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A DIRECT-GRAPHITE-RESISTANCE HEATED FLUIDISED BED : PRINCIPLES AND THERMAL PERFORMANCE OF A PILOT PLANT

by

H. J. FLAMM

1971

Joint Nuclear Research Center
Ispra Establishment - Italy

Technology

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Some cursory coating experiments showed no obvious effects on the electrical performance of the single phase system.

In extrapolation of the experimental evidence a three-phase coaxial heater system, using the upper graphite-to-graphite contact as "star-point", is thought feasible.

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KEYWORDS

HIGH TEMPERATURE
HEATERS
ELECTRIC CONDUCTIVITY
GRAPHITE
FLUIDIZED BED
COATING
PERFORMANCE
EFFICIENCY

Contents

Manuscript received on March 15, 1971

1. Introduction

In spite of extensive development programmes in HTR fuel fabrication technology 1)-4) the fluidised bed coating step still presents severe limitations with respect to scale of operation 5)-7), and, hence, economical processing conditions 8)-11). Apart from empirical relationships, equipment and process design data are sparse. Even model studies 13) and parametric surveys 14) have, so far, shown limited applicability in scale-up, which is not surprising, considering the complexity of process and product 15)-19).

Further, radial and axial temperature gradients become more pronounced with increasing column diameter and fluid throughput in coaters of conventional design, i.e. cylindrical columns with conical base and water-cooled gas injection system. Such systems are enveloped (spacing between heater and column wall approx. 5 mm) by single or three-phase graphite resistance heaters and operating temperatures of up to 2000°C are commonly employed. Beds of up to ca. 150 mm internal diameter are known to be in operation and a schematic presentation of the nature of interacting parameters in such beds is given in Fig. 1. Conductive and convective heat transfer to the column is negligible compared with the radiation component. Similar aspects govern induction heated systems.

Economically interesting unit sizes require column diameters in excess of 200 mm, preferably up to 300 mm, invariably enhancing the problems of radiation shielding and transient or unsteady state heat flow. The latter, although far from having been analysed for presently operated systems, is likely to affect on scale-up the temperature-time history of the gas inlet region drastically (present day coating techniques employ several temperature step-changes of relatively short duration, accompanied by fluid composition changes). Further, it must be noted that the particle bed height will virtually remain unchanged in scale-up. Hence, the area for radiative heat transfer to the column remains a linear function of the bed diameter. In addition the gas distributor area will have to follow proportionally the cross section and, apart from increasing the heat sink in this region, heat will only flow by conduction through the structural parts of the column or be drawn from the bed of particles itself.

For these reasons it is of paramount importance for scale-up of high temperature fluidised bed coaters to concentrate heat generation in the zone wetted by the fluidised bed and, because of the large quantities of gas to be heated to operating temperature, also in the vicinity of the gas distributor.

2. Principles of a direct-resistance heated fluidised bed system.

For the purpose of general illustration of the high-temperature fluidised bed coating process, as applied to nuclear fuel particle coating, a block diagram is given in Fig. 2, presenting schematically for the high-temperature part a graphite resistance heated system of conventional design, i.e. the reaction chamber is concentrically displaced inside a separate cylindrical graphite resistance heater.

Fig. 3 gives in more detail the principles of a direct-resistance heated single-phase system without examining mechanical design details or operation aids. Except for the heater arrangement all furnace parts function in the conventional way $^{5)-7)}$, as can be deduced from the legend in Fig. 3. Essentially the heater consists of two easily separable components, i.e. an outer cylindrical section permanently attached to a water-cooled power connector block and a coaxial inner section containing the reaction chamber mounted on a second water-cooled connector block in a manner which simplifies removal and maintenance. At the reversal point (top) both sections are in intimate contact for electrical power transmission. Design must warrant this contact, irrespective of differential thermal expansion in the two sections or machining tolerances. One way of achieving this goal is to slightly taper the contact area and to provide elasticity by appropriate slotting of the male taper. Although there are other possible solutions to this problem, experimental work described below has shown that such graphite-to-graphite elastic compression contacts are feasible electrical conductors at high temperatures and for high currents.

Physical and electrical properties of graphite limit heater design with respect to minimum wall thickness and effective electrical resistance. The latter is frequently achieved by tortuous slotting, thus leading to fragile geometries susceptible to thermal distortion and enhanced degradation by corrosion. In the system under discussion the heater length is virtually doubled by design, providing the desired increase in electrical resistance, but retaining the full mechanical strength and stability of a cylindrical body.

Furthermore, the fluidised bed coaters mentioned above are approaching a bed height to diameter ratio of one, where heat losses over the end faces are no longer negligible. Compact design and heat generation at or close to the point of consumption become a command and it is obvious that in the directly heated system heat generation per unit bed height is about twice that in a conventionally heated system. In addition the double wall array forms a graded thermal barrier with reduced radial heat losses from the inner section, which can be further underlined by disproportionating of heat generation between inner and outer section.

Depending upon the optimisation of gas injection systems for large-scale coaters 20) reaction chamber geometries may be devised, where a large part of the heat is generated within or near the zone of gas injection, i.e. the point of highest heat consumption (~10 - 20% of total heat generated).

Three-phase operation may be desirable for more uniform grid loading. This can be achieved by simply employing a third concentric heater tube in the manner described and using the reversal point (top) as "star-point" in the conventional way.

There is no generally successful method for estimating heat flow in critical regions of high temperature fluidised bed coaters and its effect on product quality or in the first instance on local process parameters. Therefore, following only practical reasoning, power supplies and (water) cooling rates are balanced on the merits of minimising cooling times, i.e. down-times or waiting-times. Preliminary heat balance, and heat transfer calculations for a scaled-up system make this practise appear extravagant and indicate not only the need for better thermal shielding all around, but even increased heat generation in the gas inlet region, in order to check unsteady state conditions. The system under discussion follows these principles and it seems necessary for any scaled-up system to provide secondary means for rapid cooling.

3. Experimental

3.1 Design limitations

A single-phase high temperature coating furnace, as developed by the OECD High Temperature Reactor Project DRAGON, and manufactured by WDM Limited, Bristol, U.K. (for basic features see Fig. 4) was made available by the CCR Materials Division for the experimental work described below. In addition a 55 mm I.D. coating column with heater of conventional design (Fig. 5) aided comparative experimental work.

Design of the direct-resistance heated experimental system had to adhere to the basic features and dimensions of the above furnace including those of the lower power connector block. This allowed for a maximum diameter of the outer heater of 110 mm and a graphite felt insulation to the water cooled furnace casing of 28 mm. Inner diameter of the outer heater, and outer and inner diameter of the reaction chamber were 95,90 and 82 mm, respectively. The upper end of the outer heater was internally tapered to an included angle of 10^{o} and fitted with three equi-spaced slots 60 mm in length. The matching male taper of the inner heater thus provided an effective contact area of the order of 60 to 70 cm^{2}.

In order to retain experimental flexibility for further work on gas injection systems and to obtain further valuable information on flexible graphite-to-graphite compression contacts at operating temperature, the reaction chamber was screwed onto a graphite block with a tapered section for electrical contact. Flexibility was again provided by six equi-spaced slots in the tapered and threaded section of the graphite block. General details of this type of connection and the reaction chamber geometry may be taken from Fig. 6. The graphite block was fitted with a central 5 mm diameter gas passage and mounted on a special water-cooled power connector block.

Design limitations were encountered, in so far as the furnace length to diameter ratio could not be optimised, i.e. the reaction chamber had to be lined up with the existing viewing ports for temperature measurement, thus rendering the column length excessive. Furthermore, both the positions for water-cooling and the cooling rate were fixed.

By design approximately 60% of the heat was generated in the inner section, and 40% in the outer section.

3.2 High Temperature Performance Tests

Work so far was primarily concerned with testing various novel features of the system and to establish confidence criteria for future design, particularly with respect to:

a. electrical power transmission through graphite-to-graphite compression joints.

b. the effect of temperature gradients and, hence, differential thermal expansion on the mechanical stability of the compression joints.

c. arcing in a coaxial system and the compression joints

d. pin-pointing unexpected technical problems in mounting and removal of the reaction chamber
e. temperature distribution and power consumption
f. the effect of a bed of particles and coating deposits on the overall heater resistance.

Under the given circumstances the experimental programme followed basically go-no-go procedures in the order of the above tabulation. Thus, in the first approach to temperature, the system was heated under argon to approximately 1200°C, checked for signs of arcing, hot spots, electrical power stability, etc., then dismantled after cooling and rechecked for signs of wear, distortion or mechanical damage. As there were no visible defects the cycle was repeated, but the temperature increased to approx. 2200°C. This temperature was then held for about two hours. Even this test revealed no signs of deterioration.

Only two points were noted for future design, i.e., allowance should be made for in-situ concentric alignment of both heater legs, and the connector block for the inner section should have a positive stop in both axial directions when in contact position for the compression joints. Before start-up the furnace is evacuated and slight inward movement of the connector block may lead to destruction of the upper graphite-to-graphite compression joints. This feature has to be stressed, since in order to absorb machining tolerances, fine axial adjustment of the inner section is needed on assembly.

Items e. and f. are treated separately in the following chapters.

3.3 Comparison with a conventional system

The principle features of the high-temperature section for both systems may be taken from Fig. 5 (conventionally heated bed) and Fig. 6 (direct-resistance heated bed). Graphite felt insulation was adapted to the latter system. Temperatures were determined by micro-optical pyrometry on the outer wall of the heater, and through 3 mm diameter holes in the latter wall on the outside wall of the reaction chamber (or inner heater for the direct-resistance heated bed). Sight ports were provided at 4 cm spacing in the vessel wall and the position of the reaction chambers with respect to these viewing facilities can be seen in Figs. 5 and 6. (Pos. 1 to 7).

In order to compare temperature profiles data are plotted in Fig. 7 against the point of gas injection as base line or reference level. For each system three representative sets of data are given over the temperature range of interest (all data refer to beds without particles and static argon atmosphere at 1.1 ata.). As expected for the conventional system the heater temperature is approximately equal to or higher than that of the reaction chamber. The temperature gradient towards the gas inlet is remarkable, increasing to several hundred degrees for high temperature operation. This feature, negative in every respect, and in particular to process control, does not, however, come within the scope of this evaluation.

The direct-resistance heated bed clearly indicates the "shielding" effect of the outer leg of the heater, increasing with temperature. Although axial temperature gradients are present, they are less pronounced than in the former system. It is thought that by further adjustments in local heater resistance (i.e. local effective heater cross-section) a virtually flat temperature profile can be achieved. This is, however, a topic to be closely investigated in connection with improved high-temperature gas injection systems [21].

At this stage of the investigation the striking reduction in power consumption for the direct-heated system is of paramount interest. Although the effective bed cross-section was more than twice that of the conventional bed (52.8 and 23.6 cm^2, respectively), the power required to heat the reaction chamber to 'equivalent' temperatures amounts only to some 50 - 60% of that required for the conventional bed (Fig. 8).

Precise formulation of all parameters relevant to power consumption and process application, such as influence of bed size and geometry, effect of local cooling on temperature gradients, differentiation of all circuit resistances, etc., is outside the scope of this presentation and would require exceptional experimental and analitical efforts. For the purpose of scale-up to beds with 200 to 300 mm I.D. reaction chambers the advantages are clearly demonstrated and further work in depth must be process-orientated.

A further process-orientated feature is the rate of temperature rise from equilibrium condition, a factor finally decisive for the lay-out of the power supply, As only manual power control and temperature recording was available, a set of manageable conditions was chosen for orientation experiments in two-temperature ranges (Fig. 9). Both systems were heated to equilibrium for a given power input and at time zero the power was increased by approximately 50% (volt-ampere reading). As the graphite resistance changes with temperature, small power adjustments had to be made in the course of the experiments. Significant differences in the rate of temperature rise are not observed, particularly as the method used for comparison of the two systems is overshadowed by the steep temperature gradients in the conventional system (see Fig. 7). The comparatively lower rate-of-temperature rise in the direct-resistance heated system is attributed to the larger mass of graphite structure to be heated , and, hence, it can be concluded that the response of the latter system to relative power changes is of the same order of magnitude or better than that of the conventional system.

Finally the rate of cooling is an important process parameter in fluidised bed coating, as waiting times have a strong effect on the process economy. Evidently the improved "shielding" in the direct-resistance heated bed has an adverse effect on the rate of cooling as shown in Fig. 10. Conclusions on this point will be drawn in Chapter 5.

3.4 Effect of particle loading and coating

Only some cursory experiments were made using improvised gear for gas flow control and 595-707 µm sieve fraction fused zirconium oxide particles for fluidisation and coating experiments. There was no evidence of changes in heater characteristics with bed filling and degree of fluidisation using argon over the range of interesting temperatures. Similarly exploratory coating experiments with methane-argon mixtures revealed no adverse effects on the heater characteristics due to impregnation of the heater by pyrolysis products. The latter effect, however, can only be studied in long time experiments and from experience this will greatly depend on appropriate gas injection system design. It is in any case current practice to replace the reaction chamber at regular intervals, so that the evidence provided so far suffices to encourage further experimental work.

4. Summary

A direct resistance heated high-temperature fluidised bed coater is a feasible solution for scale-up in coating of nuclear fuel particles, because of its thermal economy in the coating zone. Some specific advantages or features are:

a. It allows an extremely compact design of improved rigidity.

b. The length of the heater is approximately doubled, thus providing a desirable increase in electrical resistance, but retaining the full mechanical strength of a cylindrical body.

c. As the outer heater forms a graded thermal barrier with reduced radial heat losses from the internal section, power consumption can be reduced by an estimated 50% over the conventional design, thus reducing plant investment cost, cooling water consumption, problems associated with high current transmission, etc.

d. The heater cross-section can be adjusted locally within the limits of mechanical strength and performance requirements, thus allowing disproportionation of heat generation according to local demand. (It must be recognized that on scale-up, heating of the gas to working temperature can no longer be ignored. The peak heat flux will be in the gas inlet region).

e. It has been shown that elastic graphite-to-graphite compression contacts are feasible electrical conductors at high temperatures and for high currents.

f. Arcing or electrical interactions between the coaxial heater sections have not been observed.

g. Finally, a three-phase concentric system can be devised, using the upper graphite-to-graphite contact as "star-point".

5. Recommendations for further work

It may be difficult to find a base line for comparison of the different systems. Data on power consumption in conventional systems are sparse and, where available, more related to the overall furnace geometry and thermal shielding than to reaction chamber diameter. From experience some values are given in Fig. 11 for conventionally heated systems of 55, 76 and 127 mm I.D. and hypothetically extrapolated to larger beds. The direct resistance heated bed has been set in perspective making allowance for actual coating conditions. It will be necessary to verify these assumptions by experiment, in order to arrive at the correct rating for power supplies with respect to minimum permissible wall thicknesses of the reaction chamber. A reaction chamber wall thickness of as little as 5 mm may be required for a 300 mm I.D. bed (using a nuclear grade graphite) and mechanical performance tests will be necessary. Carbon or graphite yarn/resin composites in shaped form may well offer development potential in this area.

Fast cooling on temperature step-reduction (e.g. from ~2000°C to ~1600°C for application of silicon carbide coatings) and for bed unloading is another area in need of experimental and engineering attention. The problem arises on scale-up of either system, but is far more pronounced in the direct-resistance heated bed. Variable cooling of the gas injection area could be one effective solution. A retractable "cold-finger" arrangement with incorporated suction type "hot-unloading" would serve a dual purpose by submersion into the bed of particles from the top of the furnace.

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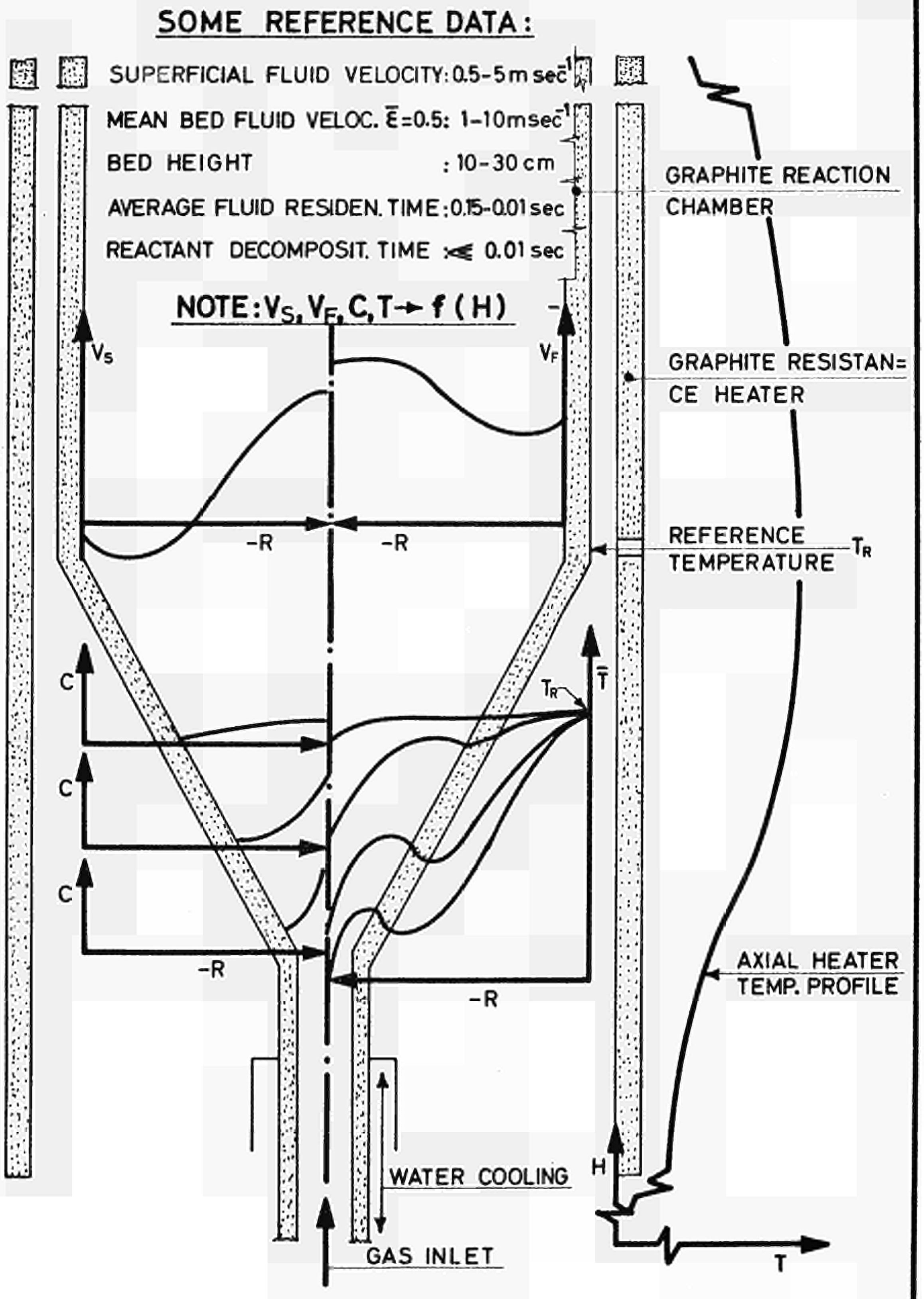
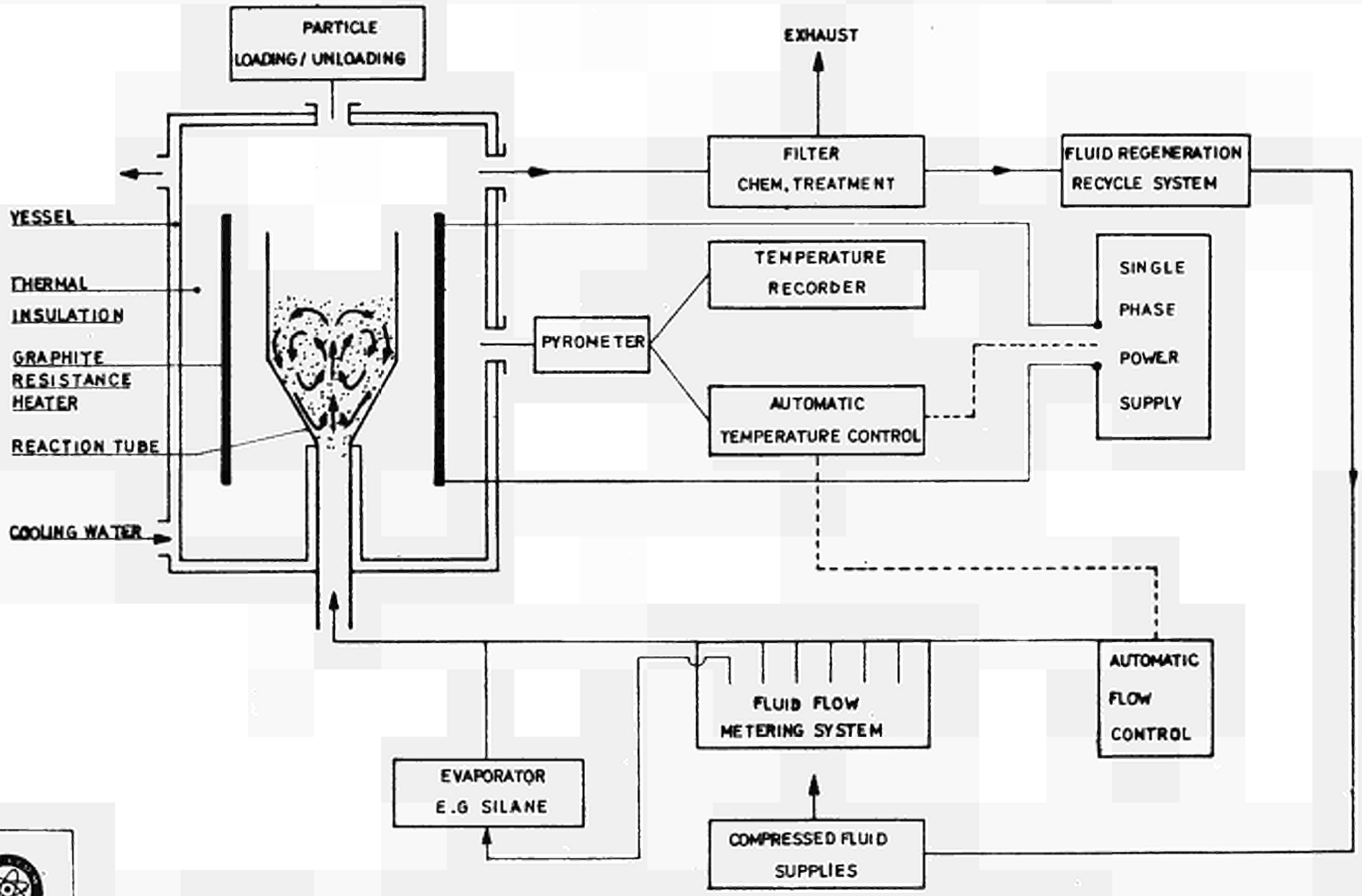


C.C.R. - ISPRA

SCHEMATIC PRESENTATION OF TEMPERATURE (T), FLUID VELOCITY (V_F), SOLIDS VELOCITY (V_S) AND COATING AGENT CONCENTRATION (C) AT RADIAL POS. (R) AND AXIAL POS. (H) IN A

HIGH TEMPERATURE FLUIDISED BED COATER

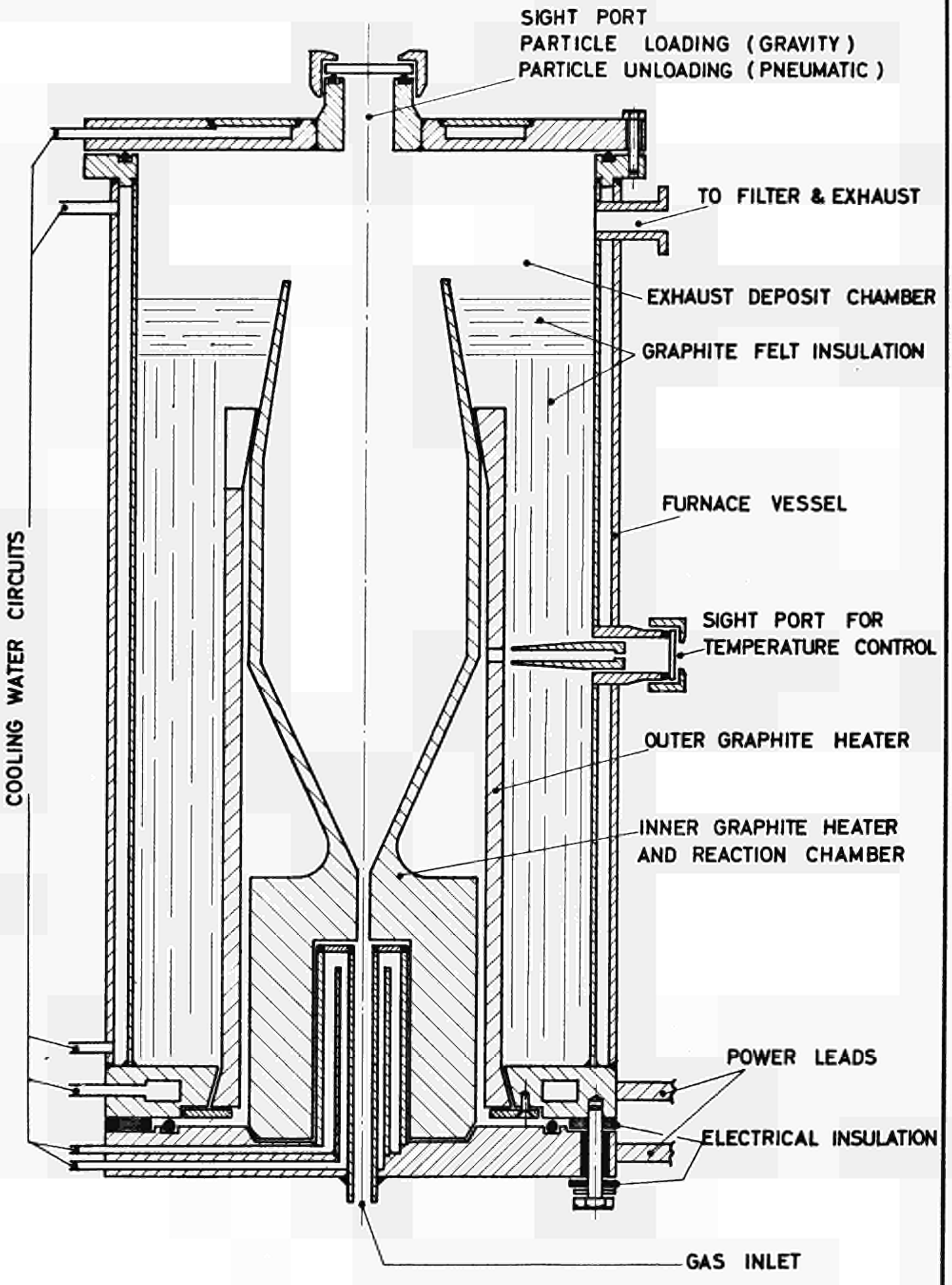
FIG. 1



C.C.R.-ISPRA

BLOCKDIAGRAMME OF A HIGH TEMPERATURE FLUIDISED BED COATER FOR NUCLEAR FUEL PARTICLES

FIG.2



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LAYOUT OF DIRECTLY HEATED FLUIDISED BED FURNACE (SINGLE PHASE)

FIG.3

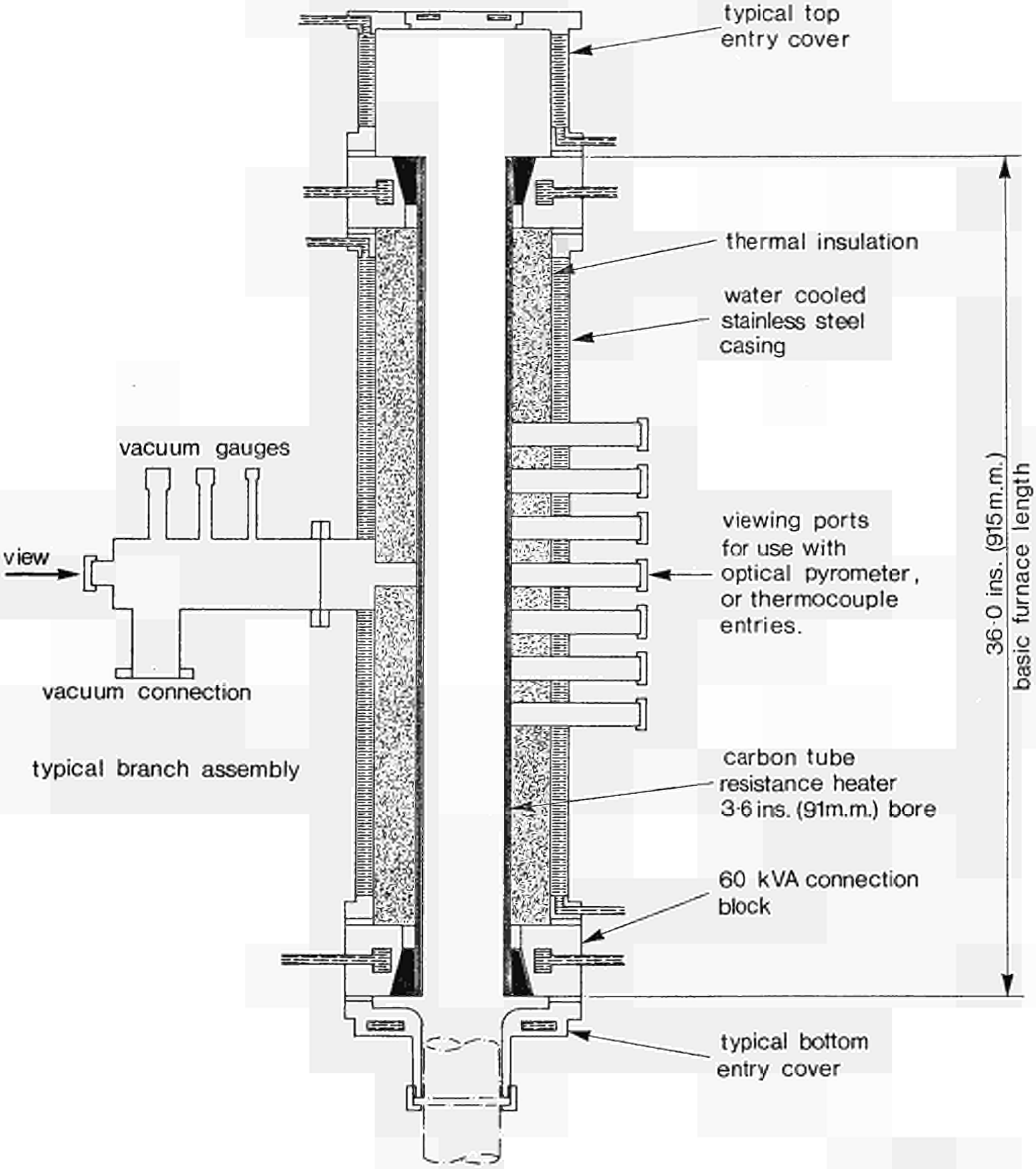
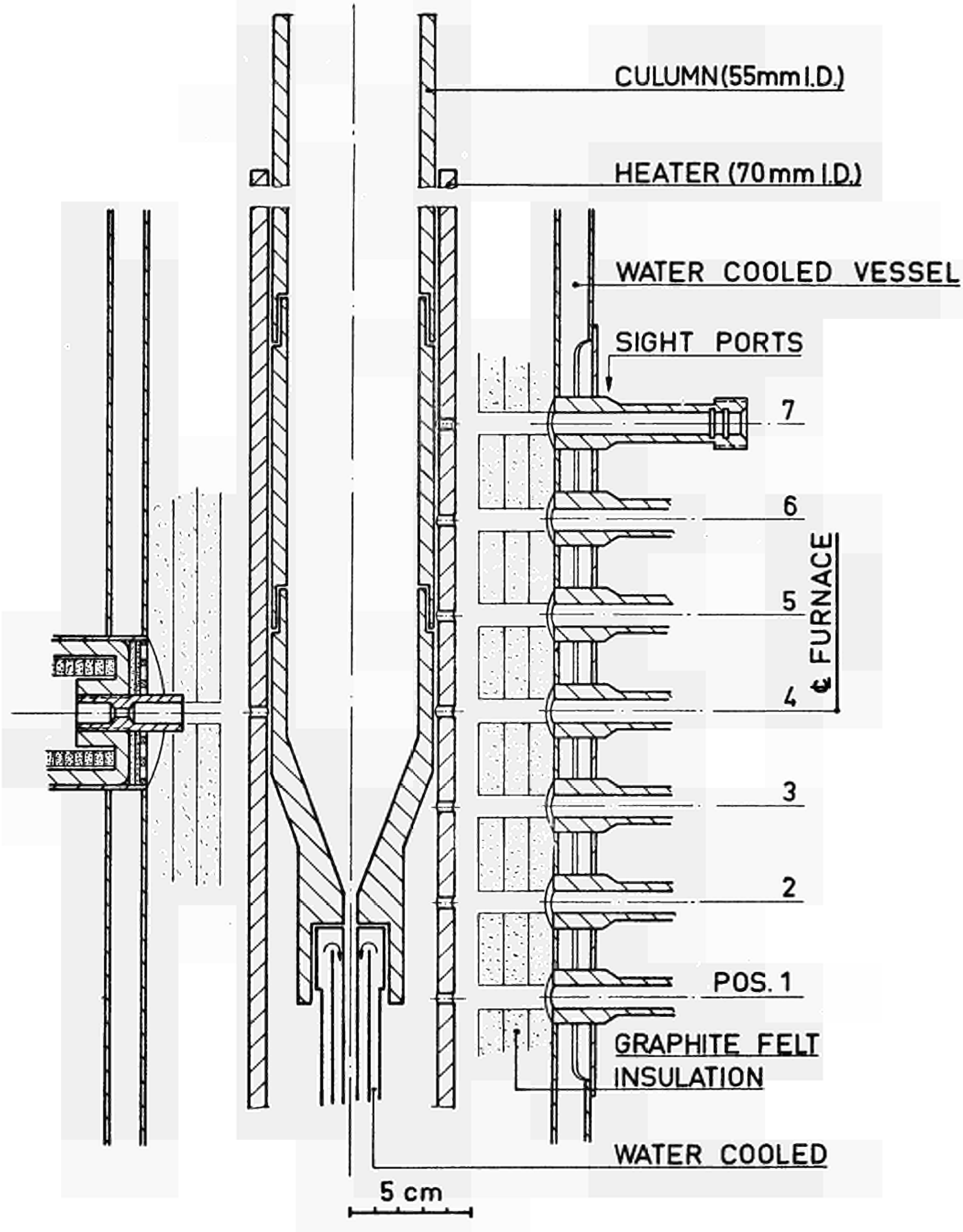


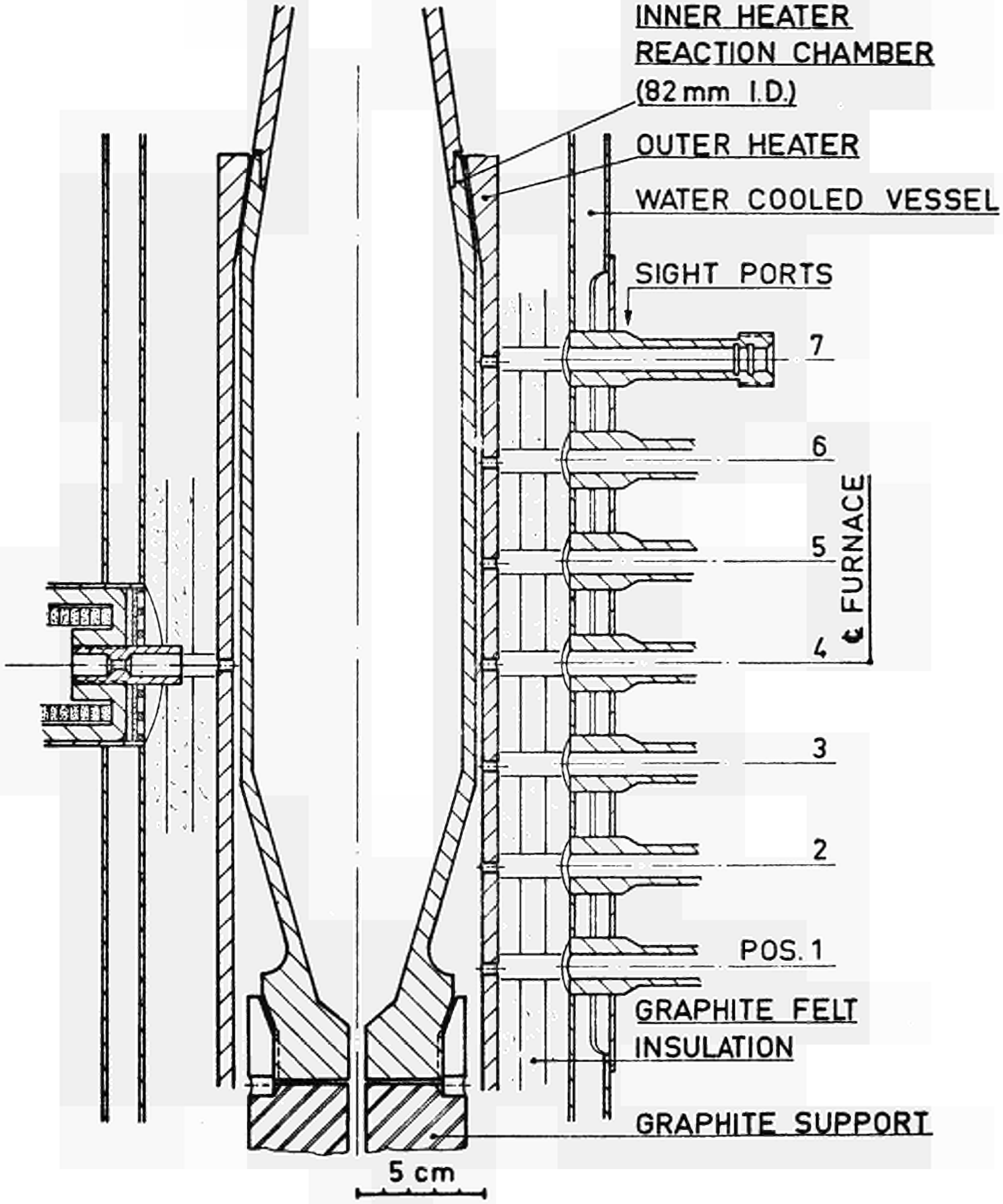
FIG. 4

BASIC FURNACE ASSEMBLY



GEOMETRY OF CONVENTIONALLY HEATED BED

FIG. 5



GEOMETRY OF DIRECT-RESISTANCE HEATED BED

FIG. 6

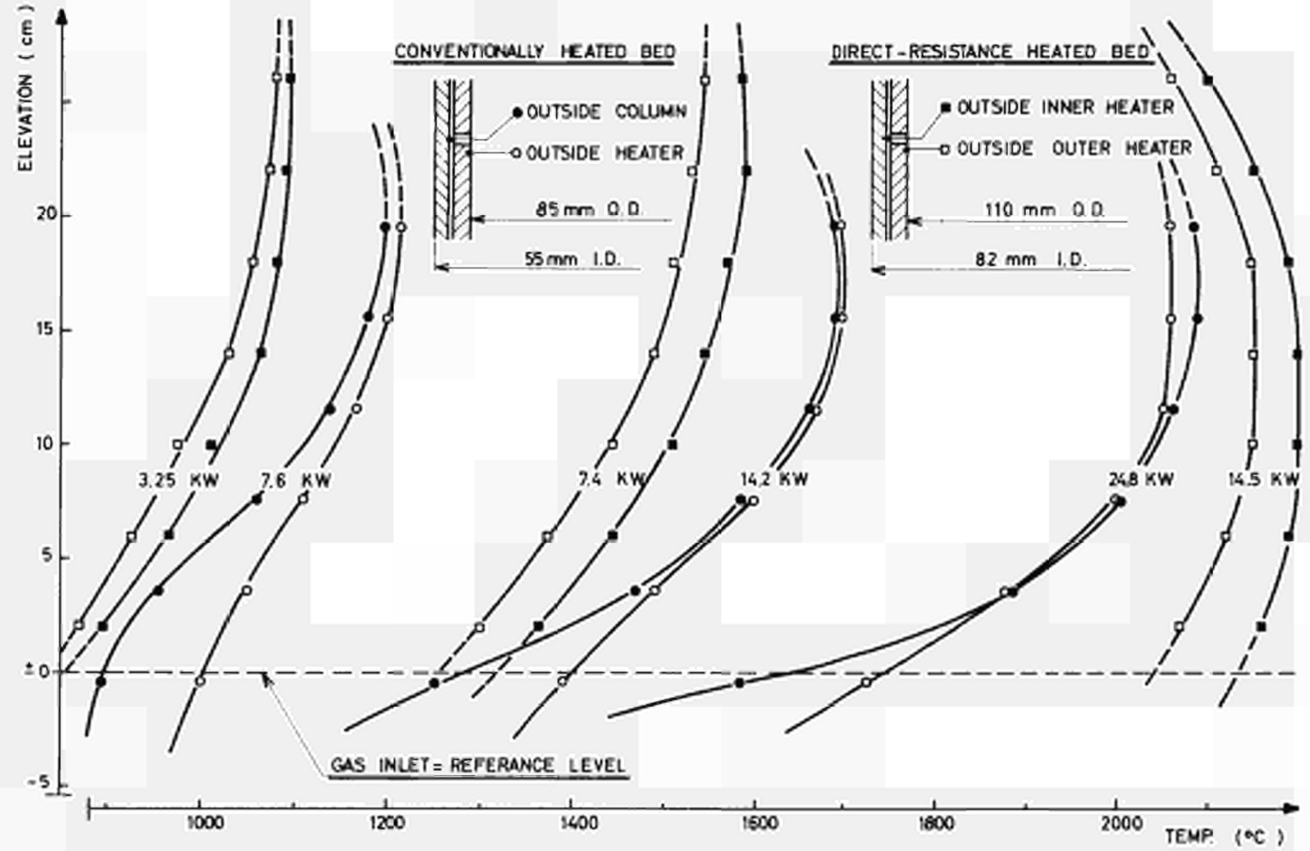
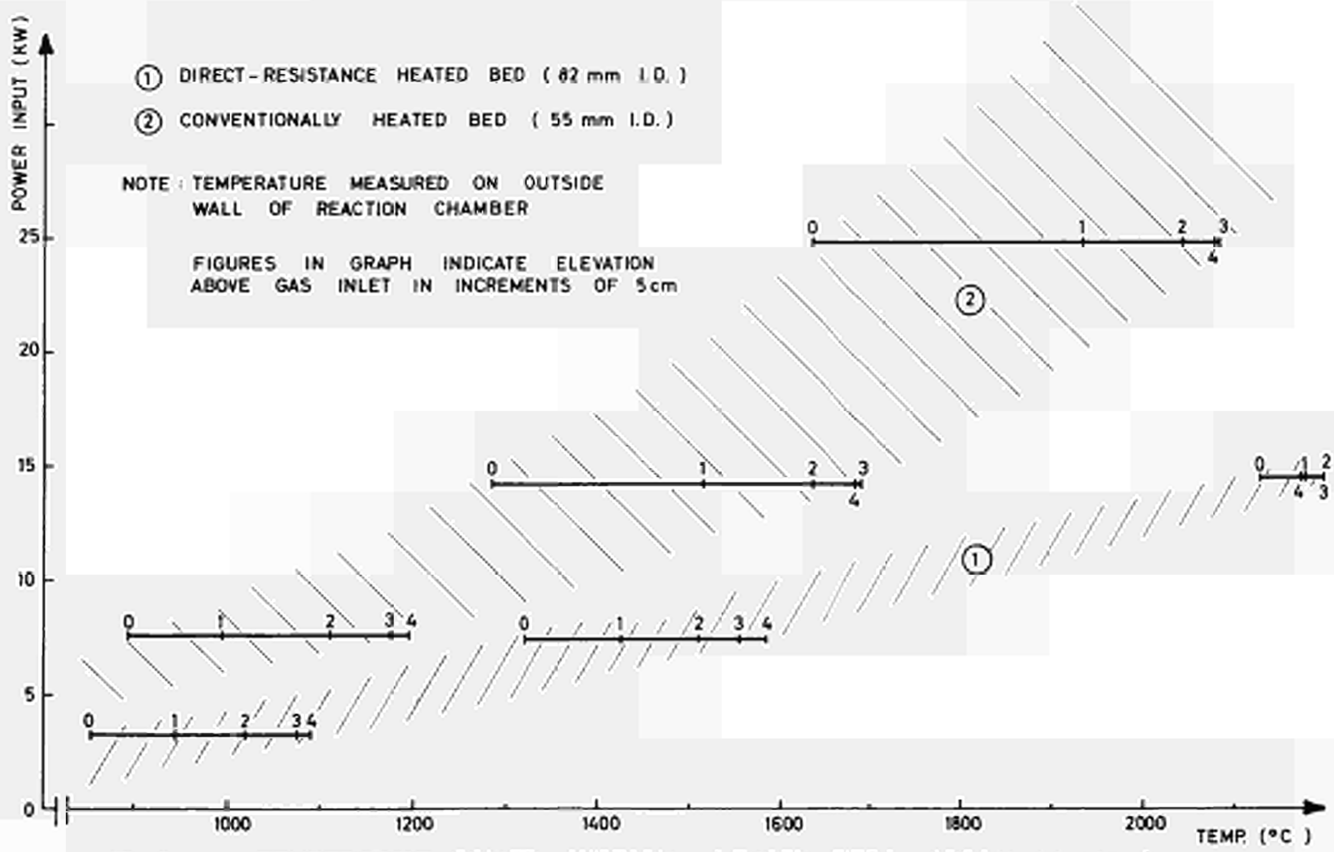


Fig. 7 – TEMPERATURE PROFILES (EMPTY BEDS - ARGON 1.1 ata)



NOTE : TEMPERATURE MEASURED ON OUTSIDE WALL OF REACTION CHAMBER

FIGURES IN GRAPH INDICATE ELEVATION ABOVE GAS INLET IN INCREMENTS OF 5 cm

Fig. 8 – TEMPERATURE - POWER HISTORY (EMPTY BEDS - ARGON 1.1 ata)

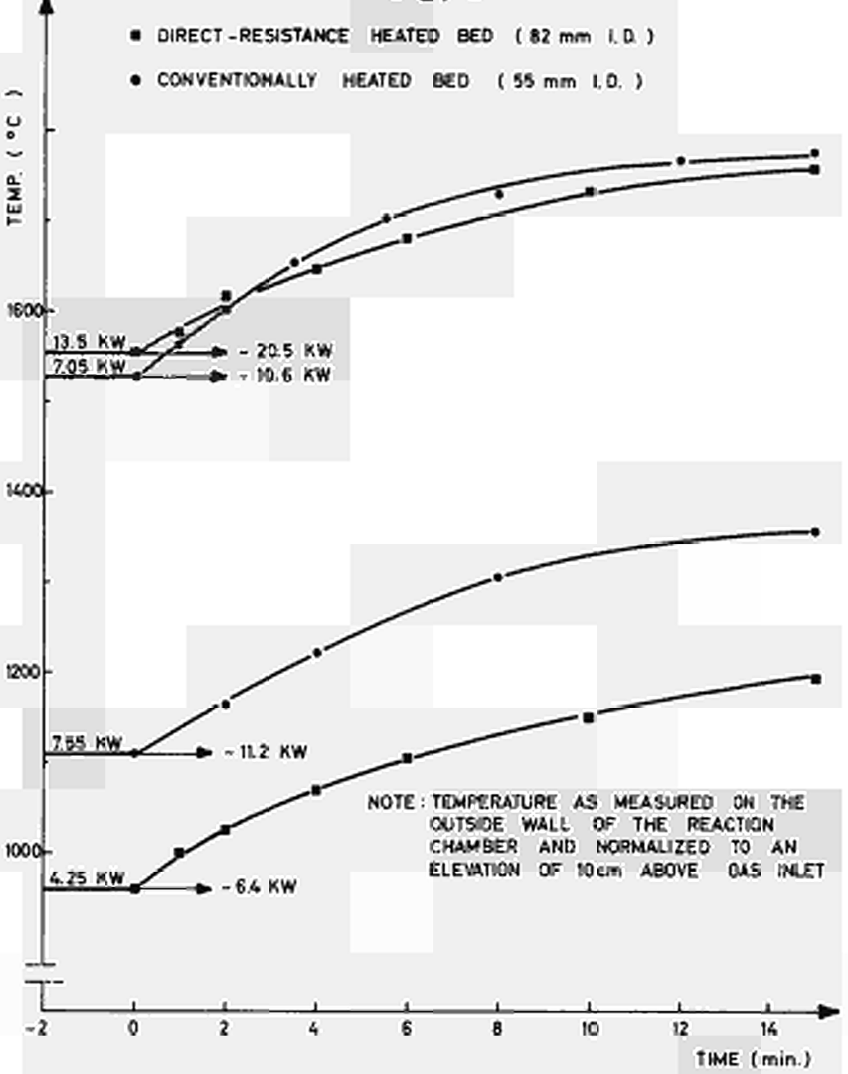


Fig. 9 - TEMPERATURE INCREASE FOLLOWING ~ 50 % POWER INCREASE FROM EQUILIBRIUM (EMPTY BEDS - ARGON 1.1 ata)

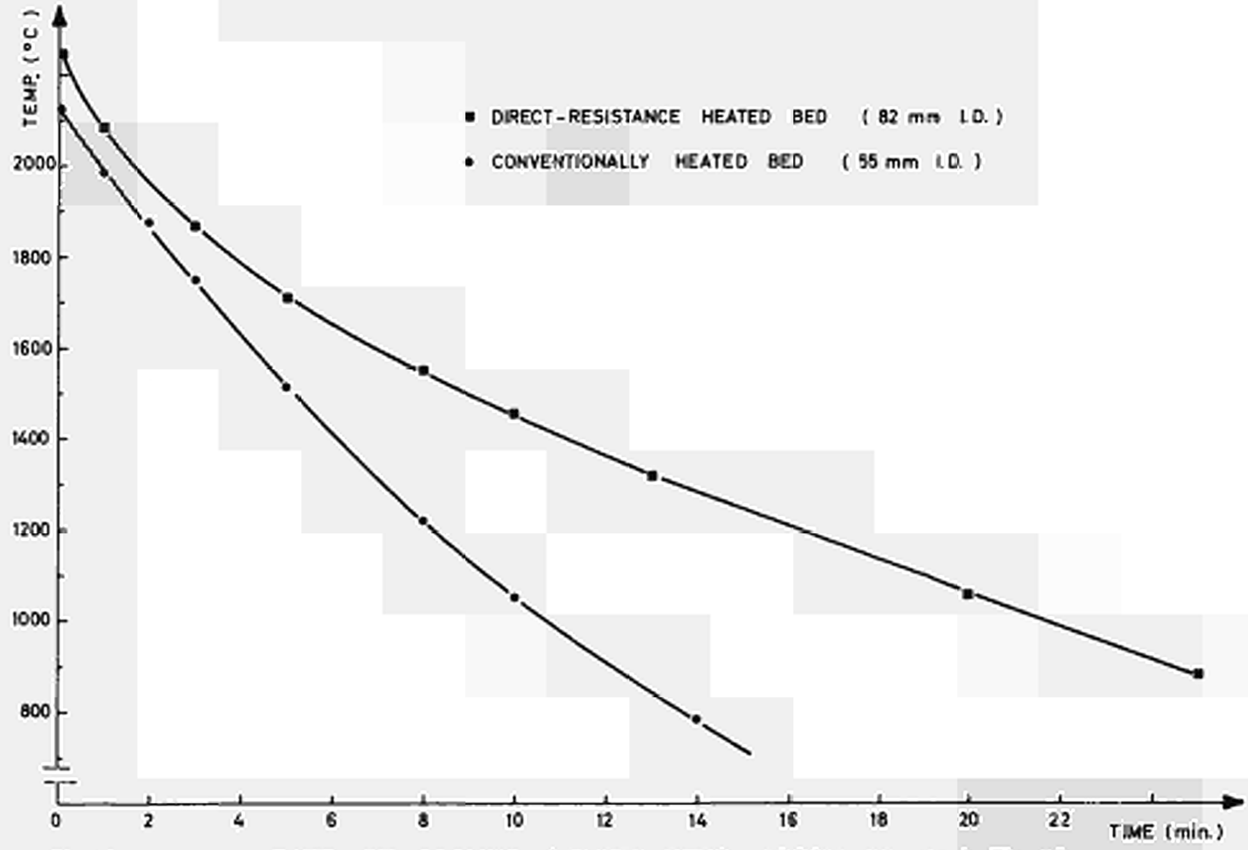


Fig. 10 — RATE OF COOLING (EMPTY BEDS - ARGON 1.1 ata) - Fig. 10

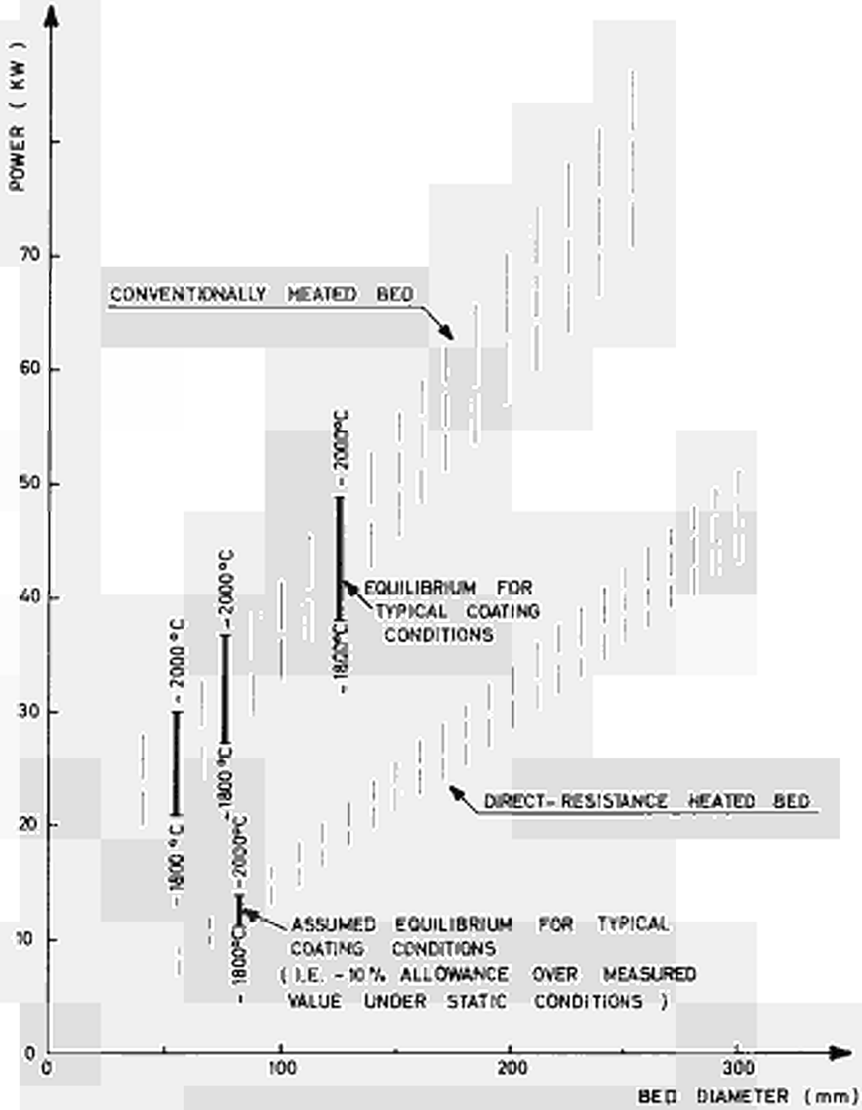


Fig. 11 ESTIMATE OF EQUILIBRIUM POWER REQUIREMENTS ON SCALE - UP

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>
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