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EMF DEVIATION OF CHROMEL/ALUMEL THERMOCOUPLES CAUSED BY NEUTRON IRRADIATION

(Irradiation Experiment TIC-01)

by

R. GOTTSCHLICH and M. BELLEZZA

1966

Joint Nuclear Research Center
Petten Establishment - Netherlands

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A further purpose of the experiment was to get experience in handling and remote welding of radioactive wires.

The capsule containing bare wires for 20 thermocouples was irradiated in HFR position F-2 for two reactor-cycles, corresponding to average neutron fluxes of 4.5 $\times$ 10^{20} cm^{-2} thermal and 1.4 $\times$ 10^{20} cm^{-2} fast.

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After a long period of cooling, the capsule has been dismantled in a cell of the hot laboratory of RCN and the calibration has been performed inside another hot cell.

Two calibration methods have been applied :

a) Calibration of remote welded thermocouples, each of them consisting of one irradiated and one non-irradiated wire of the same composition.

b) EMF comparison of thermocouple pairs welded from irradiated wires, with non-irradiated thermocouples made from wires of the same origin.

The obtained results showed a negative EMF drift of irradiated thermocouples of 0.8 % (average value).

The remote welding procedure is treated separately in the Appendix.

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SUMMARY

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CONTENTS.

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1. Statement of problems (*)

Temperature measurements in the nuclear field (measurement of temperatures in in-pile rigs and temperature measurement of reactor components) require a high degree of reliability and accuracy.

The measurements are carried out mainly by means of thermocouples, because of their simplicity for nuclear application. Thermocouples cover a wide range of temperatures, they are available in sheathed form (a favourable feature in view of compatibility problems and leaktight penetrations through pipe and tank walls), and they have neither special nor complicated in-pile parts.

However, the properties of thermocouple materials undergo changes in the presence of nuclear radiations, and the EMF may change due to transmutation, nuclear alloying and hardening effects.

Transmutation implies conversion to the next heavier unstable nuclide formed by neutron capture. This is usually followed by β^- decay.

The hardening effect is due to lattice defects produced by fast neutron and gamma irradiation. At high temperatures this effect becomes negligible because of continuous annealing.

In temperature regions up to 1000 °C Chromel/Alumel thermocouples are commonly used (low cost, availability and stable EMF).

Their behaviour under irradiation has been already investigated several years ago (ref. 2,3,4). The main conclusion obtained was that the drift of the EMF of Chromel/Alumel couples remains within the accuracy standards of unirradiated couples up to a thermal neutron dose of 10^{21} nvt.

2. Capsule assemblage and irradiation.

2.1. Description of capsule design and construction.

The capsule (shown on figure 1) was of a non-instrumented type. All parts of it were made of aluminium.

The capsule consisted of three main parts :

- the outer cylinder (50 mm o.d.)
- the sample holder with the special top cone and the loading eye
- and the 20 rings for the thermocouple wires.

(*) Manuscript received on November 1966

The capsule had a central cooling channel to prevent an unacceptably high temperature, and was filled with helium (1,5 kg/cm^2 abs.)

On each of the rings one Chromel and one Alumel wire (1 m length) were wound. The wire ends were fixed in small grooves in the inner surface of the rings. The spool shape for loading of the thermocouple wires was chosen in order to obtain an uniform flux distribution over the wire length (necessary to avoid calibration errors due to the temperature gradient of the furnace).

2.2. Pre-irradiation test of the sample material.

The thermocouple wires were furnished by the Hoskins Company via the Dutch representative.

T.C. type : Chromel - P/Alumel 3-G-178 ; 0,32 mm Ø .

Before installation in the capsule, several wires have been examined with regard to their homogeneity. The work was carried out at Mol, using the test set-up in the instrumentation laboratory of GEX.

In this test, a hot region is moved at a constant speed along the length of the wire, and the EMF thus produced is amplified and recorded on a high speed recorder.

The wires showed the following maximum inhomogeneity values :

Chromel - (temp. $\sim$ 500° C) : 15 μV max.
Alumel - (temp. $\sim$ 500° C) : 10 μV max.

2.3. Irradiation

The capsule has been irradiated for two HFR cycles in reflector position F-2.

For monitoring of the thermal and fast flux, the capsule was equiped with Al/Co, Ni and Fe foils and wires.

Monitor evaluation produced the following average values :

$4,5 \times 10^{20}$ n/cm^2 thermal (flux : $1,2 \times 10^{14}$ cm^{-2} s^{-1})
$1,4 \times 10^{20}$ n/cm^2 fast (flux : $3,7 \times 10^{13}$ cm^{-2} s^{-1})

Due to the flux distribution over the capsule length the maximum and minimum values were $\pm$ 10 %.

Cooling time before examination was 18 months.

3. Post-irradiation examination

3.1. Dismantling of the capsule

The dismantling took place in a hot cell of the LSO laboratory of RCN.

The outer cylinder of the capsule (together with the top section) was first removed. This revealed the cylindrical carrier, from which the thermocouple wires could be easily unwound by rotating the rings.

The dismantling procedure is described in detail in EURATOM Internal Note : IP-T 138.

3.2. Welding of thermocouples and remote handling.

The radioactivity of one irradiated T.C. was 100 r/h at 5 cm necessitating the remote handling and welding of the wires.

The following equipment was installed on a working table in a hot cell :

- the calibration furnace (resistance heated tube furnace with a large temperature equalization block),
- the cold junction DEWAR,
- the welding machine for the T.C. junctions,
- the support for the thermocouple carrier,
- a radiation shield between furnace and DEWAR.

After unwinding, the thermocouple wires had to be straightened. In order to avoid cold working effects, this was done as carefully as possible using manipulator claws.

Next, the wires were insulated by means of Al_2O_3 capillaries, welded to non-irradiated wires from the same batch of material, and connected at the cold junctions to extension leads. The prepared hot junctions were then inserted into the temperature equalization block in the furnace.

Calibration and control instruments were all located outside the cell.

Further information on the handling and the welding procedure is included in the appendix.

3.3. Calibration of thermocouples.

In order to detect the EMF changes caused by irradiation, two calibration methods were adopted :

- a) Welding irradiated and non-irradiated wires of the same composition together, and measurement of the EMF thus produced at known temperatures.
- b) Comparison of thermocouples made from irradiated Chromel and Alumel wires with non-irradiated thermocouples of the same wire batch.

The calibration took place under normal atmospheric environment.

Method a)

The EMF of Chromel (irradiated) /Chromel pairs and Alumel (irradiated)/ Alumel pairs were measured at 3 temperatures : $\sim$500, $\sim$750, and $\sim$ 1000° C.

Calibrations were performed by means of a Bleeker precision potentiometer (maximum sensitivity 10^{-6} volt).

Balance was achieved using a mirror galvanometer.

Temperatures were measured with a Pt/Pt- 10% Rh thermocouple, which was connected to an electronic voltmeter and a potentiometric recorder.

The measured EMF values are given on figures 6, 7, 8, 9.

The circuit diagram is outlined on figure 4.

The two photographs (Fig. 2 and 3) show the whole measuring and calibration set-up inside and outside the cell.

At the end of the measurements a "zero test" was made, using two identical non-irradiated T.C. wires (Alumel/Alumel), and showed that the switching arrangement used introduced an error of 5 μV, at 800°C.

Method b)

In this measurement, irradiated and non-irradiated Chr/Al thermocouple pairs were compared against one another.

Two types of calibration were made :

1) The EMF output of the standard (non-irradiated) and the irradiated thermocouples were both measured, using the potentiometer described above (see fig. 5 I).

2) The standard and the irradiated thermocouples were connected in opposition, and the differential output amplified and measured by means of the galvanometer incorporated in a Leeds & Northrup d.c. amplifier (see fig 5 II).

Figure 10 illustrates differences between the two thermocouples and were obtained by means of arrangement shown in fig. 5 II. These values at high temperatures were also verified using the arrangement shown in fig. 5, I and found to correspond with the values obtained by the differential method.

The results were corrected to a cold junction temperature 0° C and the temperature distribution in the furnace block due to different immersion depths taken into account.

Fig. 11 shows the axial and radial temperature distribution inside the equalization block.

The cold junction extension wires were of Chromel to facilitate welding.

4. Discussion of the results.

The results of the experiment can be summarized as follows :

Within the temperature range 250 to 1000°C the irradiated Chromel/Alumel thermocouples gave a lower EMF (μV/°C) than the non-irradiated pairs. This was measured from all the irradiated thermocouples.

It was found both Chromel and Alumel after irradiation had become "more positive" in comparison to unirradiated wires. The effect was more evident with Alumel.

This is in agreement with observations made by Kelly, Johnston, Baumann (ref. 1) , the overall effect is that the difference in polarity of irradiated T.C. wires is smaller than those of non-irradiated wires or in other words an irradiated Chromel/Alumel thermocouple gives less μV/°C.

Fig. 10 shows the drift in EMF as a function of temperature of five different couples made from irradiated Chromel/Alumel wires.

The average drift lay somewhat below 1 %, and it may be seen that the deviation caused by neutron irradiation of 4,5 x 10^{20} n/cm^2 thermal and 1,4 x 10^{20} n/cm^2 fast does not exceed the fabrication tolerances by a significant amount.

When comparing our experimental results with those of Kelly et al. (1) and Lupoli et al. (3), it can be concluded that the drift measured by us is larger.

The drift occuring in Alumel after irradiation is more temperature sensitive than that of Chromel (see figs. 6 and 7). Further investigation of this effect would require irradiations to higher doses.

It is felt that the measured nuclear effects were due to transmutation.

The effects of fast neutron (hardening) and cold working by handling damage are mainly eliminated by annealing.

In order to estimate the magnitude of these effects, samples of irradiated T.C. wires were recalibrated after a short term annealing and thermal cycling.

Although some change in EMF drift can be seen in the Alumel wire in fig. 9, results for the completed thermocouples presented in figure 10 (curve Ib) show the overall change to be negligible.

5. Conclusion.

Chromel/Alumel thermocouples are suitable for use under reactor conditions with regard to the nuclear stability of their composition up to a dose of at least 5×10^{20} cm^{-2} thermal and fast neutrons.

The experiment was not related to any compatibility test, longtime drift test or mechanical investigation.

It was felt however that much effects are probably of more importance than transmutation, even after irradiation at higher doses.

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6. Appendix.

The remote welding device.

The radioactivity of irradiated thermocouple wires involved the use of a suitable remote welding device in order to carry out the hot and cold junction welds. Since such a set-up was not available, a microwelding device for in-cell operation was developed and built in the Petten centre.

The equipment is of the arc welding type, and is assembled in two groups :

A) The control unit, which is located outside the hot cell to facilitate operation and maintenance.

B) 1. The electrode holder.
This may be positioned by means of an electric motor , incorporating a solenoid which in association with the soft iron mounting of the replacable welding electrode forms a variable timing unit.
2. The work head.
This is provided to clamp the wires in a pre-determined position during welding, and to this end includes a device to cut the wires at the point where the weld will be made. A welding chamber is formed in the front face of the block to provide an inert gas shield during welding. The rear face of the block is chamfered to facilitate remote insertion of the thin and fragile wires.
3. Supports.
The free lengths of Al_2O_3 capillary insulated wires are supported by two fixtures.

Operation.

After fixing, centering and cutting of the wires the operation proceeds semi-automatically.

First, the complete electrode holder assembly is moved forward by an electric drive motor until a pre-determined contact pressure is established between electrode and wires. The supply to the motor is then cut, by means of limit switches.

Next, welding is initiated by manually closing a pair of contacts in the control console, one of which completes the welding circuit whilst the other energises the solenoid which retracts the electrode itself. The time for this operation is governed by an adjustable core within the coil. Movement of this core, alters the magnetic gap between itself and the soft iron electrode holder/armature. This operation both strikes the arc, and controls its duration. The junction thus welded takes the form of a compact ball-shaped bridge connecting the wire ends. Observation of the completed weld is made through windows in the walls of the welding chamber using binoculars located outside the hot cell. In the case of an unsactisfactory fusion at the first attempt, welding may be repeated without re-setting the wires.

Finally, the drive motor is reversed and the assembly withdrawn from the work head.

A simplified diagram of the welding circuit is given in fig. 12.

The technical and electrical data of the welding machine are :

- overall length	: 90 cm
-max. cross section	: 10 x 10 cm
- weight	: 24 kg.
- welding voltage	: up to 60 volt (d.c. or a.c.)
- welding power	: ~ 2 kw
- welding capacity	: wire sizes 0,15 to 0,5 mm Ø
- inert gas	: argon.

For the experiment TIC-01 an a.c. version of the welding device was used because of its simplicity.

The welding of thermocouples of the irradiation experiment TIC-01 was carried out in the following way.

After straightening, the radioactive wire was welded to a pulling wire which was already threaded inside the ceramic capillary. This was then pulled through one of the capillary holes.

The next step was the welding of the hot junction itself either with an unirradiated wire (which was already pulled into the second hole of the capillary) or with an irradiated one, which had already been pulled into the capillary as described above. Finally the cold junctions were welded.

x

x x

ACKNOWLEDGEMENT.

The author wishes to acknowledge the contribution of several persons who took part in the experiments.

Homogeneity tests on the sample material were carried out by Mr. Mathieu,G.E.X., BR.2, Mol.

The Reactor Department of RCN took care for assembly and irradiation of the capsule.

The RCN dosimetry group performed the flux measurement.

Operators of the LSO hot cells participated during several phases of the post-irradiation work.

The remote welding of the irradiated wires was developed and carried out by Mr. Bellezza (see appendix).

Mr. Bufalari performed the EMF measurements.

x

x x

7. References.

1. M. J. Kelly, W.W. Johnson, G.D. Baumann
"The effects of nuclear radiation on thermocouples".
Temperature, its measurement and control in science and industry.
(Reinhold Publishing Corp. New York 1962). Vol. III

2. G. Bianchi, S. Moretti
"Thermocouple irradiation test experiment".
CNEN arch. (62) 184

3. P. Lupoli, S. Moretti
"Post-irradiation calibration of Chromel/Alumel thermocouples".
CNEN arch. (63) TR 21.

x

x x

8. List of tables and figures

x

x x

TABLE I - Calibration data of Chromel irr./Chromel and Alumel irr. /Alumel pairs (belonging to curves on figures 6, 7, 8, 9)

Curve	Temp. level (°C)	Chr.irr/Chr (μ V)	Alirr/Al (μ V)
Ia	502	65	125
	742	70	130
	997	20	150
Ib	518	50	93
	760	65	110
	1014	22	132
II	508	35	116
	732	45	161
	974	22	185
III	487	11	93
	723	21	116
	977	23,4	
IV	548	23	77
	751	39	125
	979	92	120
V	518	77	99
	745	83	125
	989	4	137

TABLE 2 - Calibration data of irradiated Chromel/Alumel thermocouples.
(belonging to curves on fig. 10)

Thermocouple Ia		Thermocouple Ib		Thermocouple II	
Temperature (°C)	(negative) drift (μV)	temperature (°C)	(negative) drift (μV)	temperature (°C)	(negative) drift (μV)
248	15	552	158	313	12,5
334	47	591	210	366	20,5
419	69	659	250	419	37
491	105	688	270	551	39
541	166	735	315	620	65
591	222	810	330	649	88
640	265			688	120
688	302			735	160
735	331			782	196
828	387			828	240
873	425			873	275
				918	330
				935	380

Table 3 - Calibration data of irradiated Chromel/Alumel thermocouples
(belonging to curves on fig. 10)

Thermocouple III		Thermocouple IV		Thermocouple V	
temperature (°C)	(negative) drift (μV)	temperature (°C)	(negative) drift (μV)	temperature (°C)	(negative) drift (μV)
610	210	211	76	234	130
640	210	257	110	257	156
688	220	280	124	280	174
735	235	334	140	345	190
754	245	398	152	387	185
800	260	501	160	450	176
846	284	591	182	521	186
873	294	688	235	551	200
		770	320	662	204
		809	404	735	228
				782	235
				872	235

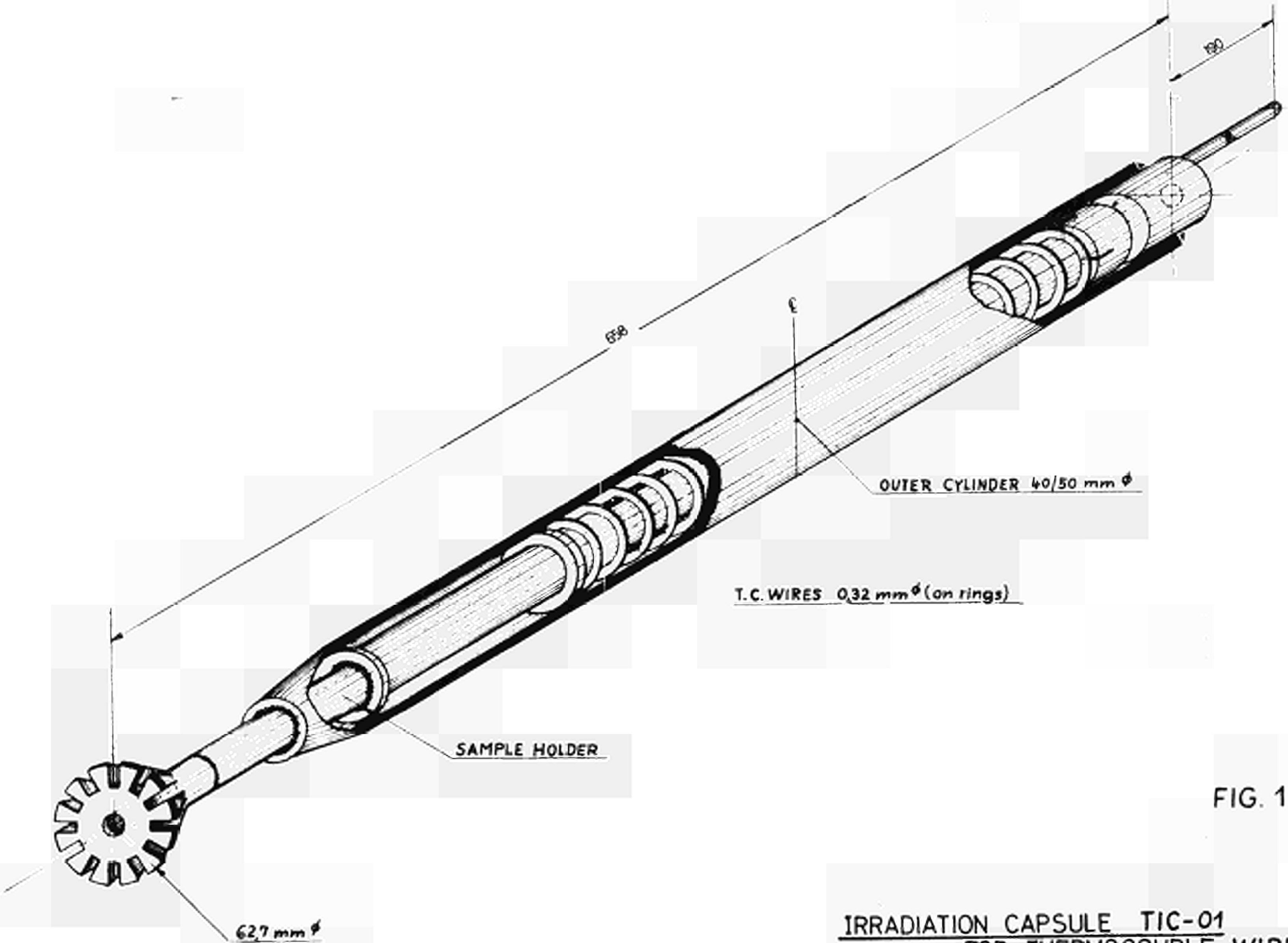


FIG. 1

IRRADIATION CAPSULE TIC-01
FOR THERMOCOUPLE WIRES

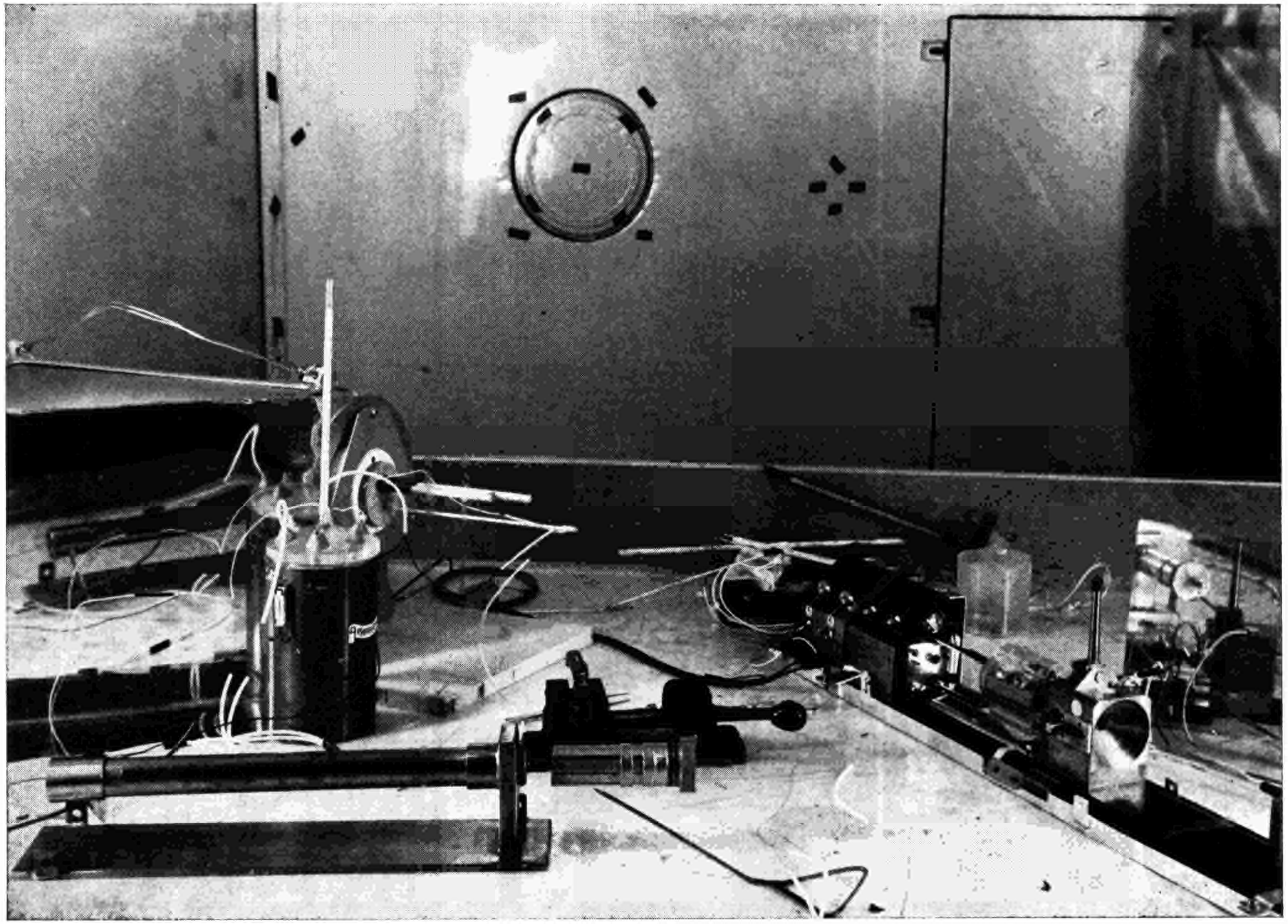


Fig. 2 : View into hot cell showing the calibration set-up
(Left side : furnace, DEWAR, sample holder - right side : welding device)

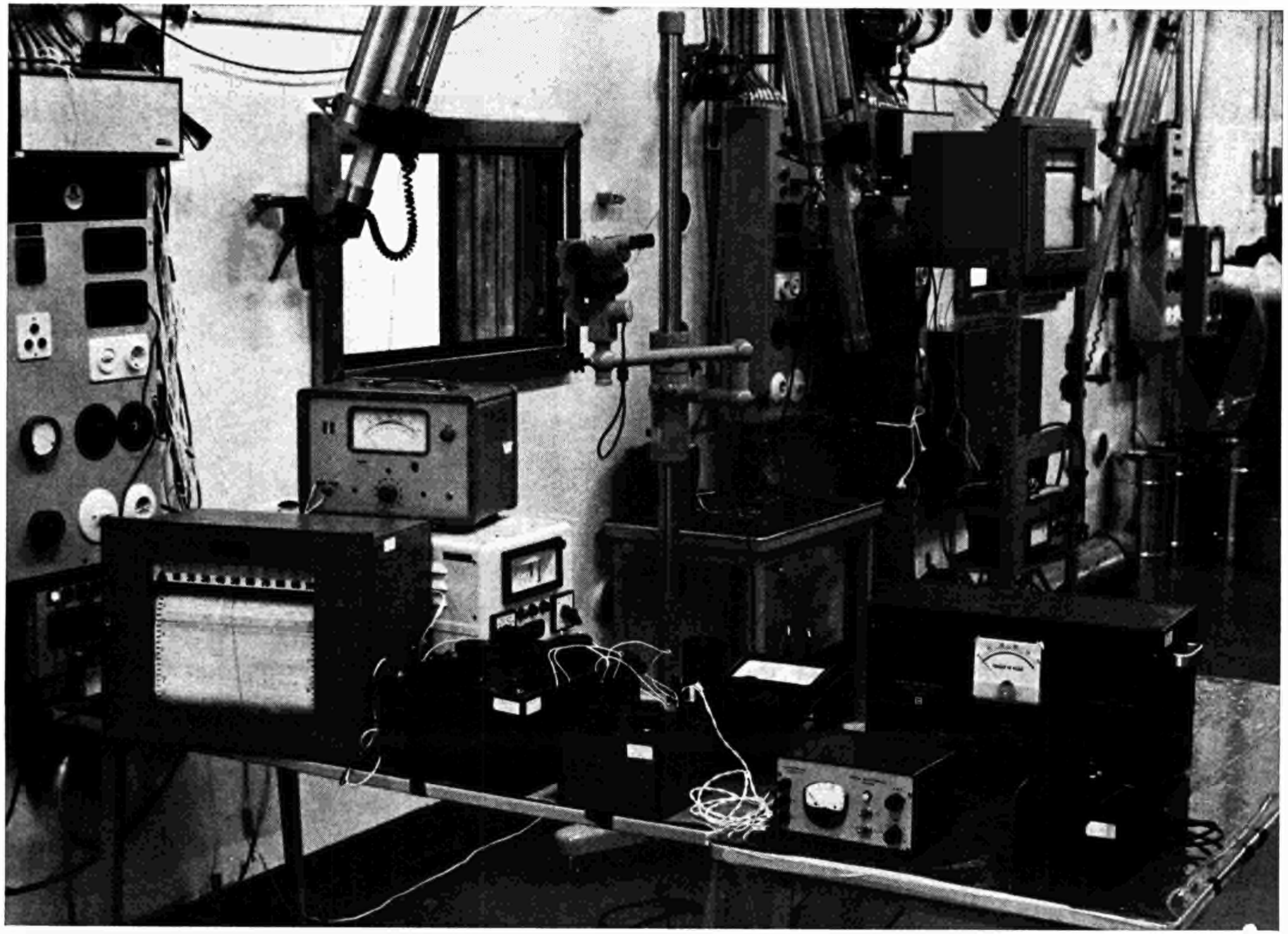
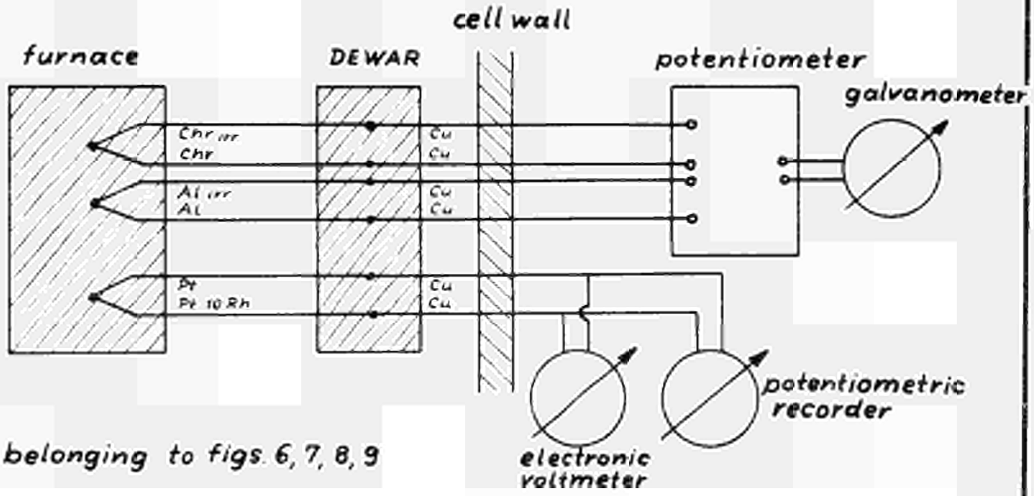


Fig. 3 : Instrument set-up outside hot cell



belonging to figs. 6, 7, 8, 9

FILE

Calibration circuit diagram

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FIG. 4

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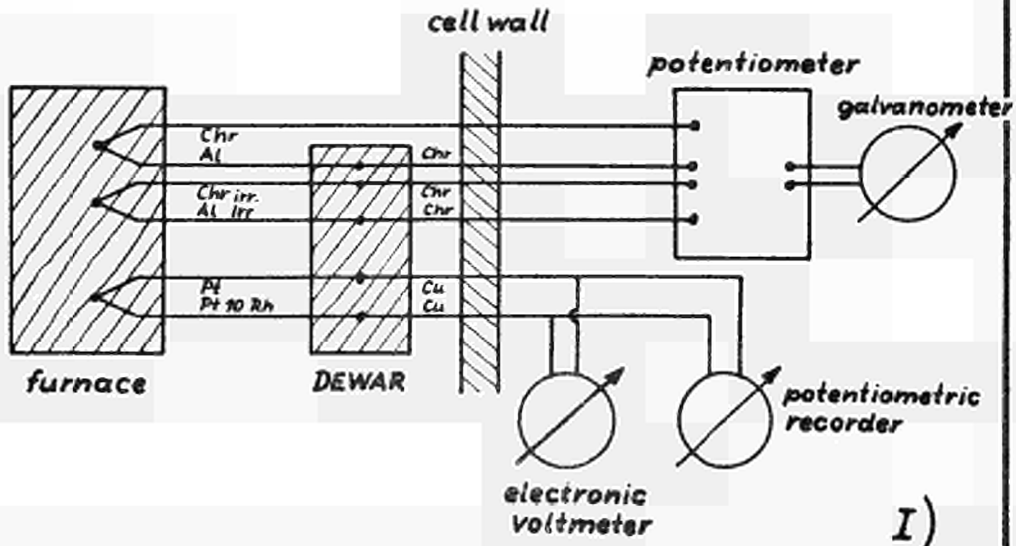

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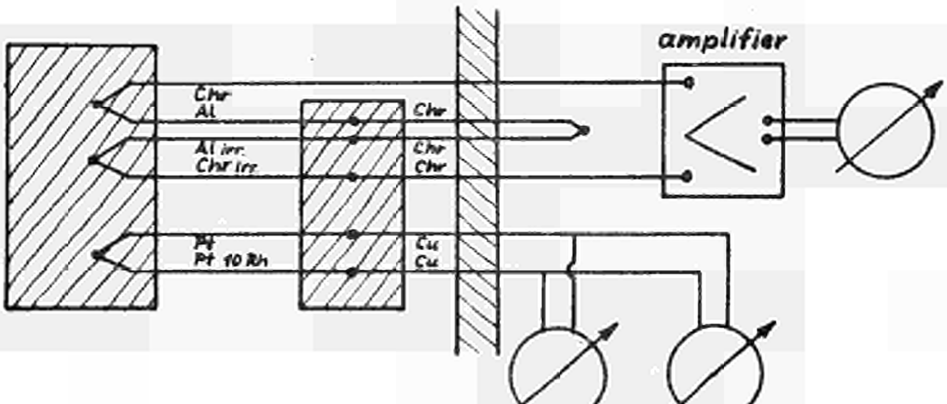

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FILE	Calibration circuit diagrams (belonging to figure 10)	EURATOM PETTEN
I I I I I I I I I I	DATE BY	FIG. 5

Utilisation

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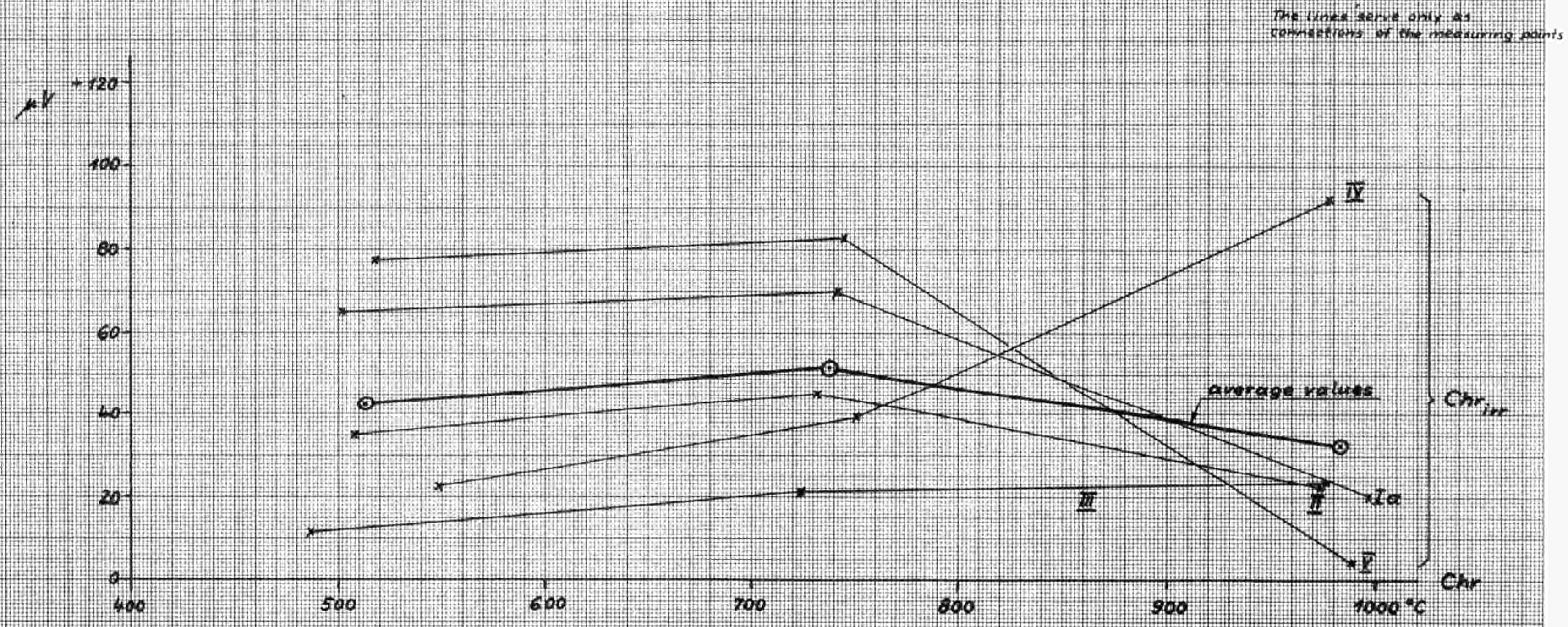
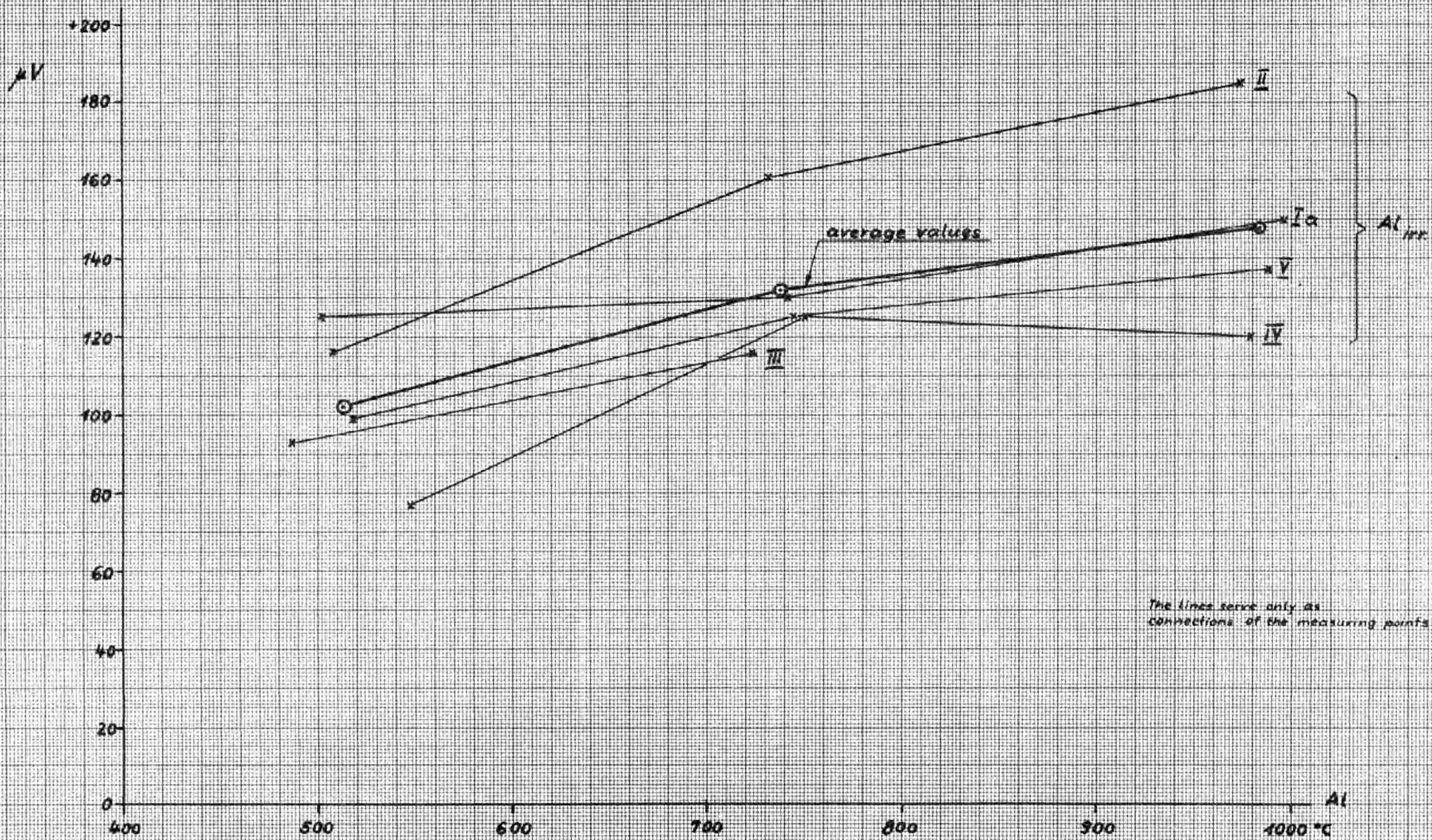


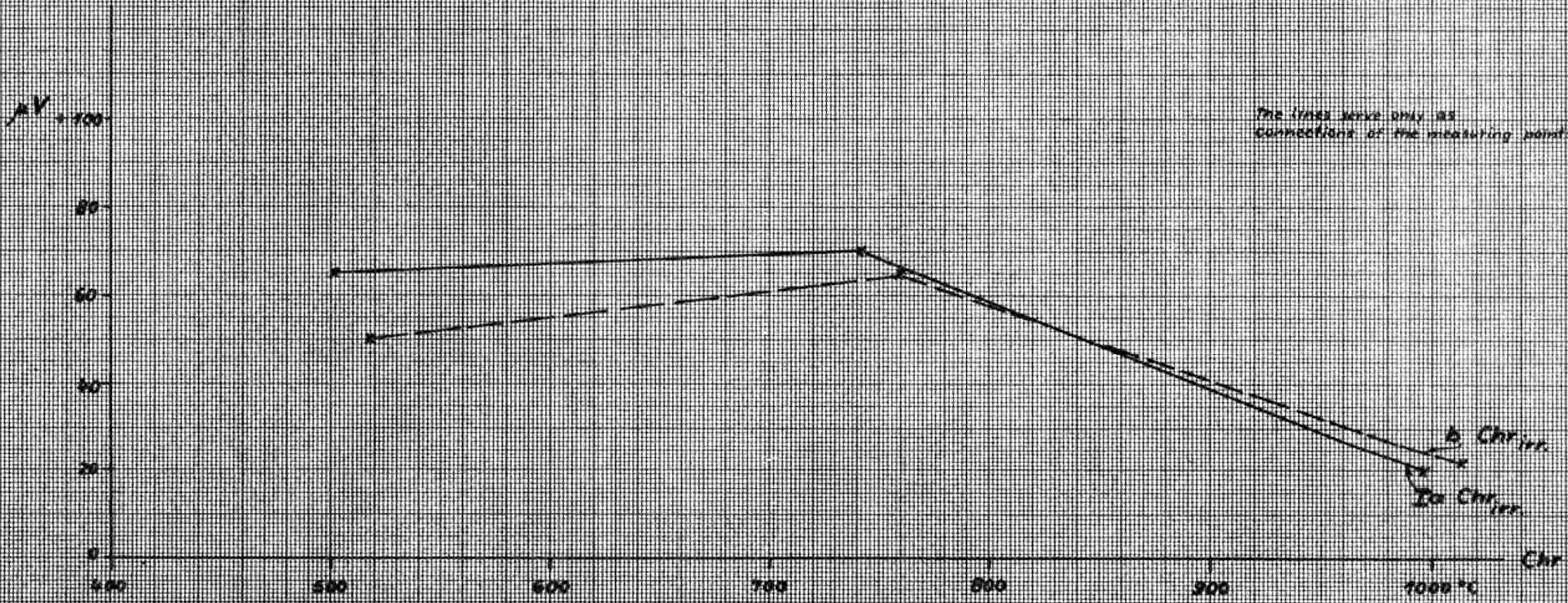
EMF of Chromel$_{irradiated}$ against Chromel$_{unirradiated}$ pairs

FIG. 6



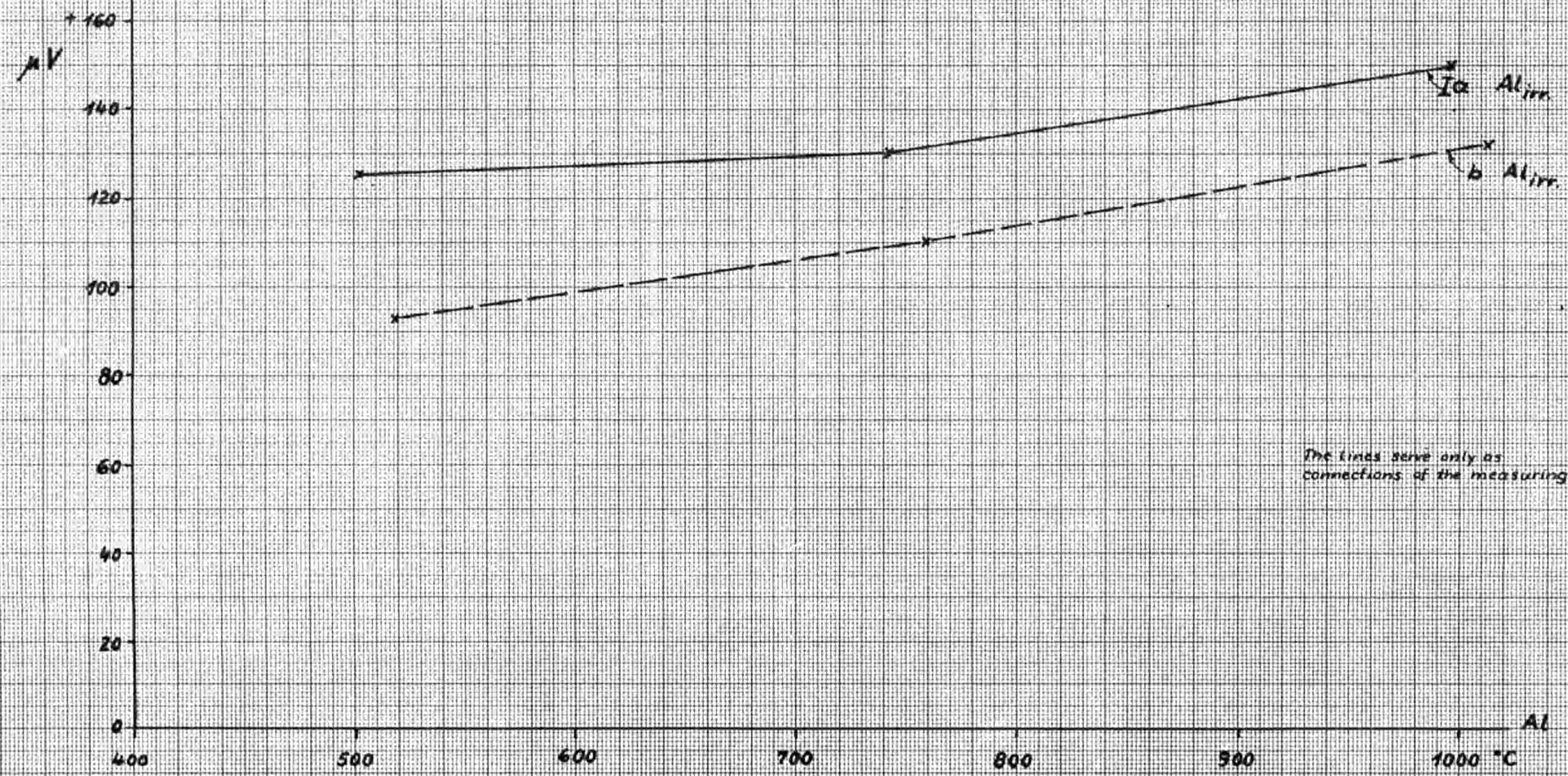
EMF of Alumel irradiated against Alumel unirradiated pairs

FIG. 7



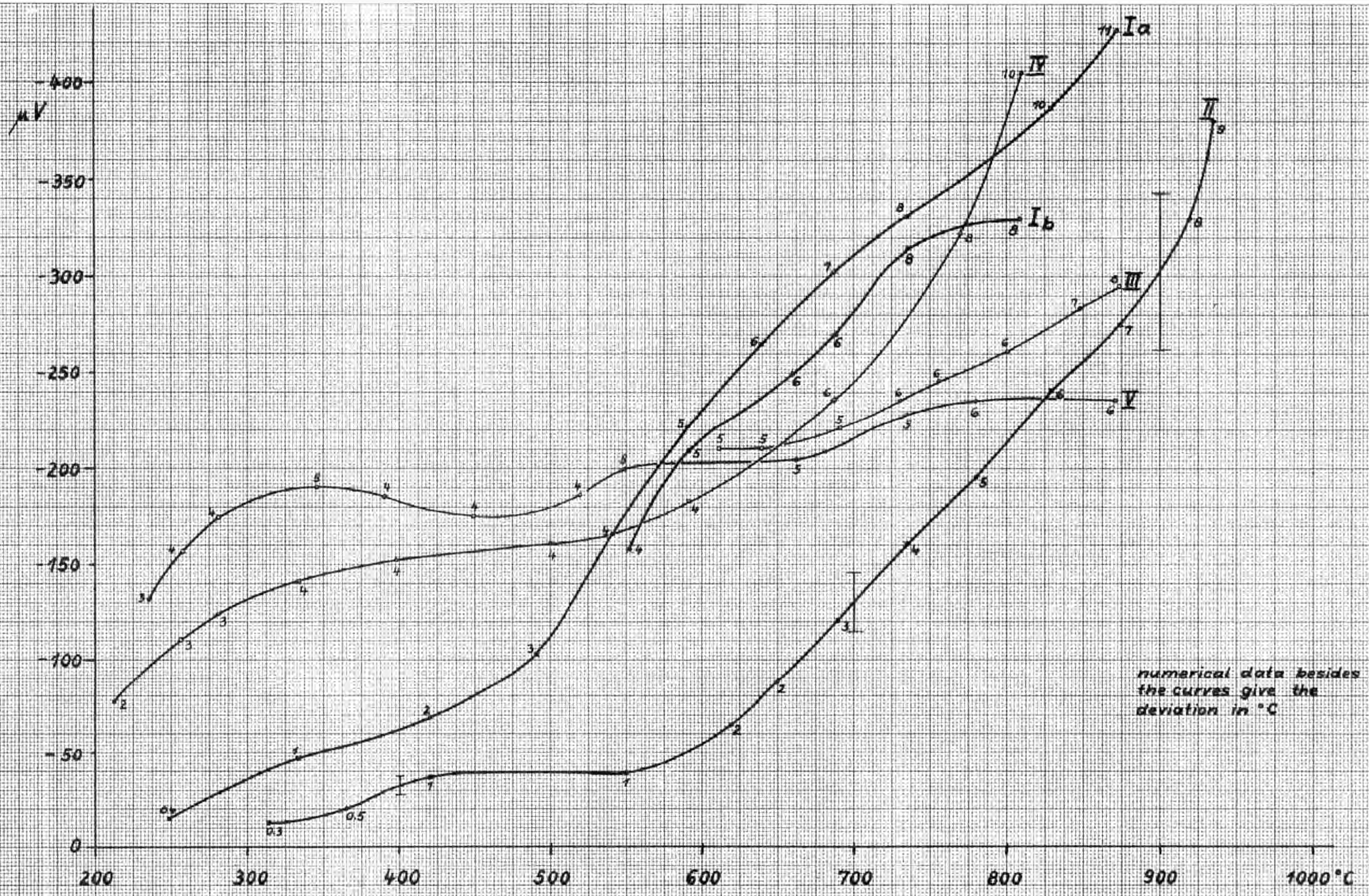
EMF of Chromel$_{irradiated}$ against Chromel$_{unirradiated}$ pair (curve b after annealing)

FIG. 8



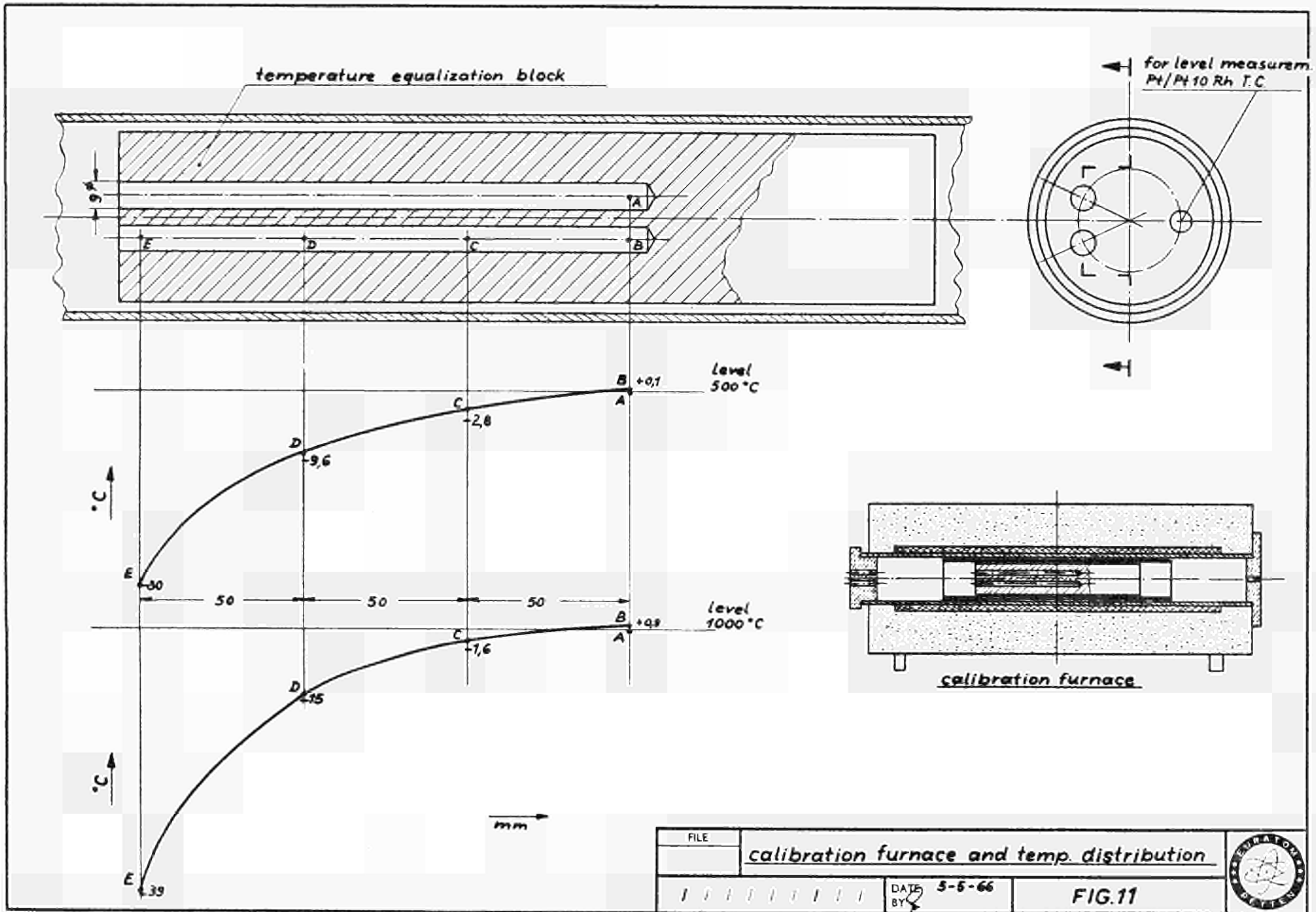
EMF of Alumel$_{irradiated}$ against Alumel$_{unirradiated}$ pair (curve b after annealing)

FIG. 9



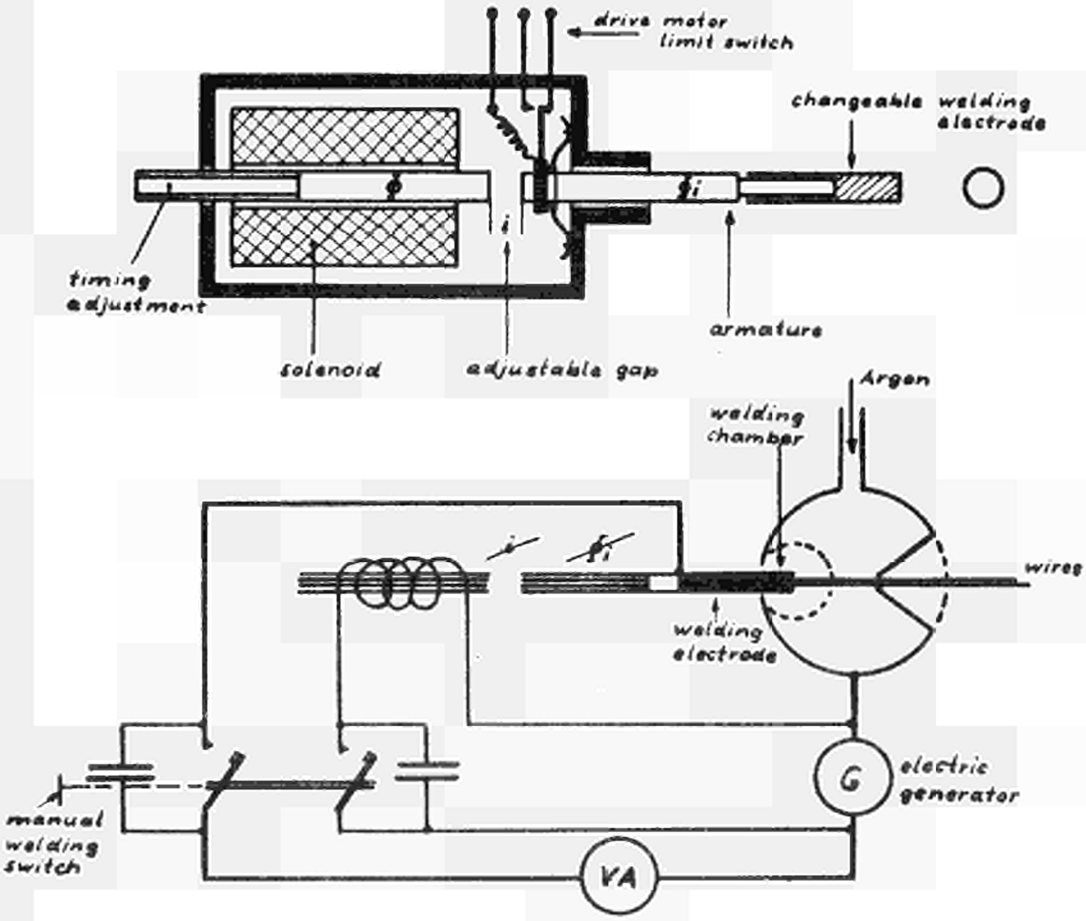
ΔEMF (negative drift) of irradiated Chromel/Alumel thermocouples
in comparison with non-irradiated Chr/Al thermocouples

FIG.10



calibration furnace and temp. distribution

FIG.11



FILE	simplified welding circuit
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Alfred Nobel

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