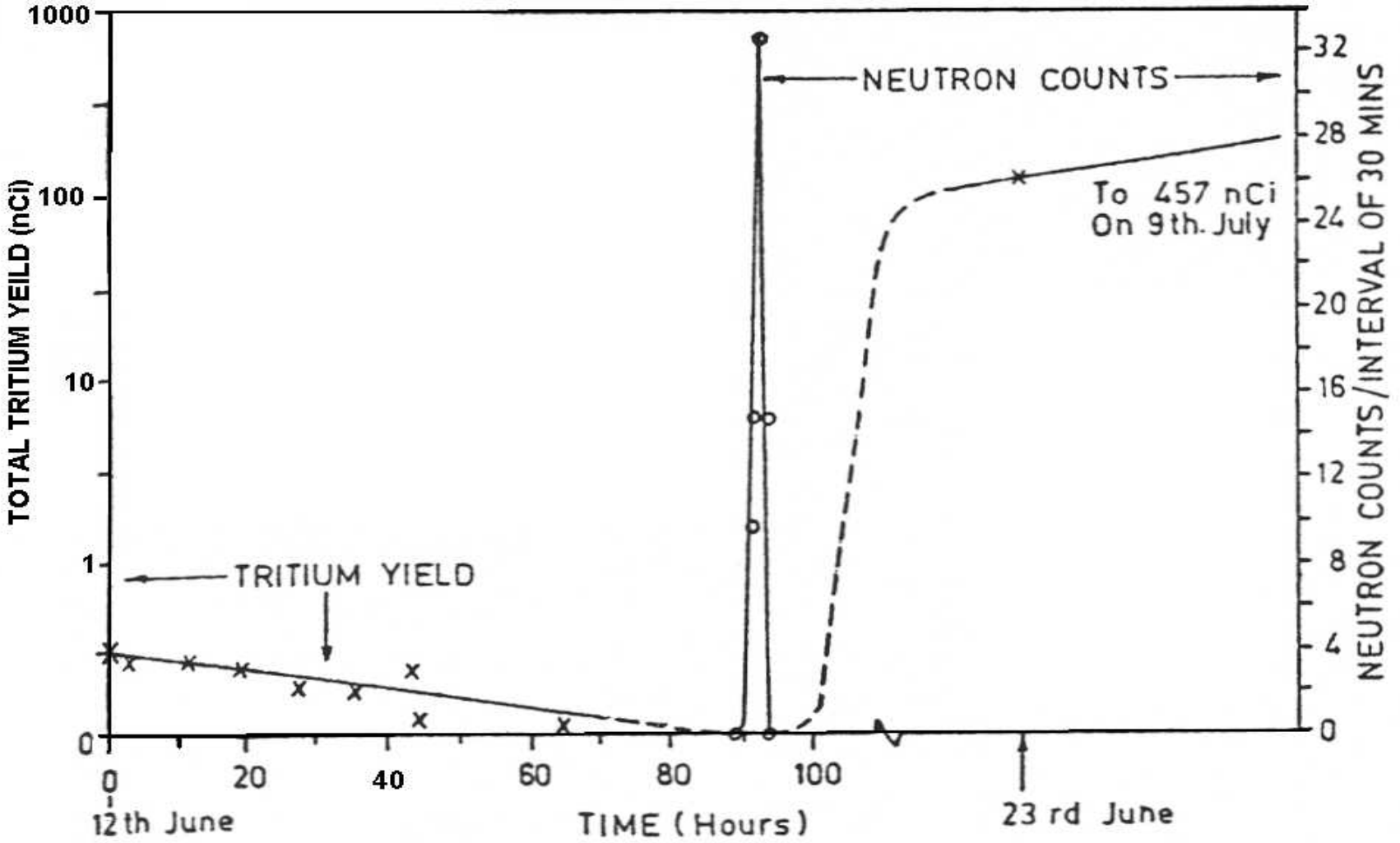
	Division	Cathode Material	Geom	cm^2 Area	Anode	Neutron Yield	Tritium Yield	n/T Ratio
1	Desalin *	Ti	Rod	104	SS pipe	3.10^{+7}	$1.4\ 10^{+14}$	2.10^{-7}
2	Neut. Phy.*	Pd-Ag	Tubes	300	Ni Pipes	4.10^{+7}	8.10^{+15}	5.10^{-7}
3	HWD *	"	"	300	"	9.10^{+7}	$1.9\ 10^{+15}$	5.10^{-7}
4	HWD *	"	5 Disks	78	Porus Ni	5.10^{+4}	4.10^{+15}	$1.2\ 10^{-9}$
5	Anal.Ch.@	Pd	Hol.Cyl.	5.9	Pt Mesh	3.10^{+6}	$7.2\ 10^{+13}$	4.10^{-8}
6	ROMg @	"	Cube	6.0	"	$1.4\ 10^{+6}$	$6.7\ 10^{+11}$	$1.7\ 10^{-4}$
7	ROMg @	"	Pellet	5.7	"	3.10^{+6}	4.10^{+12}	1.10^{-4}
8	App.Chem @	"	Ring	18	"	$1.8\ 10^{+8}$	$1.8\ 10^{+11}$	1.10^{-3}

– Electrolyte : * 5M NaOD @ 0.1M LiOd

TABLE II: SUMMARY OF OTHER TRITIUM PRODUCING ELECTROLYTIC EXPERIMENTS

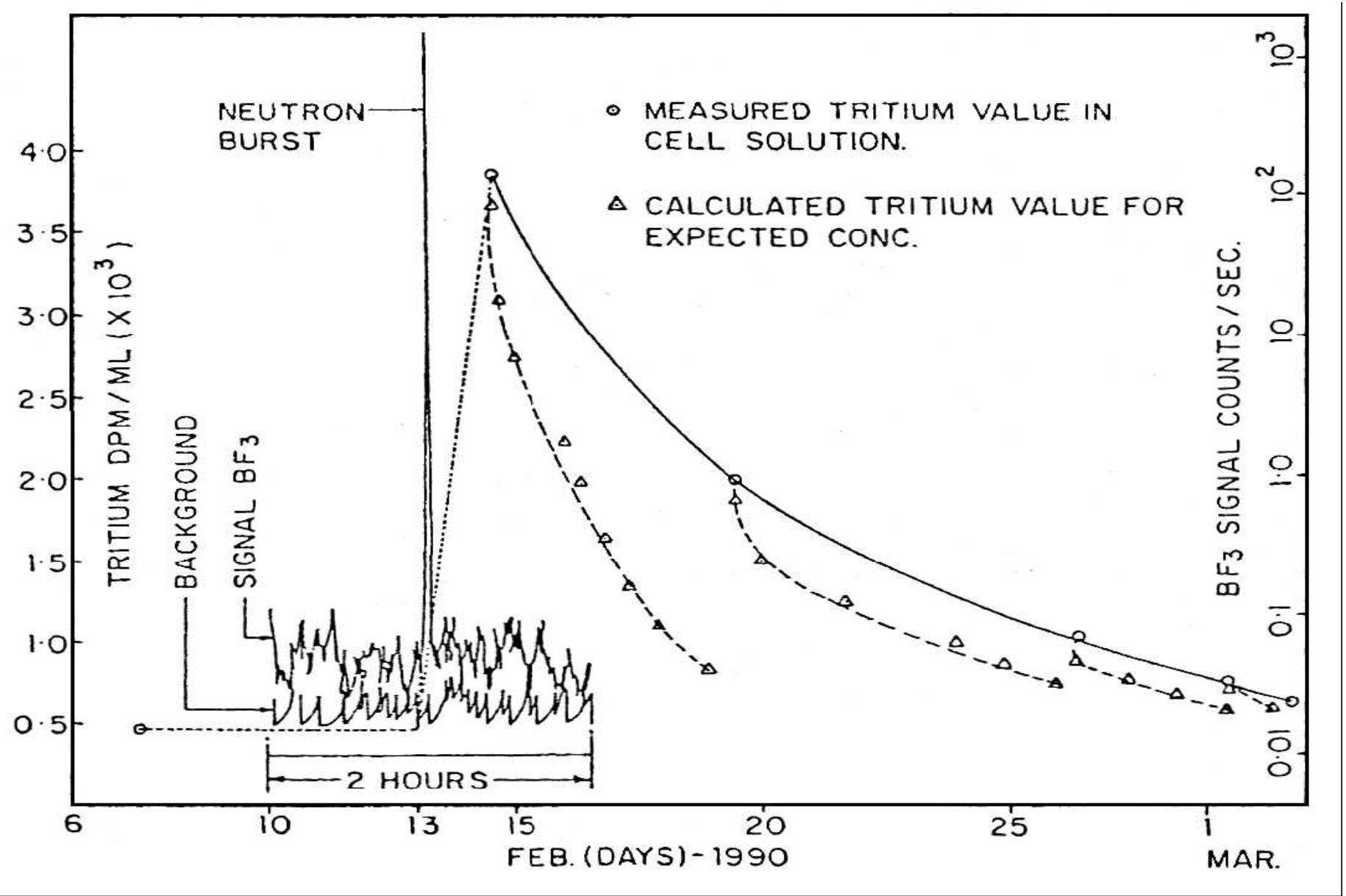
Sr. No.	#1	#2	#3	#4	#5	#6	#7	#8	#9	#10	#11
Division Cell (Name)	Heavy Water Division		ROMG	Analytical Chemistry Division					Chemistry Division		
Cell (Name)	MR (Jr)-I	MR (Jr)-II	RCS-18	PDX-0	PDC-II	PDC-III	PDC-IV	PDR-1	CD-4	CD-6	CD-5
Date	1989	1990	1989	1989	1989	1989	1989	1989	1989	1989	1989
	21 Sept	5 March	24 Oct.	24 April	10 July	6 Sept.	29 Sept.	9 Nov.	21 July	22 Dec.	24 Oct.
Cathode:											
Material	Pd-Ag Alloy	Pd-Ag Alloy	Cold Rolled Pd	Pd	Pd	Pd	Pd	Pd	Pd ingot	Pd	Pdwire
Initial Concn. (Bq)	1.44	3.33	3.6	2.7	2.81	2.77	2.70	2.68	4.6	2.0	2.5
Maximum Concn. (Bq/ml)	225.7	18.5	–	0.93×10^{4}	5.88×10^{4}	4.6	–	–	72.1	65.0	22.9
Output to Input Ratio	156.7	5.6	3.36	3425	20,925	1.66	2.5	1.91	15.7	32.5	9.16
(Bq)	3.3×10^{4}	2.28×10^{3}	2.71×10^{3}	6.02×10^{5}	2.08×10^{6}	2.96×10^{3}	6.29×10^{2}	1.1×10^{3}			
(Atoms)	1.76×10^{13}	1.2×10^{12}	1.44×10^{12}	3.2×10^{14}	1.1×10^{15}	1.56×10^{12}	3.96×10^{11}	5.83×10^{11}	10^{12}	10^{11}	2×10^{10}
t/cm^2	1.6×10^{11}	1.1×10^{10}	0.8×10^{11}	2.2×10^{13}	1.7×10^{14}	2.4×10^{11}	6.2×10^{10}	2.12×10^{11}	1.8×10^{12}	0.8×10^{12}	0.5×10

Several independent cells indicated that n & T are probably generated concomitantly



Neutron & Tritium Output - ROMG Cell

(11 mm dia Pd Pellet cathode -13th Feb 1990)



Deuterium Gas/Plasma Loaded Titanium Targets

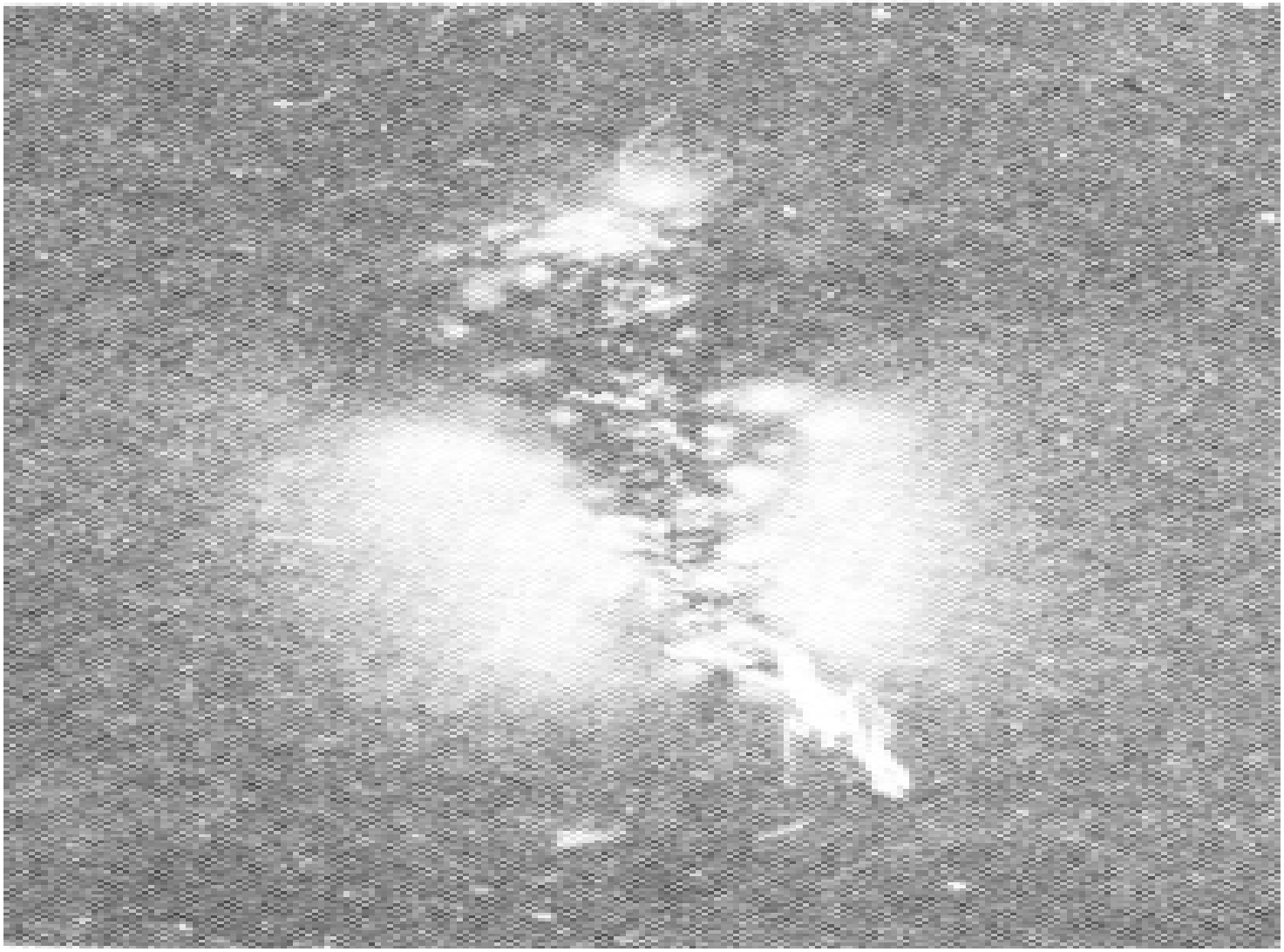
Got interested in TiD_x targets following Frascati Group's (De Ninno et al, 1989) work

We used autoradiography as a very effective tool !

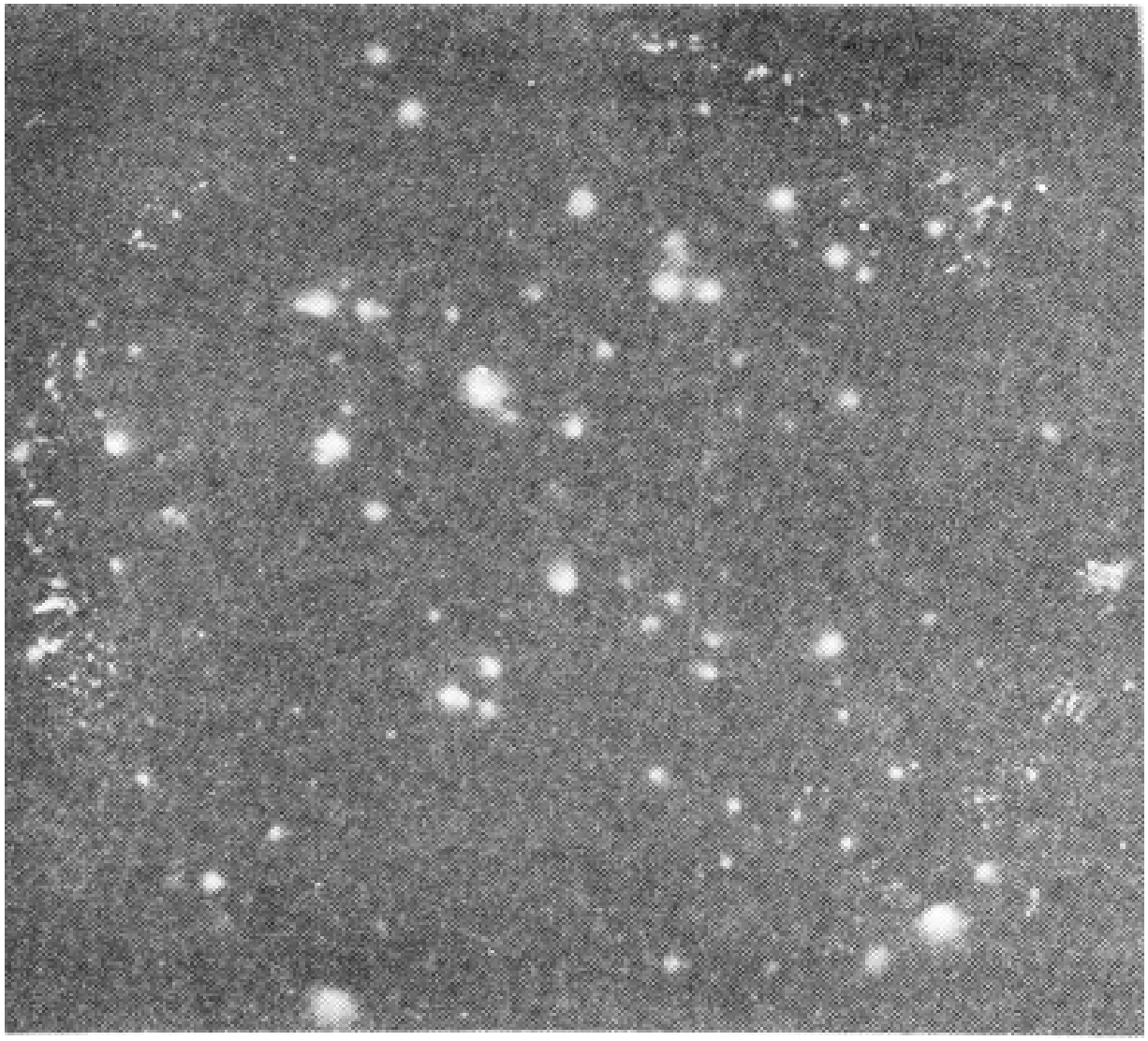
Autoradiography, a Powerful Tool

- Simple technique of detecting radiation emitting zones; free from electromagnetic interference
- Standard medical X-ray films of ~10 μm grain size; At times stack of several films used.
- High sensitivity if integrated over long exposure times. Overnight to 60 hr exposure gives good space resolved images.
- Developing time was typically 4 to 5 minutes.
- Polaroid films too used; gave images directly.
- In some cases films were placed on both sides of the sample.

Autoradiograph of a Conical Ti Electrode (After many Glow Discharge Shots in D_2 gas)



Autoradiograph of Deuterated Ti Disc



Ti K_α X-Ray Measurements

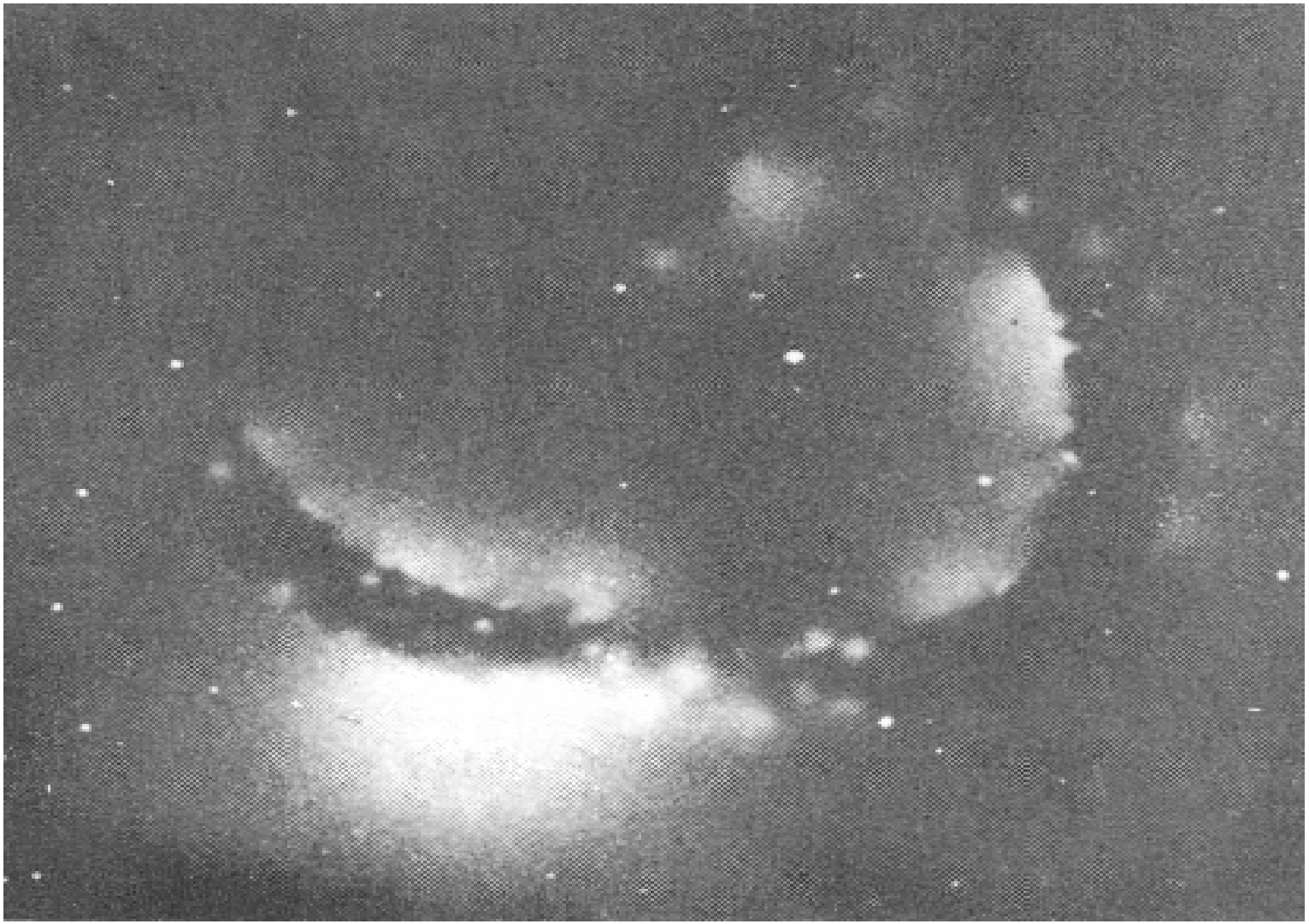
- 18 Kev Tritium βs able to excite characteristic X-rays of Ti; But efficiency of conversion only 10^{-4}
- Si(Li) surface barrier detector shows K_α (4.5 Kev) and K_β (4.9 Kev) peaks of Ti ;
- Autoradiography demonstrates existence of highly localized hot spots on target surface wherein tritium is concentrated;
- Occurrence of spots all along periphery of disc, points to important role of lattice defect-sites in T production phenomenon.

Tritium generation when Deuterated Ti metal lathe shavings subjected to thermal shock

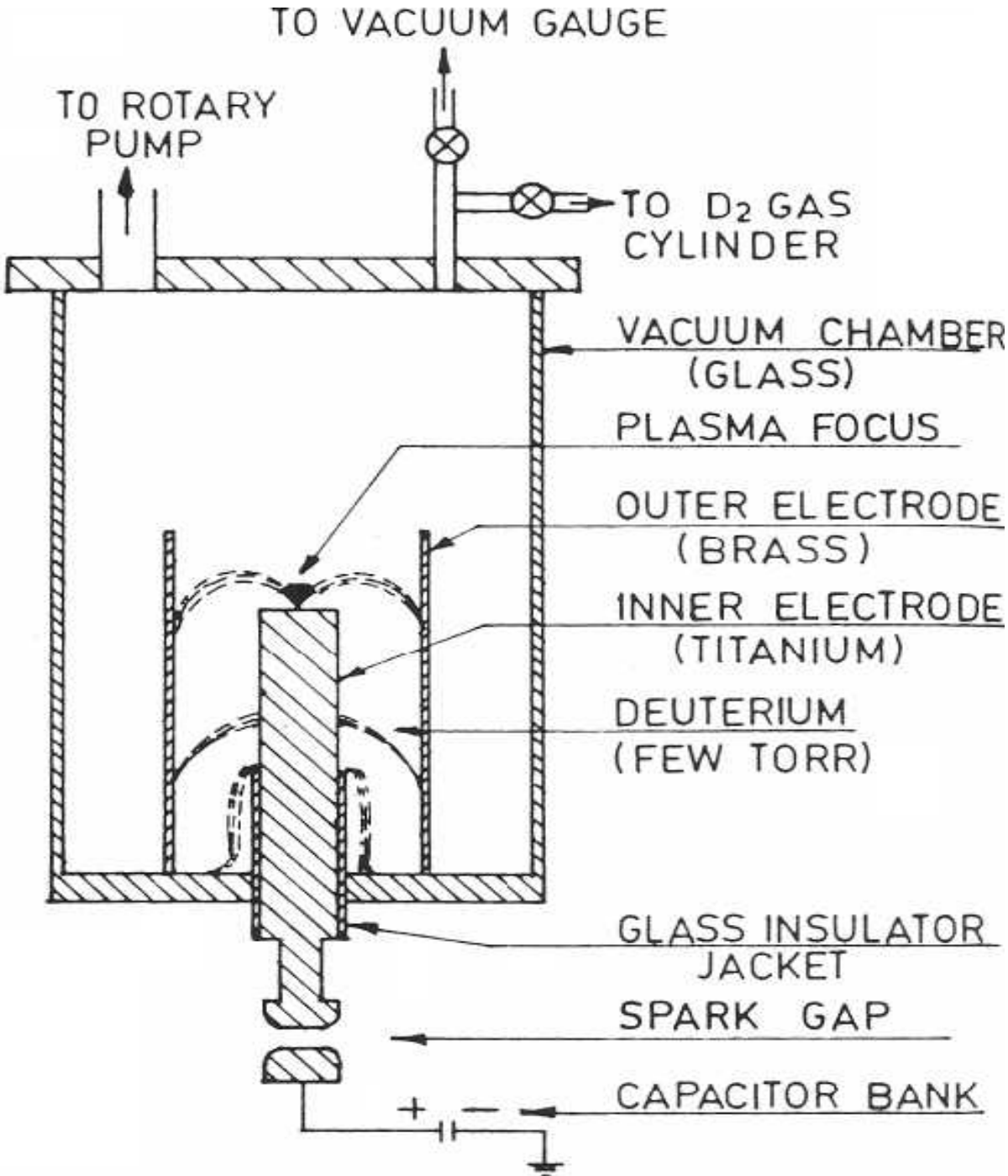
(Kaushik et al, Indian J. Technol, 1990, 28, 667)

- Ti metal shavings made using a lathe (3 ~ 8 mg)
- Thoroughly cleaned in acid/distilled water etc.
- Evacuated and heated to 850C, cooled in D_2 gas
- Amount of gas absorbed measured thru Pr. Drop
- TiD_x Chips dropped in cylinder containing LN_2
- Cylinder refilled with LN_2 repeatedly.
- Looked for T in lots of 50 chips using NaI detr.
- 4 out of 1000 chips showed MBqs of Tritium
- (T/d) for these high activity chips was 10^{-4} !
- (T/d) ratio of initial D_2 gas measured as (10^{-13})

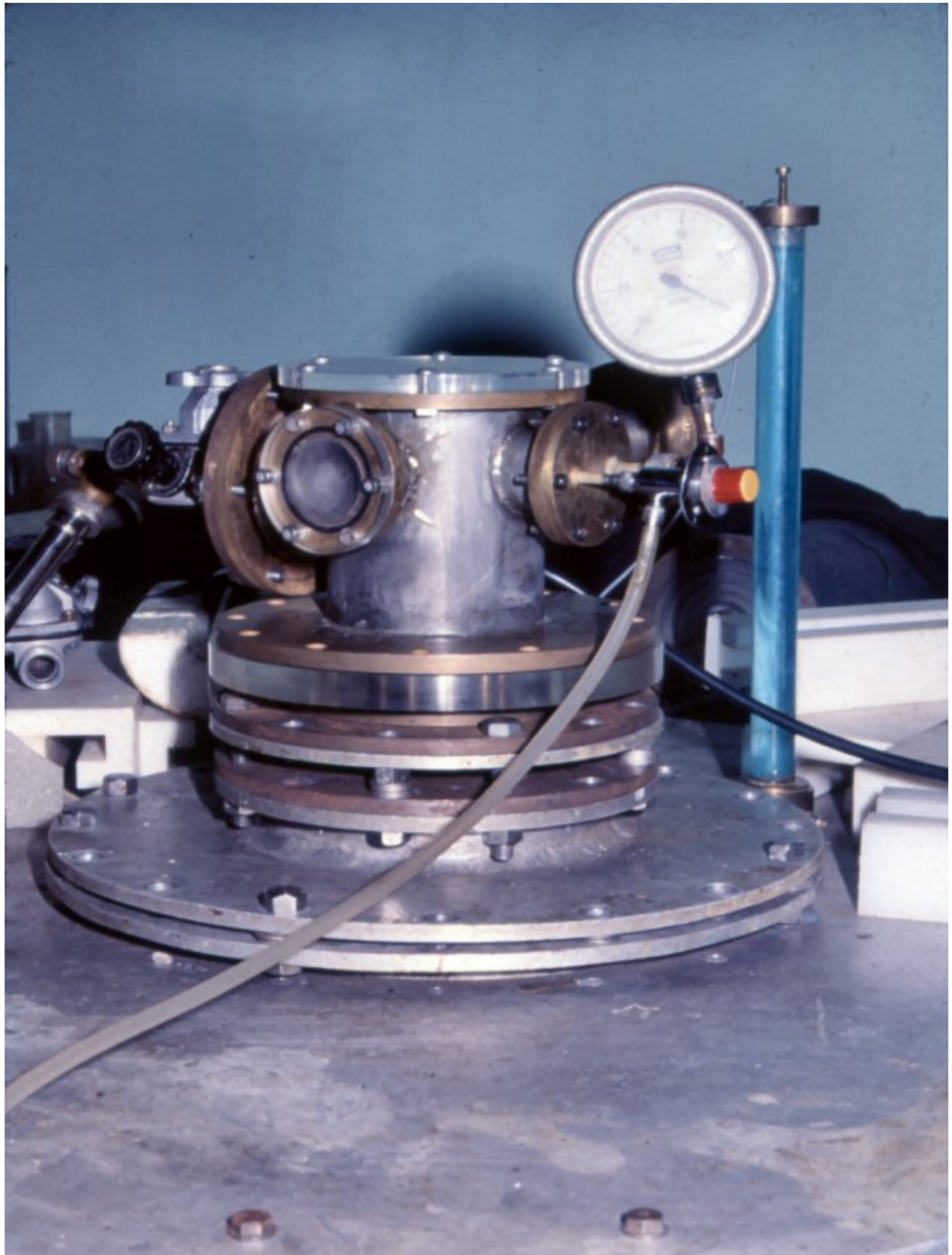
Autoradiograph of the most Active TiD_x Shaving following dunking in Liquid Nitrogen Again shows spotty nature of T distribution



Plasma Focus Experiments



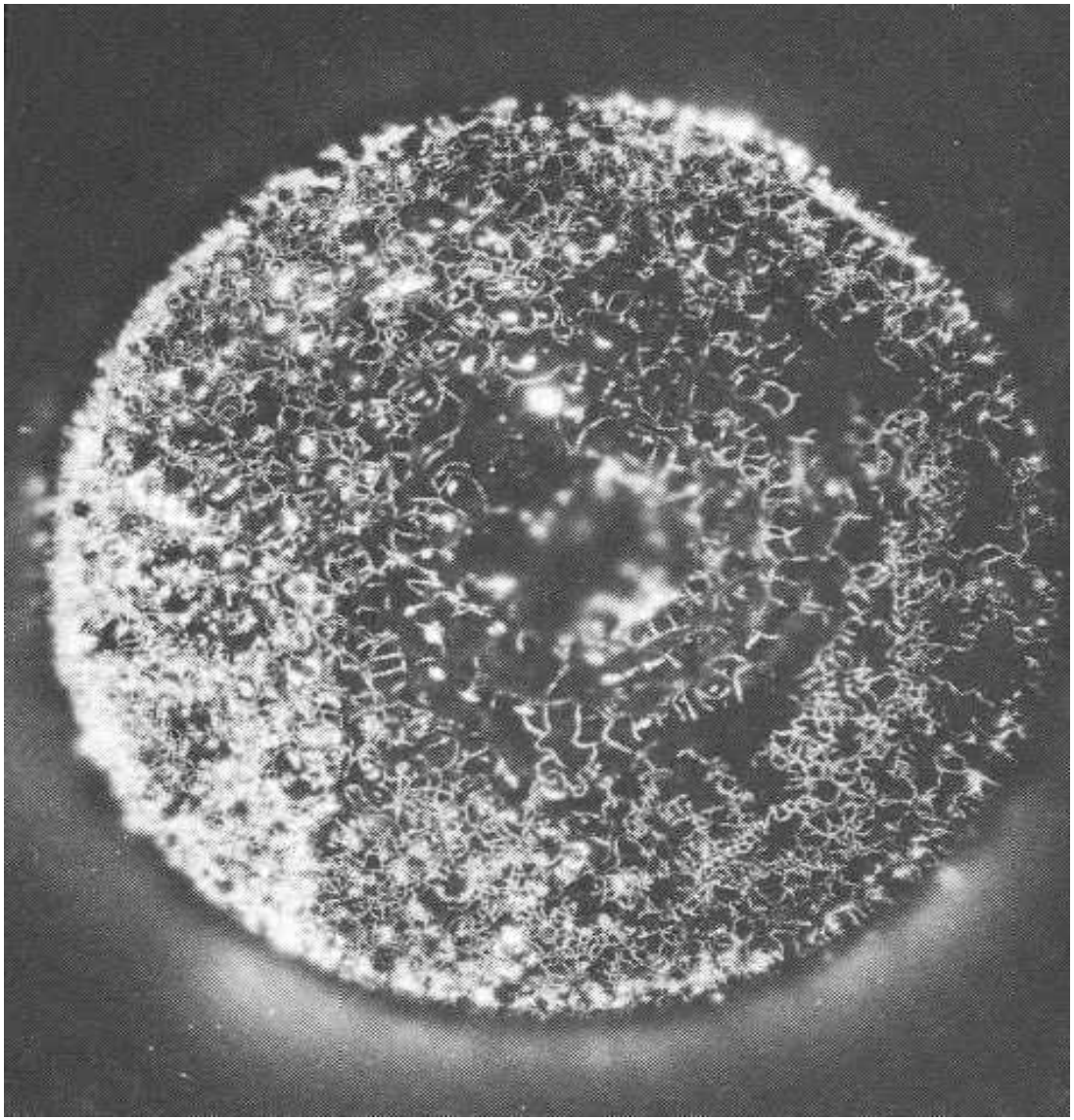
Schematic of Plasma Focus Device



Plasma Focus Set up

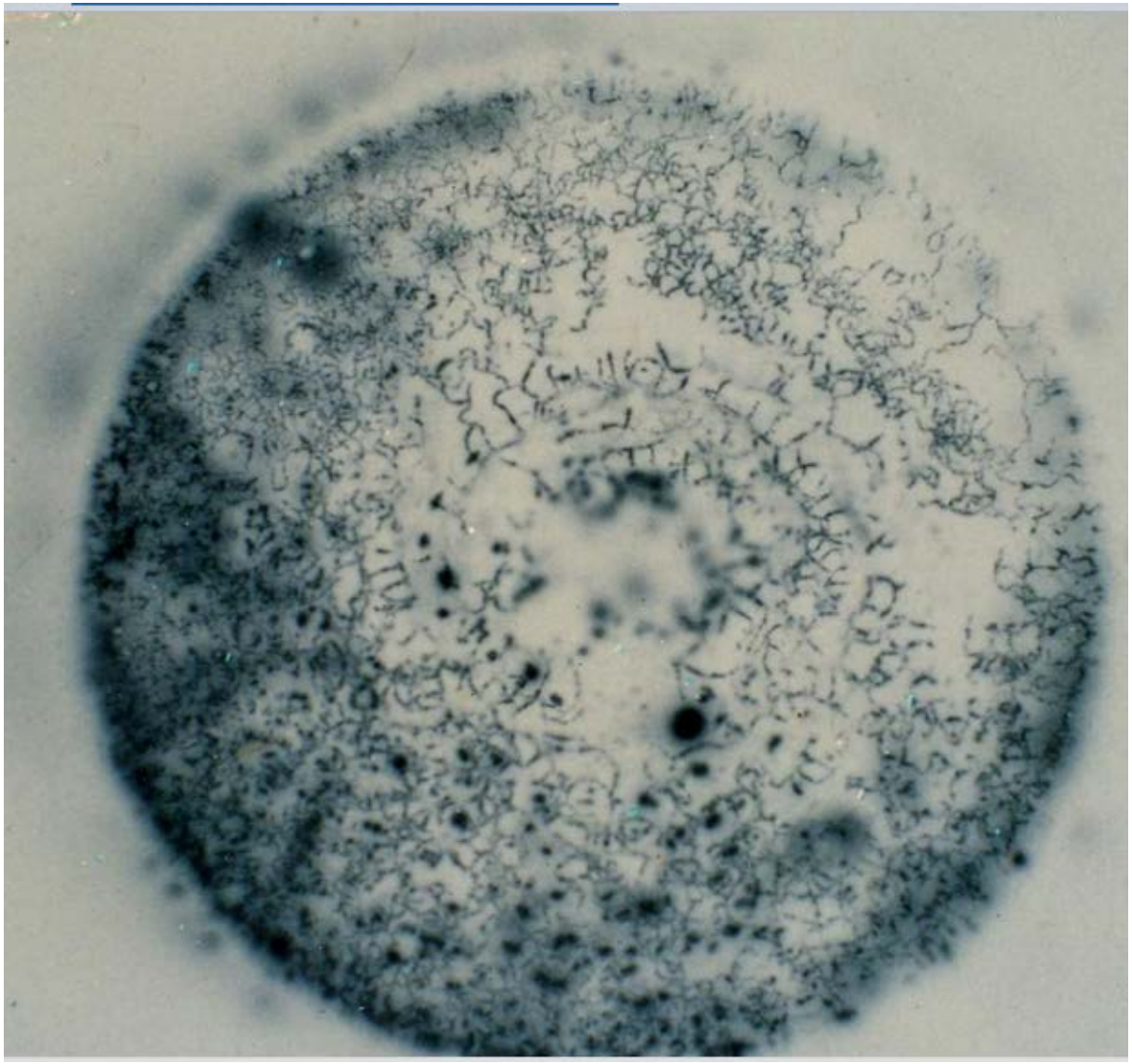
Ti Anode of PF Device after 50 shots

Autoradiograph of Ti Anode of Plasma Focus Device taken after 50 Charge/Discharge Shots with D_2 gas



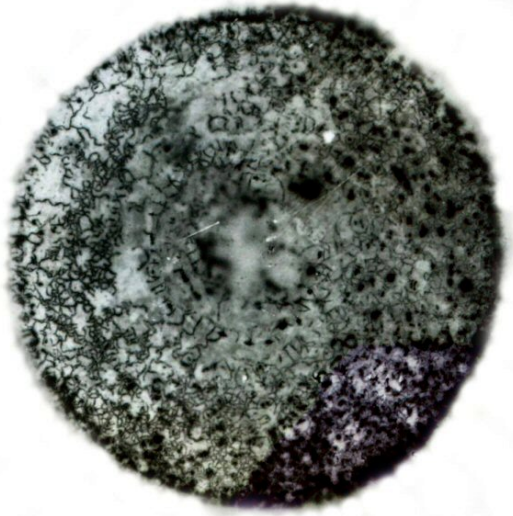
Radiograph taken on Polaroid Film

Disk dia.is 6.7 cm; Image is 200 dpi.

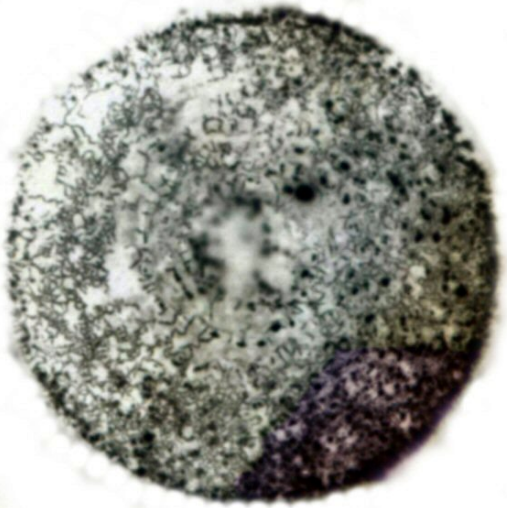


Remarkable Reproducibility of Ti autoradiograph

- This Anode was repeatedly autoradio-graphed several times over 5 year period.
- Image changed very little, indicating amazing stability of tritium (and hence all hydrogenous isotopes ?) in Ti lattice.
- Top surface had 10^{16} atoms of tritium



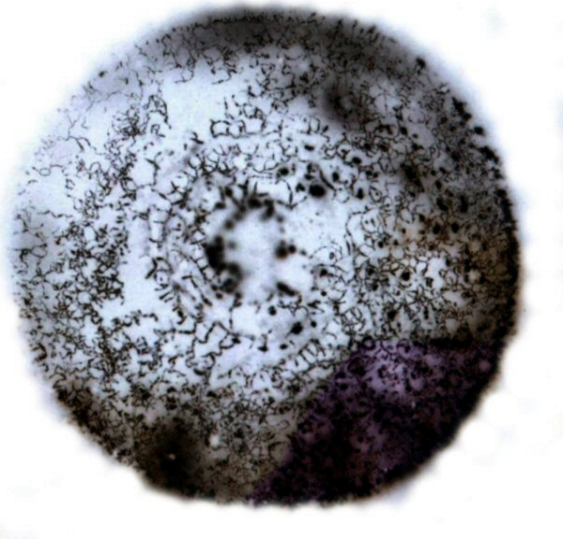
12th Feb 1990



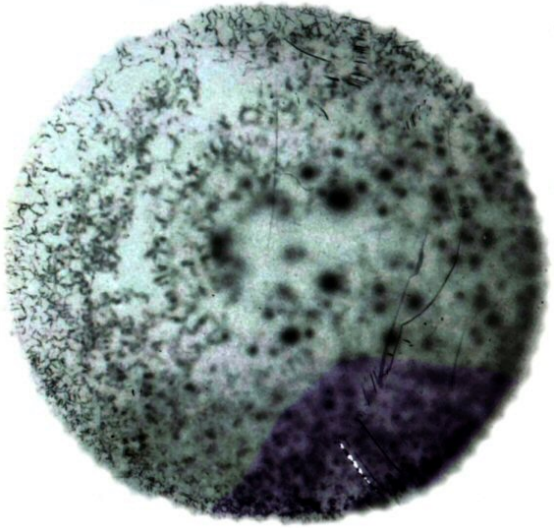
13th Feb 1990

15th Feb 1990

26th June 1990



15th April 1991



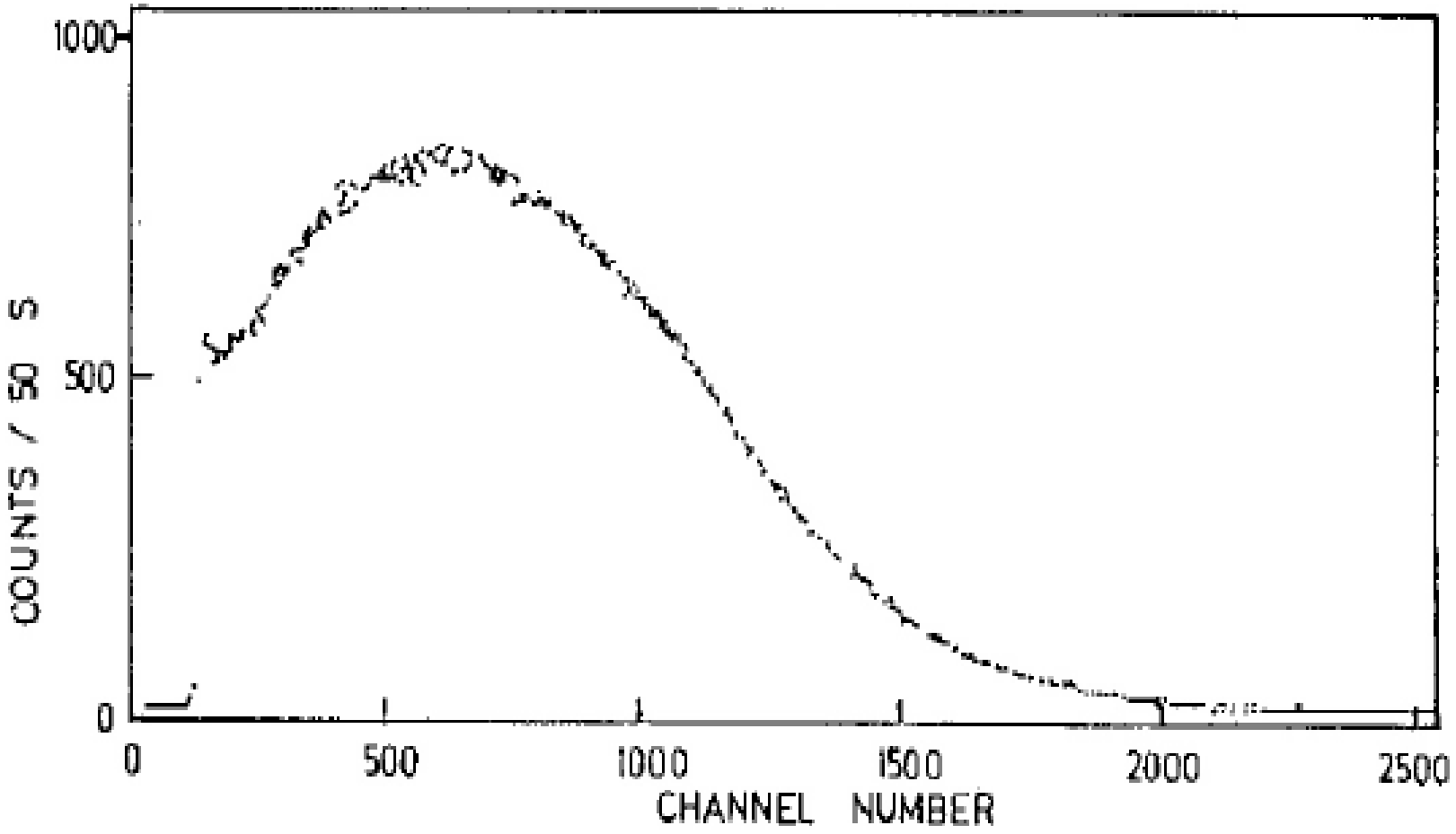
17th Feb. 1995

"Aged" TiD Targets Study

- Deuterated thin film Ti targets (on Cu backing) imported during 1972 -1981 for dosimetry studies using accelerator generated (d,d) neuts
- Ti thin film thickness ~ 3 mg/cm^2; (D/Ti) ~ 1
- Speculated CF reactions might have occurred and Tritium generated over 9 to 18 yr period.
- All 12 aged targets-> autoradiographic fogging
- Images were uniform, not spotty (thin film?)
- Mostly 10s of MBq; Highest was ~ 200 MBq
- Presence of Tritium confirmed by 5 techniques
- (T/d) ratio ~ 10^{-4}; much > Candu reactor D_2O
- Suppliers (Amersham Labs) vehemently refuted possibility of T contamination during fabrication

Observation of Tritium in Aged TiD Targets

β Spectrum of CDX 996 Target



"Bhabha Atomic Research Centre Studies on Cold Fusion"
Fusion Technology, August 1990 Issue..Thank you George !

- **Iyengar, P. K. et al (50 Authors)…… 94 pages**
- **Co-authors are listed below :**
- Srinivasan, M., Sikka, S. K., Shyam, A., Chitra, V., Kulkarni, L. V., Rout, R. K., Krishnan, M. S., Malhotra, S. K., Gaonkar, D. G., Sadhukhan, H. K., Nagvenkar, V. B., Nayar, M. G., Mitra, S. K., Raghunathan, P., Degwekar, S. B., Radhakrishnan, T. P., Sundaresan, R., Arunachalam, J., Raju, V. S., Kalyanaraman, R., Gangadharan, S., Venkateswaran, G., Moorthy, P. N., Venkateswarlu, K. S., Yuvaraju, B., Kishore, K., Guha, S. N., Panajkar, M. S., Rao, K. A., Raj, P., Suryanarayana, P., Sathyamoorthy, A., Datta, T., Bose, H., Prabhu, L. H., Sankaranarayanan, S., Shetiya, R. S., Veeraraghavan, N., Murthy, T. S., Sen, B. K., Joshi, P. V., Sharma, K. G. B., Joseph, T. B., Iyengar, T. S., Shrikhande, V. K., Mittal, K. C., Misra, S. C., Lal, M., Rao, P. S.
- ----------
- **We presented three papers at ICCF 1 March '90**

PART II : Tritium in Ni-H systems (1992 - 1996)

- We have observed Tritium both in :
- Ni cathode light water electrolytic cells (at ICCF 3 & 4)

  as well as

- H_2 gas loaded self heated Ni wires (at ICCF 5)

Studies in Ni Cathode-Light Water Systems

- Triggered by Randell Mills Fusion Technology paper of 1991 on Excess heat in Ni-H electrolytic cells;
- Mills however insisted excess heat in his cell NOT of nuclear origin, rather due to formation of compact hydrogen atoms or Hydrinos (Regretted publishing in FT)
- We Published Four Papers in Ni-H_2O (1992-96) :
- "Tritium and Excess Heat Generation During Electrolysis of Aqueous Solutions of Alkali Salts With Nickel Cathode" (ICCF 3, Nagoya,1992)
- "Further Studies on Excess Heat Generation in H_2O-Ni Electrolysis Cells " (ICCF 4, Maui, Dec,1993)
- "Two-Balance Method of Faraday Efficiency Measmnt. for Identifying Origin of Excess Heat in Ni-H_2O Electrolytic Cells" (SRI Intl.1994)
- (Have since withdrawn Excess Heat claims in open cells!)
- "Investigation of low-level tritium generation in Ni-H_2O electrolytic cells", (Fusion Technology,1996)

Tritium in Ni Light Water Cells

- In three different labs of BARC (Jan-Oct '92)
- Protocol compiled by Jed Rothwel (Jan '92)
- Open cells with Ni cathode & Pt wire Anode
- Porous Ni cathodes preferred; 2cm x 6cm long
- Carbonate solns of K, Na, Li (nat. & enrch Li^6)
- Typical Run time : a few weeks;
- Some runs with mixture of H_2O & D_2O
- 10 ml samples - distilled before counting for T
- Tritium carried away by the gas stream not included in the results
- At Nagoya : T in 18 out of 29 cells (>50%)

ICCF 3 (Nagoya) Paper
(7 out of 18 first set cells indicated Tritium)

Table I. Experiments Where DT was Measured for Various Input Power Levels.

Srl No.	Expt. No.	Date Comp—leted	No. of days	Alkali Type	Sol—vent	Max. Input Power P_j(w)	Max. Excess Power (%)	Tritium Content in Elect. (Bq/ml)
PISD GROUP (CYL. & PLANAR GEOMETRY; SOLID & POROUS Ni)								
1	X—8/12*	May,1	44d	K_2CO_3	H_2O	2.0w	40%	NIL
2	X—14†	Jun,3	16d	Li_2CO_3	H_2O	3.5w	30%	177
3	X—15†	May,21	10d	K_2CO_3	25%D_2O	1.6w	130%	3390
4	XA—1‡	Oct,20	30d	K_2CO_3	H_2O	1.2w		NIL
5	XA—2‡	Oct,20	30d	K_2CO_3	H_2O	1.0w		NIL
6	XA—3‡	Oct,20	30d	K_2CO_3	H_2O	1.3w	23%	NIL
7	XA—4‡	Oct,20	30d	Li_2CO_3	H_2O	1.0w	70%	NIL
8	XA—5‡	Oct,20	30d	Na_2CO_3	H_2O	0.8w	35%	NIL
HALL 5 GROUP (CYLINDRICAL GEOMETRY : POROUS Ni)								
9	A1	Jun,29	21d	K_2CO_3	25%D_2O		36%	NIL
10	A2	Jun,29	21d	K_2CO_3	25%D_2O		28%	NIL
11	A3	Jun,29	21d	K_2CO_3	25%D_2O	0.23w	28%	NIL
12	A4	Jun,29	21d	K_2CO_3	H_2O		24%	46
13	A5	Jun,29	21d	Li_2CO_3	H_2O		70%	1454
14	B1	Aug,25	28d	*Li_2CO_3	H_2O		40%	NIL
15	B2	Aug,25	28d	*Li_2CO_3	H_2O		58%	NIL
16	B3	Aug,25	28d	*Li_2CO_3	50%D_2O	0.3w	46%	513
17	B4	Aug,25	28d	Li_2CO_3	50%D_2O		49%	69
18	B5	Aug,25	28d	*Li_2CO_3	D_2O		53%	195

ICCF 3 Paper – Additional Cells

(7 out of 11 second set cells generated Tritium)

Table II. Other Excess Heat and Tritium Experiments.

Srl No.	Expt. No.	Cathode	Alkali	Solvent	Excess Power (%)	Tritium Level (Bq/ml)
1	NtPD–1	Solid Ni	K_2CO_3	25% D_2O	< 5%	88
2	NtPD–2	Por. Ni	K_2CO_3	H_2O	65%	NIL
3	NtPD–3	Solid Ni	Na_2CO_3	25% D_2O	72%	NIL
4	NtPD–4	Solid Ni	Li_2CO_3	H_2O	68%	NIL
5	TiB–1	Ti Button	Li_2CO_3	H_2O	10%	205
6	TiB–2	Ti Button	Li_2CO_3	H_2O	10%	124
7	TiF–1	Ti Foil	LiOD	D_2O	*	147
8	KSR–1	Por. Ni	K_2CO_3	H_2O	*	NIL
9	FP–1	Por. Ni	K_2CO_3	H_2O	*	310
10	OM–1	Por. Ni	K_2CO_3	25% D_2O	*	74
11	OM–3	Por. Ni	$^6Li_2CO_3$	H_2O	*	223

New Dedicated Liquid Scintillation Spectrometer Procured in May 1993

- Installed in Chem. Eng. Division where no other Tritium work done;
- Low Background ; Sensitivity ~ 1 Bq/ml
- Microdistillation of electrolytic solution samples done before counting;
- All stock solutions first checked for initial Tritium content prior to electrolysis !
- These results presented at ICCF 4

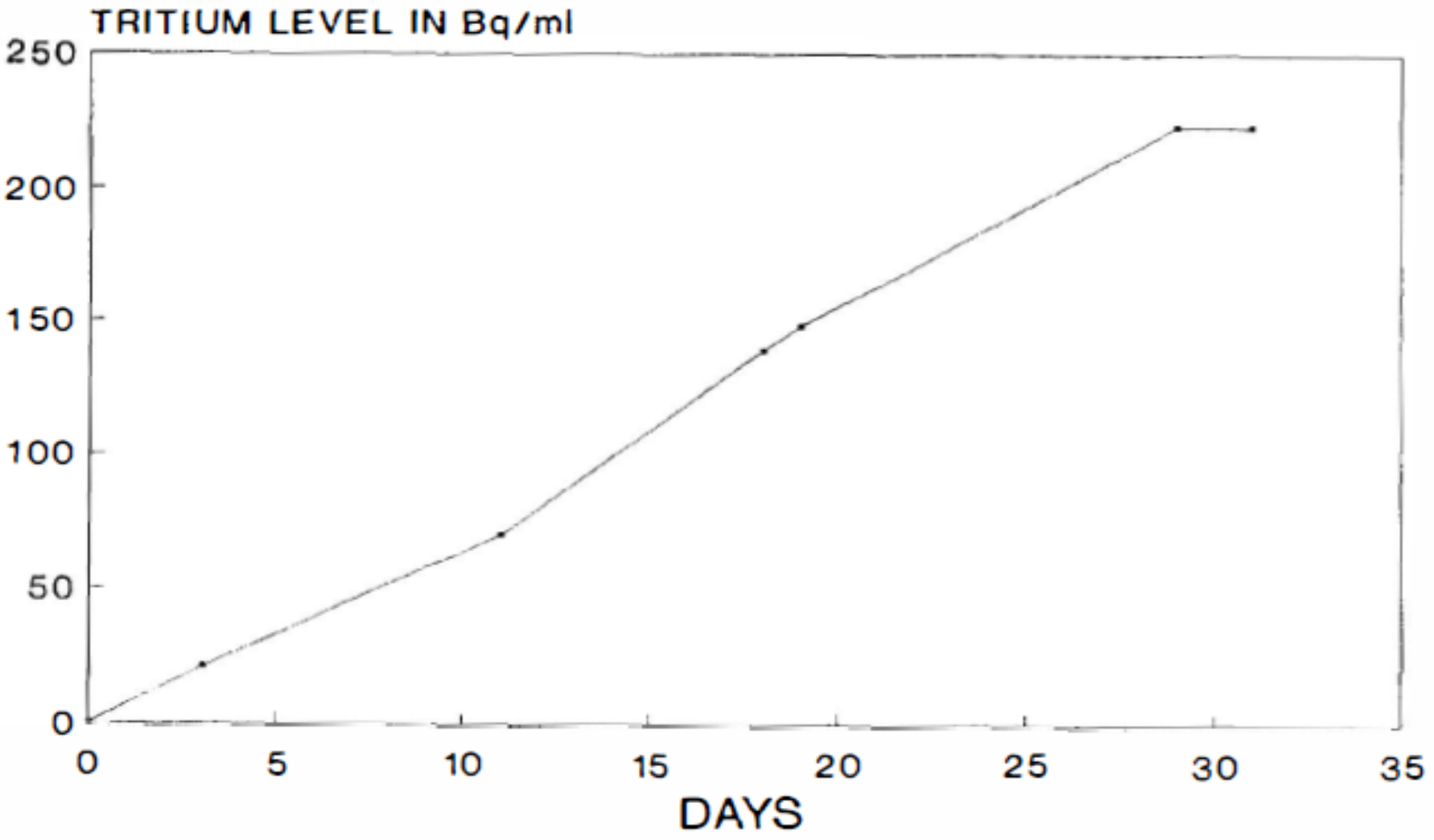
ICCF 4 Paper (FT-'96): Results from New Ni-H_2O Cells - June to Sept 1993 (8 out of 17 yielded Tritium)

Serial Number	Cell	Electrolyte[a]	Date of Sampling (1993)	Total Count/9 min	Tritium Activity (dpm/ml)
1	X0-1	K_2CO_3	July 5	252	---
2	XO-2	K2CO3	July 5	219	---
3	XO-3	(N)Li_2CO_3	July 5	228	---
4	XO-4	(N)Li_2CO_3	July 5	783[b]	260
5	XO-5	K_2CO_3	July 5	243	---
6	XP-1	(N)Li_2CO_3	July 12	246	---
7	XP-2	(N)Li2CO3	July 12	363[b]	56
8	XP-3	(N)Li2CO3	July 12	267	---
9	XP-4	(E)Li_2CO_3	July 12	278	---
10	XP-5	(E) Li_2CO_3	July 12	300	30
11	XQ-1	(N)Li_2CO_3	August 7	327[b]	40
12	XQ-2	(N)Li_2CO_3	August 7	264	---
13	XQ-3	(N)Li_2CO_3	August 7	396[b]	73
14	XQ-1	(E)Li_2CO_3	August 10	429[b]	88
15	XQ-2	(E) Li_2CO_3	August 10	360[b]	56
16	XQ-1	K2CO3	August 13	267	---
17	XQ-2	K_2CO_3	August 13	870[b]	290

Study of Temporal Variation of Tritium

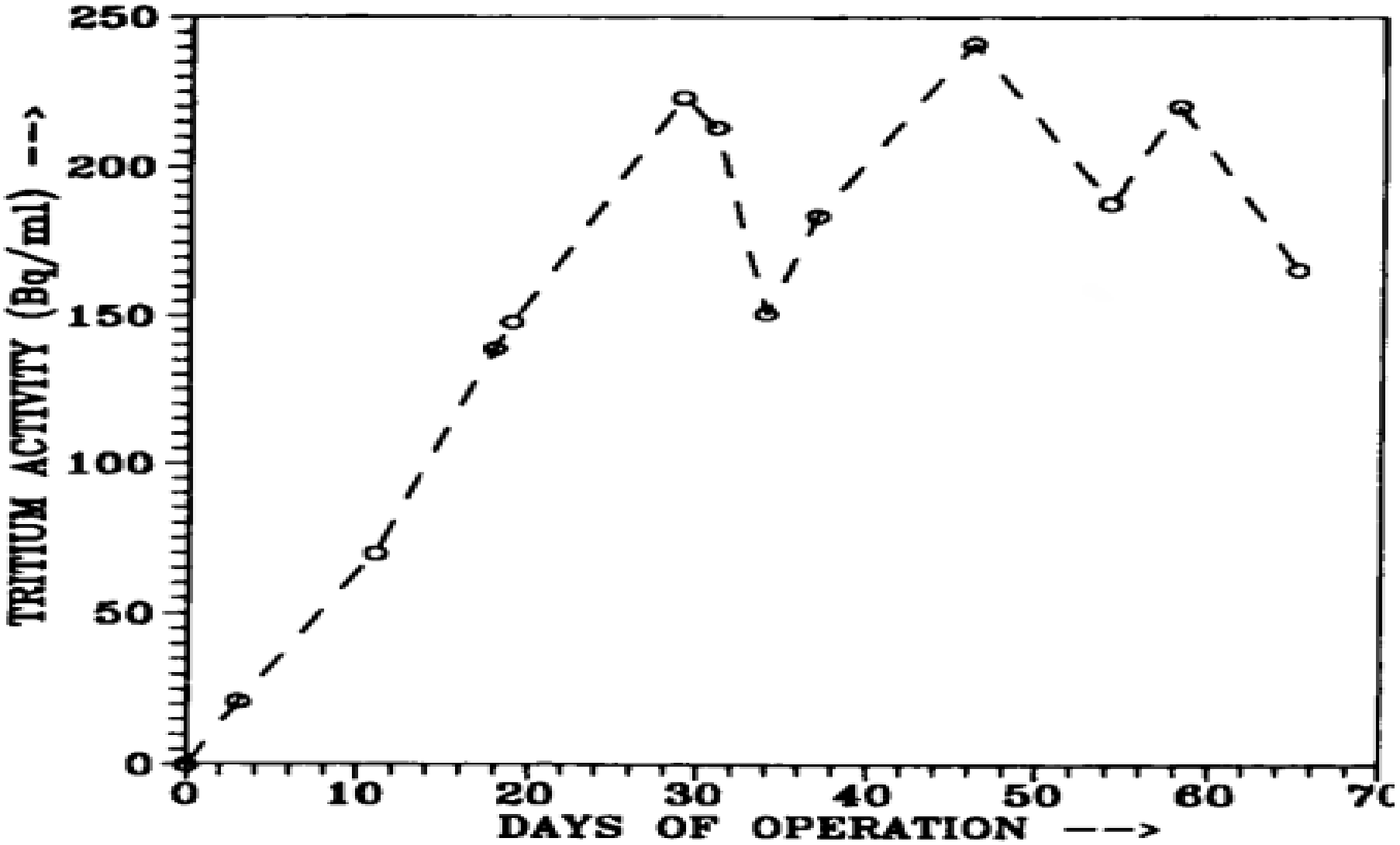
- 6 new cells set up using separating funnels so that samples can be drawn from bottom without disturbing cell (Designated OM-3 to OM-10).
- Flat plate cathodes, Pt wire Anodes
- Samples drawn at intervals of 3~10 days

Ni Cathode - Separating Funnel Cell # OM-3 : 54% Enr. $^6Li_2CO_3$ in H_2O - Showed Continuous Build up of T (Sept. 5th - Oct. 6th '92)

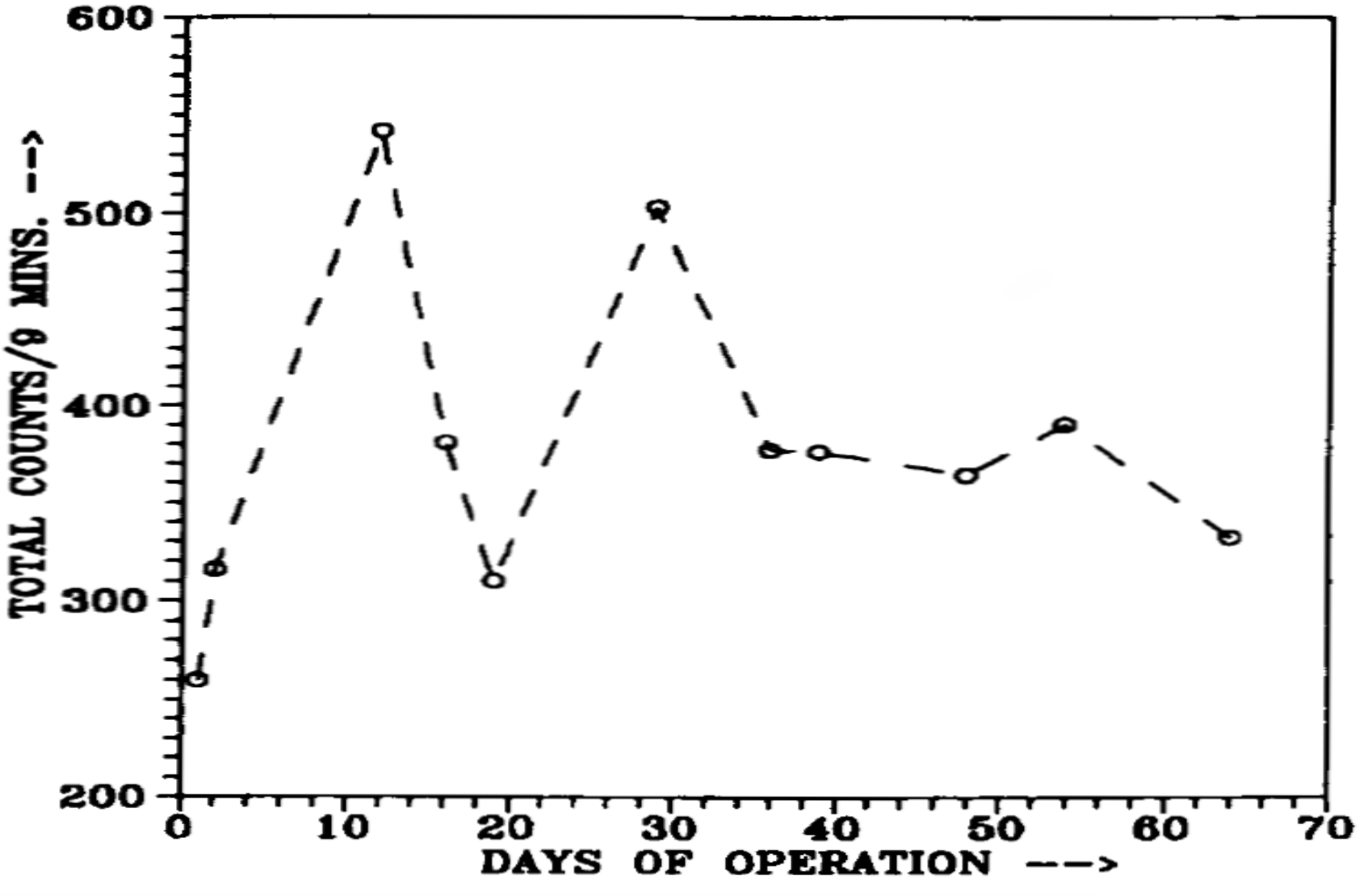


Cell #3 : Variation of T (Post Nagoya)

Suggests Scavenging & Saturation



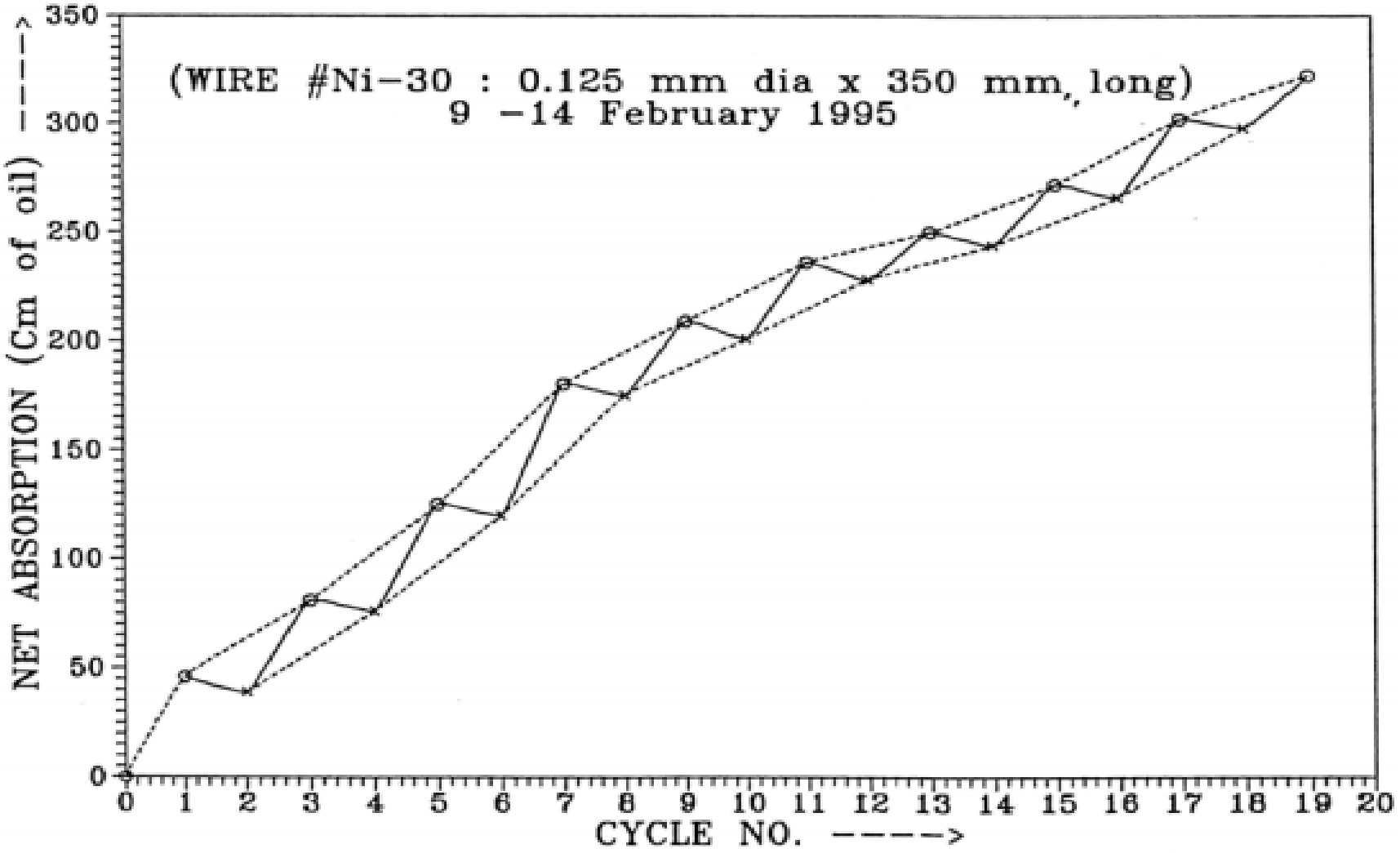
Cell # OM-7 : Also shows two opposing Processes at work : Production & Removal



Self Heated Ni Wire Experiments

- 125 or 380 μm, 35 to 50 cm long; coiled as spring
- Wire temp. controlled through (R/R_0) ratio
- Standard Glass Vacuum line
- Double walled quartz glass cell with tungsten leads
- Absolute Pr. of H_2 gas in system Hg manometer
- Absorption of gas measured through a sensitive silicone oil differential manometer
- V-I characteristics firstly measured both in Vacuum & H_2 gas atmosphere
- Best absorption in 200 C to 300 C region
- Quantum of absorption surprisingly high (> unity !) due to self heated method (magnetic field role?)

Absorption/Desorption of Hydrogen During Repeated cycles of Heating/Cooling



Quantum of H_2 Gas Absorbed : Pr. Drop Cm of Silicone oil in Differential Manometer

Sr. No.	Wire #	Dimensions Dia mm	length cm	Mass mg	Duration of loading	Number of cycles	Net Loading
1	Ni-23	.125	50	53.8	7th OCT to 13th OCT	5	*
2	Ni-501	.38	50	500	24th OCT to 27th OCT	5	125
3	Ni-501	.38	50	500	23rd OCT to 9th DEC	15	203
4	Ni-24	.125	50	54.1	12th DEC to 15th DEC	5	58
5	Ni-27	.125	45	48.6	18th JAN to 25th JAN	6	436
6	Ni-30	.125	35	39.4	9th FEB to 14th FEB	10	325

Self Heated Ni Wires following several H_2 loading cycles :

Variation of Tritium in cut piece (Last Col gives Bqs)

Background was 250 cts/10 mins; Excess over BG given in col 3.

Sample No	Piece id No	Counts/10 mins	Bq/5 ml
1	**Standard**	4200	170
2	501/1	532	22
3	504/4	152	6
4	504/5	70	3
5	504/5/11	103	4
6	24/1	410	18
7	24/2	690	28
8	24/3	1150	47
9	27/1	950	38
10	27/2	704	28
11	27/3	57650	2333
12	30/1	1560	63
13	30/2	220	9
14	30/3	550	22

Summary of BARC Tritium Measurements

- Bockris & I had this friendly dispute as to who reported Tritium in CF/LENR first!
- We presented T and branching ratio anomaly at Karlsruhe in July 1989 !
- We first reported T in Ni-H_2O at Nagoya in 1992; Wonder if ours the first reports of a nuclear signature in a Ni-H system?
- Todate we have reported finding T in 25 out of 52 Ni-H_2O cells; (~ 50% success?)
- Our Plasma focus T results triggered interest in plasma loading (Romodanov)

"Reproducible Tritium Generation using Glow Discharge Plasma interacting with Metals such as W, Mo etc with Pulsed Currents and External Magnetic Field"

(>16 papers during 1991 to 2003)

V.A.Romodonav et al

SRI of SPA Lutch

Podolsk, Moscow region, Russia

Papers in 6 ICCF Confs. (3 to 10)

- Planar Glow Discharge; (Geometry important)
- Hollow Cathodes (Moly, Tungsten, Nb and Ta);
- H_2 flows through wall of cathode into plasma discharge chamber; "Transfusion thru membrane"
- Gas evacuated into rubber "pillows"; burned with air to form water; 0.2 ml of this mixed with liquid scintillator for counting tritium level;
- Mag. Field of 0.2 to 0.2 Tesla Perp. to cathode using a Permanent Magnet; increased tritium production rate by two orders of magnitude !
- Reported reproducible generation rate of 10^{10} T atoms/s, even with Nat.H_2

Comment

In the light of our extensive Tritium Results I would be VERY surprised if Rossi & Defkalion and others observing large excess heat in Ni-H systems don't detect Tritium in their spent fuel samples or hydrogen gas !

Thank you !