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Institute for Safety Research and Reactor Technology

# Helium Purification in AVR and THTR

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| Title | Helium Purification in AVR and THTR |
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| Customer | EUROPEAN COMMISSION |
| | |
| Customer reference | FIKI-CT-2001-20177 |
| | |
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| | |
| File reference | FZJD44 |
| | |
| Report number | HTR -E-FZJD44 |
| Report status | Issue 1 |

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**Summary**

This report contains state of the art information on the helium purification of the German HTR's AVR test reactor and THTR-300. It is the contribution of Forschungszentrum Jülich GmbH to deliverable D44 WP6 of HTR-E (EC project: FIKI-CT-2001-00177)

## Contents

# 1 INTRODUCTION

Helium is used as the primary coolant for the HTR power plants. The helium purification system provides the necessary degree of helium purity. The tasks for the system are:

1. Reduction of corrosive impurities in the primary coolant
2. Reduction of gaseous radioactive impurities in the primary coolant
3. Support of operation procedures like:

- Helium supply and storage
- Evacuation of the primary circuit
- Fuel charging (pebble bed type)
- Storage of radioactive gaseous materials

In detail, the helium purification system has the following objectives:

- Removal of gaseous contaminants especially oxidising and there reaction products with carbon from the primary coolant to maintain design values, in particular for H2O, CO, CO2, H2, CH4
- Removal of tritium
- Removal of inert gases, e.g. nitrogen
- Removal and decay of radioactive noble gases
- Start-up purification of the primary system before initial start-up and after inspections and maintenance
- Purification of newly delivered helium

# 2 AVR

The AVR purification already contains most of the purification steps of the later reactors.

## 2.1 Description of the Flow sheet

In a gravel bed (0,8-4mm) the dust content of a by-pass of 1000m³/hr is reduced. Afterwards the by-pass flows through filters, where the remaining suspension materials are reduced. A smaller part of the pre-cleaned gas (50m³) flows to an oxidisation catalyst bed to reduce the $H_2$ content. The drying and separation of $H_2O$ and $CO_2$ follow. In the stage RA2 (see Figure 1) the gaseous fission products Krypton and Xenon are removed. The separation is executed in absorber beds (activated charcoal) located in a cold-box..

The Gas purification unit is based on the following stages:

**Dust filter**

**Oxidizing**

In a metal oxide bed corrosive gas components like H2, CO, CH4 are oxidized to H2O and CO2. In this step the tritium is oxidized too. After a certain time the oxygen content of the catalyst has been exhausted and the catalyst has to be regenerated with oxygen.

**Cooling to ambient temperature**

**Condensing of liquid gas contents**

**Cooling**

of the incoming gas with clean gas in a cross counter flow heat exchanger.

**Drying (RA2/1a and 3a)**

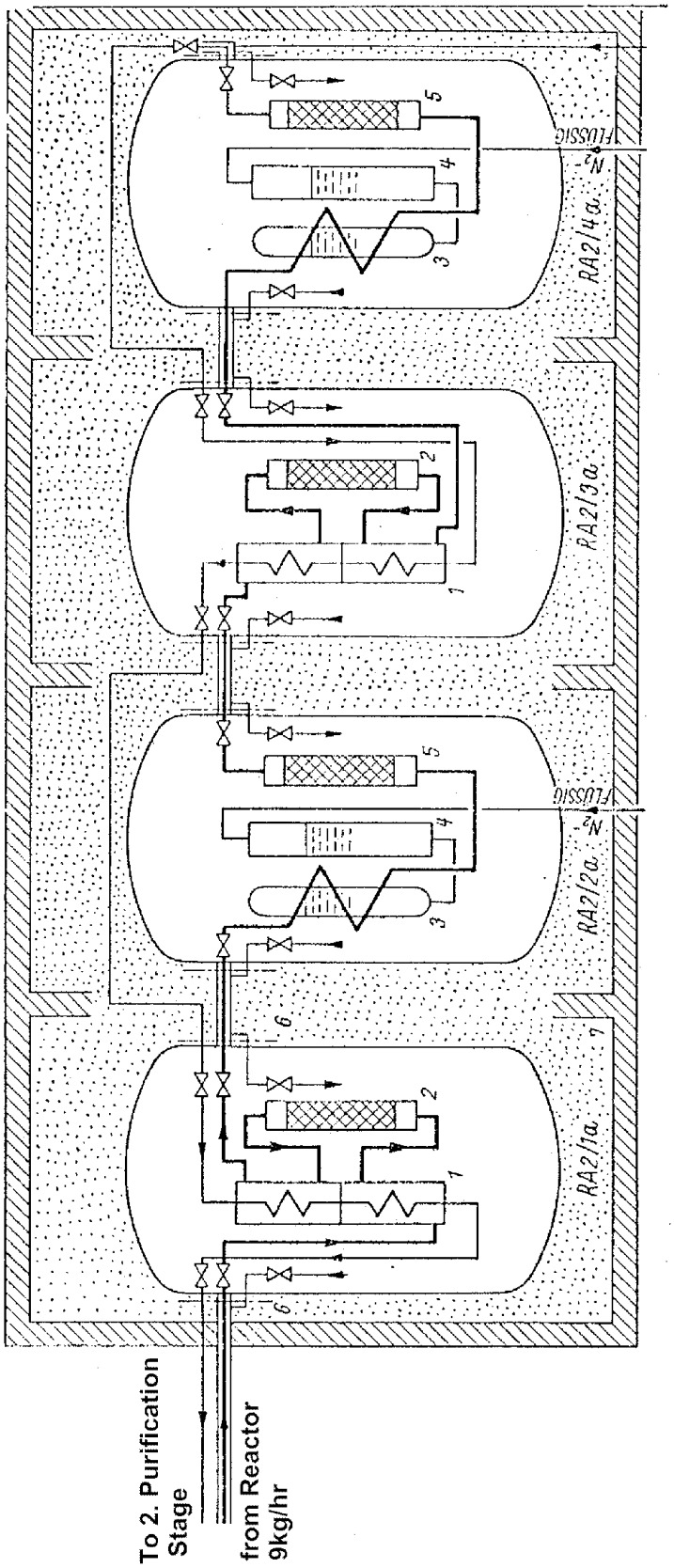
In a silica gel bed the final drying takes place. $H_2O$ and $CO_2$ are removed..

**LN cooler (RA2/2a and 4a)**

The incoming gas is precooled with purified helium and afterwards with boiling liquid nitrogen to a temperature of –175°C. The liquid nitrogen is delivered externally.

**Activated charcoal**

In a bed with activated charcoal noble gas impurities krypton and xenon (radioactive fission products) are adsorbed. The dimensions of the absorbers are chosen in a way that for the reduction of the activity enough decay time remains e.g. 2000hr for Kr and 350hr for Xe.




adsorber

Perlite)

Crosscounterflow HX

adsorber

Figure 1 Schematic flow sheet of the AVR He-purification

## 2.2 Design Philosophy

The AVR purification already contains most of the purification steps of the later reactors. But the specific guide lines, expressed in the safety report, are total other. Two examples:

- The reason for the dust separation was the abrasion of graphite surfaces with the dust (sandblast)
- The $CO_2$ removal to overcome the Carbon-transport via Boudouard and reverse Boudouard reaction. This was a well known effect of the Magnox-reactors, but there because of the $CO_2$ cooling.

## 2.3 Activity of the primary coolant

Continuous measurement of noble gas activity were conducted in the AVR. Particle failures in test fuel elements could be detected by a considerable increase of the activity (1975 - 1978). Also the raising of the average outlet temperature from 850°C to 950°C caused a prompt increase (by 25%) of the noble gas activity. Elevated values were also found after the water ingress accident 1978. Since 1983 the values of fission gas were low. This was caused due to the increased fraction of "clean" LEU fuel, where the outer contamination fraction was estimated to $10^{-5}$.

A typical distribution of specific activities in the hot coolant as measured in the period 1984 to 1987 at full power and 950°C gas outlet temperature is:

| | |
|---|---|
| fission noble gases | $4.6*10^{8}$ Bq/m$^3$ |
| Tritium | $3.7*10^{7}$ Bq/m$^3$ |
| $^{14}$C | $1.9*10^{7}$ Bq/m$^3$ |
| $^{60}$Co | $1.0*10^{1}$ Bq/m$^3$ |
| $^{90}$Sr | $2.0*10^{2}$ Bq/m$^3$ |
| $^{110}$Ag | $4.9*10^{1}$ Bq/m$^3$ |
| $^{131}$J | $5.2*10^{2}$ Bq/m$^3$ |
| $^{137}$Cs | $3.0*10^{2}$ Bq/m$^3$ |

## 2.4 Experience

In the last years of operation there was no radiological necessity to operate the low temperature part of the gas purification because of the improvement of fuel elements. The only purpose was to have enough delay time before transport to the clean gas storage.

The nitrogen concentration in the helium was also no serious problem with delivered fresh helium. Nevertheless this has to be observed carefully because of $^{14}C$.

The tritium production from $^{3}He$ could be neglected compared to that from the lithium in the carbon stone, which is a permanent source because of the large amounts of carbon stone inside the ceramic structures of the AVR.

A separator for larger amounts of water (partial condenser) was missed after the water ingress accident in 1978. After that event a possibility for faster drying was missed too.

# 3 REMARKS ON DUST IN AVR EXPERIMENTS

## 3.1 Primary Circuit Data

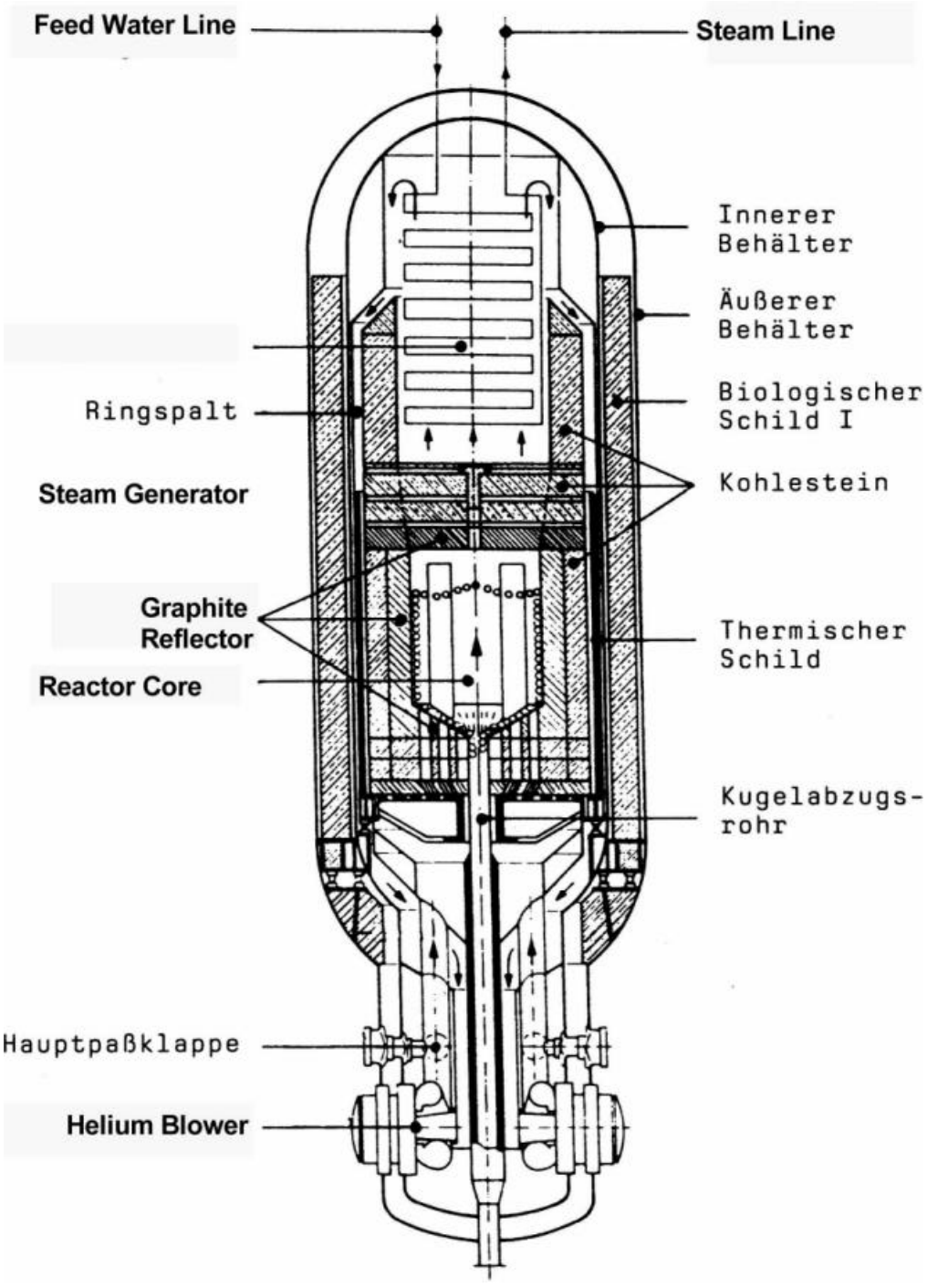


Figure 2 Vertical Section of the AVR

In Figure 2 the vertical section of the AVR is shown. The flow of the helium through the core is upwards in contrary to the today's discussed HTR's. The main data are given in Table 1

| | |
|---|---|
| Thermal Power | 46 MW |
| Electrical Power | 15 MW |
| Power Density | 2.6 MW/m$^3$ |
| Helium Inlet Temperature | 270 °C |
| Helium Outlet Temperature | 950 °C |
| Helium Pressure | 10.8 bar |
| Helium Flow | 12.6 kg / s |

Table 1 Main Data of the AVR

## 3.2 Experiments at AVR

The AVR was not only used, to show the operation of a Pebble Bed Reactor, but also for a lot of experiments. Therefor a couple of fuel element types were tested. This was not only in the direction of improving the coated particles but also in direction of the outer graphite layer.

Later work in the direction of inherent safe reactors was done, that means experiments for beyond design accidents. After the successful experiment with the blocking of all 4 control rods and the stopping of the blower as an additional experiment was planned with the conditions but with pressure relief to normal pressure. In preparing and licensing of this experiment, some work was done to estimate the possible activity release. In the frame of this work the activity of the dust was discussed.

The activities were measured in the different experiments (Vampyr I, Cold Gas Filter and Dust Experiment) shown in Figure 3.

A calculation (not experiment) estimated a dust inventory of 60 kg after 20 years operation or a dust production of 3 kg / year. It was assumed, that 50% of that amount is deposited in the fuel handling system and some other absolute sinks, while the other 30 kg are deposited evenly on the steam generator and the cold gas parts.

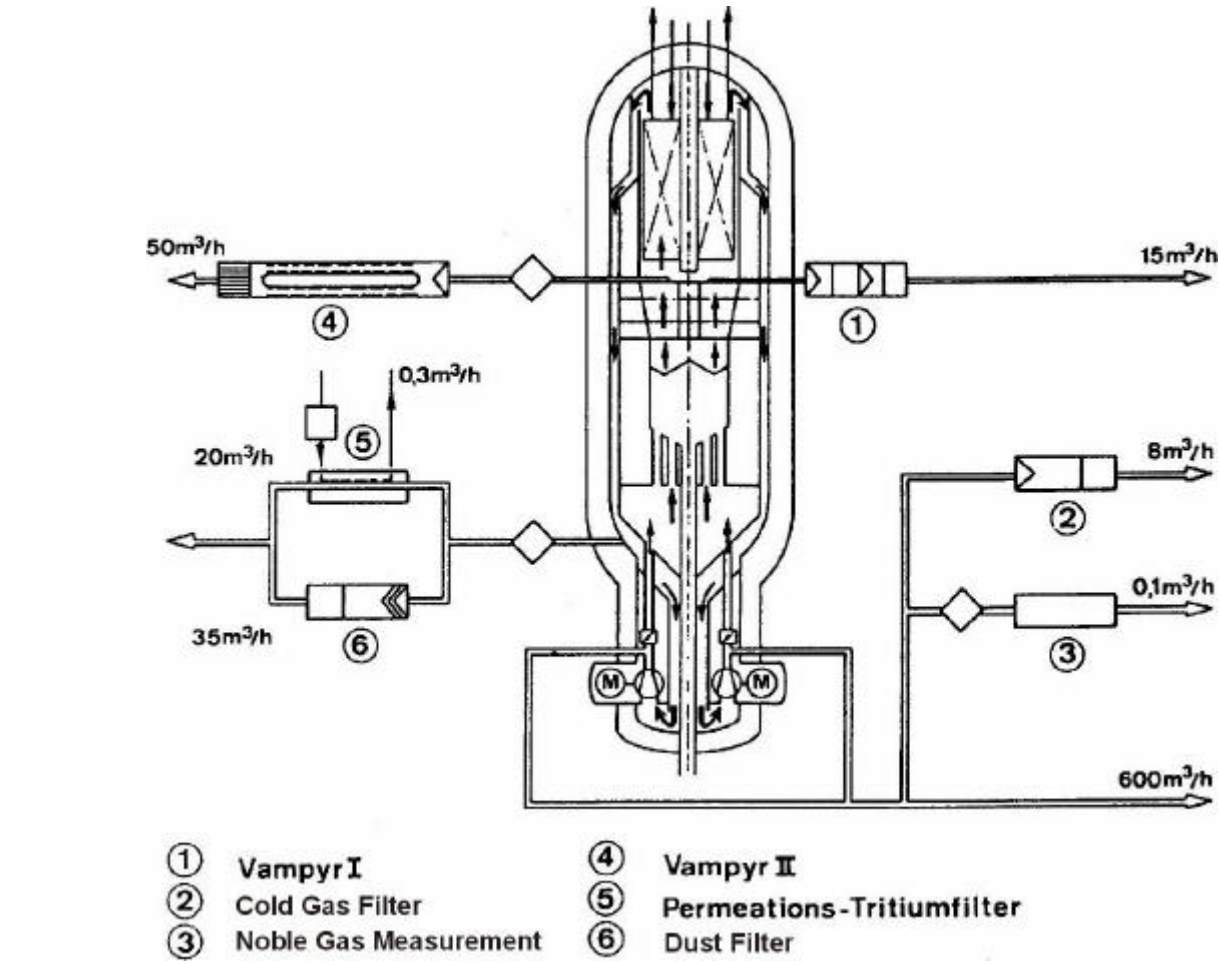



Figure 3. Experiments in the primary circuit of the AVR

## 3.3 Experimental Results

In Figure 4 the dust concentration in the gas is shown. A concentration of 5 ìg / $m^3$ means 8 mg dust in the primary circuit carried by the helium. It has to be mentioned that the concentration is sensitive against operational changes caused by the fuel handling system or vibration of the steam generator (see T1 and T2). But also then less than 0.01% of the expected deposited dust is remobilize.

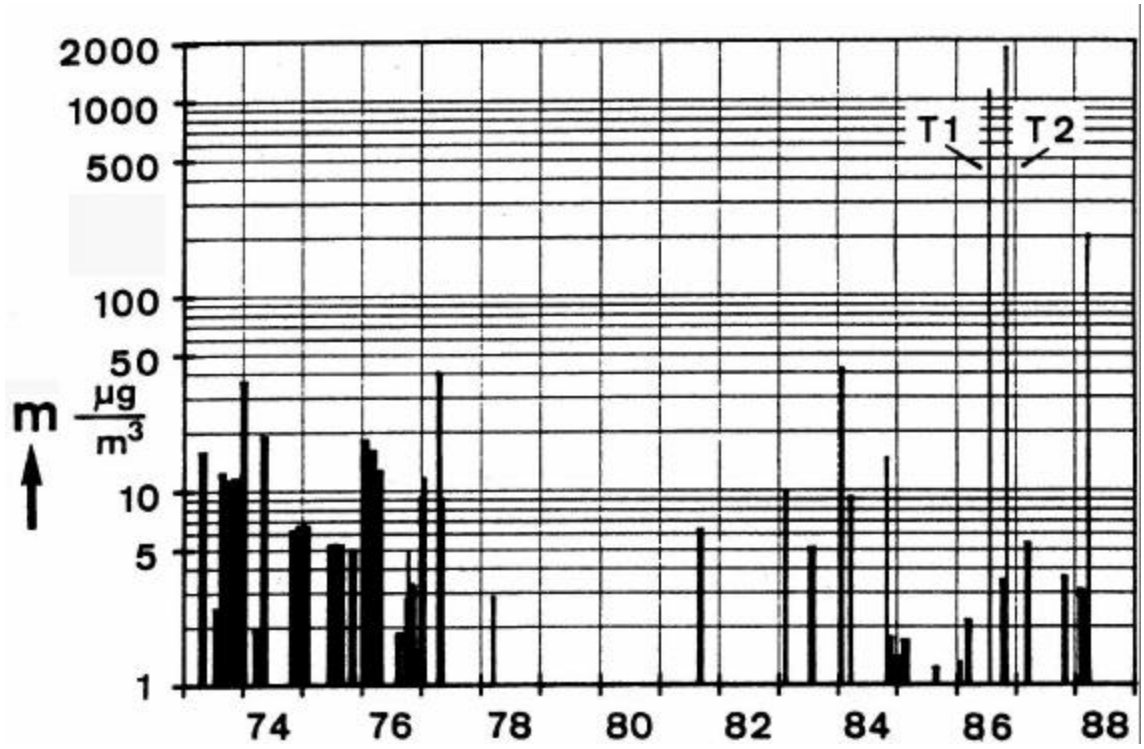



Figure 4. Dust Concentration in the primary Helium

In Figure 5 the particle size measured in the AVR experiments is shown. The frequency maximum of the particle size was measured with < 1 ì m. The part of dust deposit per circulation was determined to 0.5 – 2 %. This small value is not surprising , if one considers the small particle size.

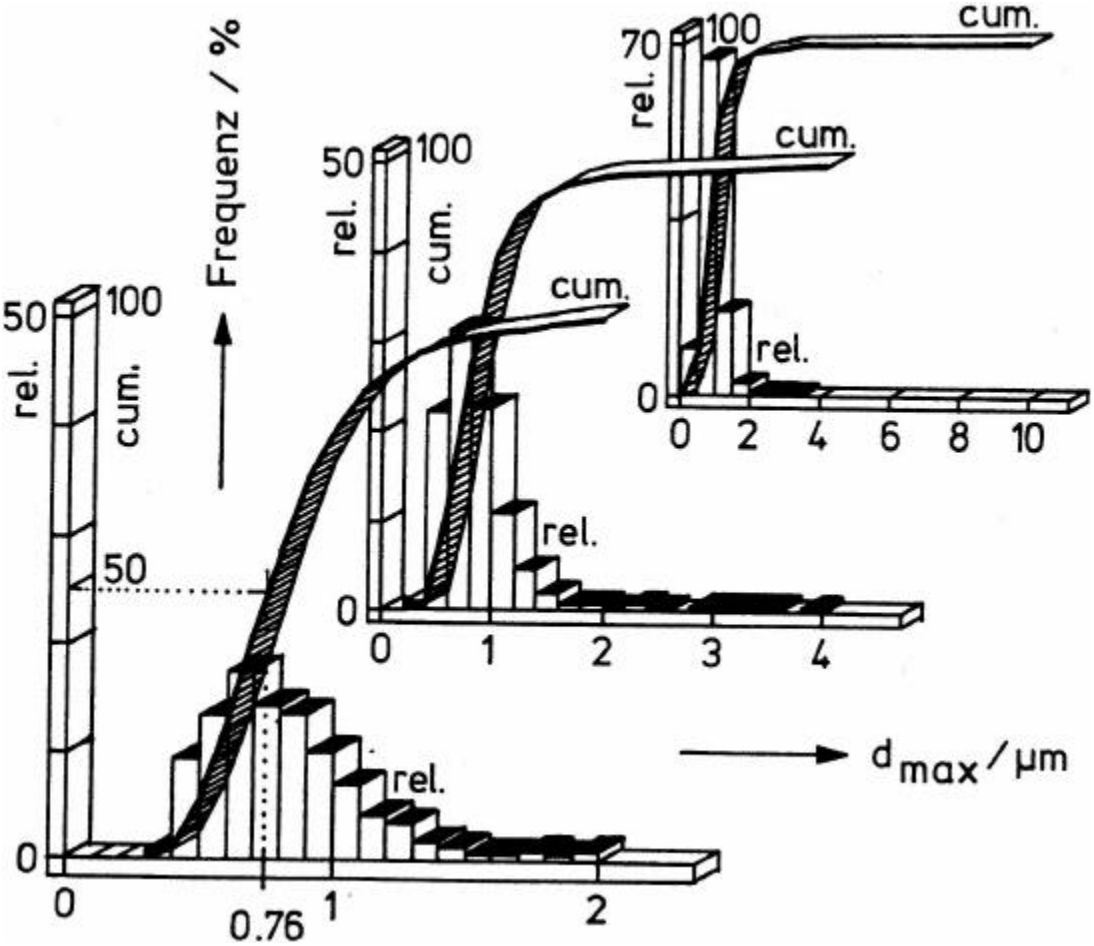



Figure 5. Particle Size in the AVR Dust

# 4 THTR

The helium purification unit of the THTR took advantage of the meanwhile established coated particles and the better knowledge of the release. The design criteria mentioned in the safety report are:

- limit of the water content in the circuit at a given maximum leakage of the steam generator
- carbon deposition on the steam generator
- preparation of pure gas for the auxiliary users like sealing gas, cleaning gas etc.
- The unit was divided into two stages:

## 4.1 Auxiliary helium systems (Figure 6)

The tasks for the auxiliary helium systems are

- Purification of the primary cooling gas to limit the content of impurities
- Supply of clean helium for several reactor components e.g. fuel handling, control rods
- Helium supply and storage
- Evacuation of the primary circuit prior to commissioning and after maintenance
- Purification of supplied fresh helium

## 4.2 Gas purification

Simplified flow-sheets of the purification units are given in Figure 7 and Figure 8.

### 4.2.1 First purification stage (Figure 7)

The helium is delivered from the higher pressure side of the helium blower and the clean gas delivered to the same pressure side. The flow through the stage is 0.8‰ of the helium flow. The stage consists of the following elements

**Copper oxide bed**

Task is the oxidation of chemical impurities carbon monoxide, hydrogen and methane to carbon dioxide and water. The copper is reduced. The ignition temperature of this BTS-catalyst is above 150°C. Therefor an additional heating of the incoming helium is not

necessary (260°C at normal operation). If the incoming gas should contain some oxygen works the reduced as an excellent trap. Normally the bed has to be regenerated with oxygen after more than 120hr.

**Molecular sieves**

The helium enters the molecular sieves with a temperature of 15°C (the last water cooler is operated with water from the cold water system 10°C). Molecular sieves with a porosity of 4Å are used. That avoids a considerable co-adsorption of fission noble gases Xenon (4.02Å molecular diameter) and Krypton (3.8 Å molecular diameter). Because of the better adsorption of water and the inhibition of the bed for carbon dioxide at a water contents >1% the bed is splitted in two parts one for water and one for carbon monoxide. The regeneration for the water adsorber will take place after 1000hr and for the carbon monoxide every 100hr. Because of the regeneration two beds of each type are necessary. The regeneration of the water sieve is done after an considerable increase of the water content in the outlet of the first bed.

The purified gas is compressed and recycled with a two stage blower in counterflow to the incoming gas.

4.2.2 Second purification stage ( Figure 8)

The 2$^{nd}$ stage of the purification has three tasks:

At **normal operation** cleaning of a gas flow of 100m³/h from radioactive noble gases (Xe, Kr) and removal of volatile impurities like nitrogen and methane.

At **commissioning and after maintenance** removal of volatile gases like nitrogen, argon, methane. For this purpose the 2$^{nd}$ purification stage is operated in series with the 1$^{st}$ stage and the flow is 2000m³/hr.

For **shut-down with depressurization** cleaning of the helium inventory of the primary circuit and transport to the gas storage.

Besides heat exchangers and a liquid nitrogen supply is the most important component

**Activated charcoal adsorber**

This adsorber is filled with activated carbon. The adsorber bed is cooled with liquid nitrogen and operated at a temperature of –192°C. The time between two regeneration cycles is more determined by start-up and shut-down than normal operation. In normal operation the operation time between two regeneration is 3000hr. For the regeneration the adsorber and the freezing heat exchanger are heated up to 150°C with preheated clean gas. The desorpted gas is collected in a storage and controlled released to the atmosphere.

To reach a continuos operation and reach also for $^{133}$Xe a sufficient decay time two parallel trains are available.

### 4.2.3 Experience

There were not many experience with the THTR because of the short operation time of the reactor. That for modern fuel elements a special purification in direction of radioactive noble gases is not a design criteria was confirmed. Also with the several maintenance actions the absolute necessity for a low temperature purification could not be confirmed.

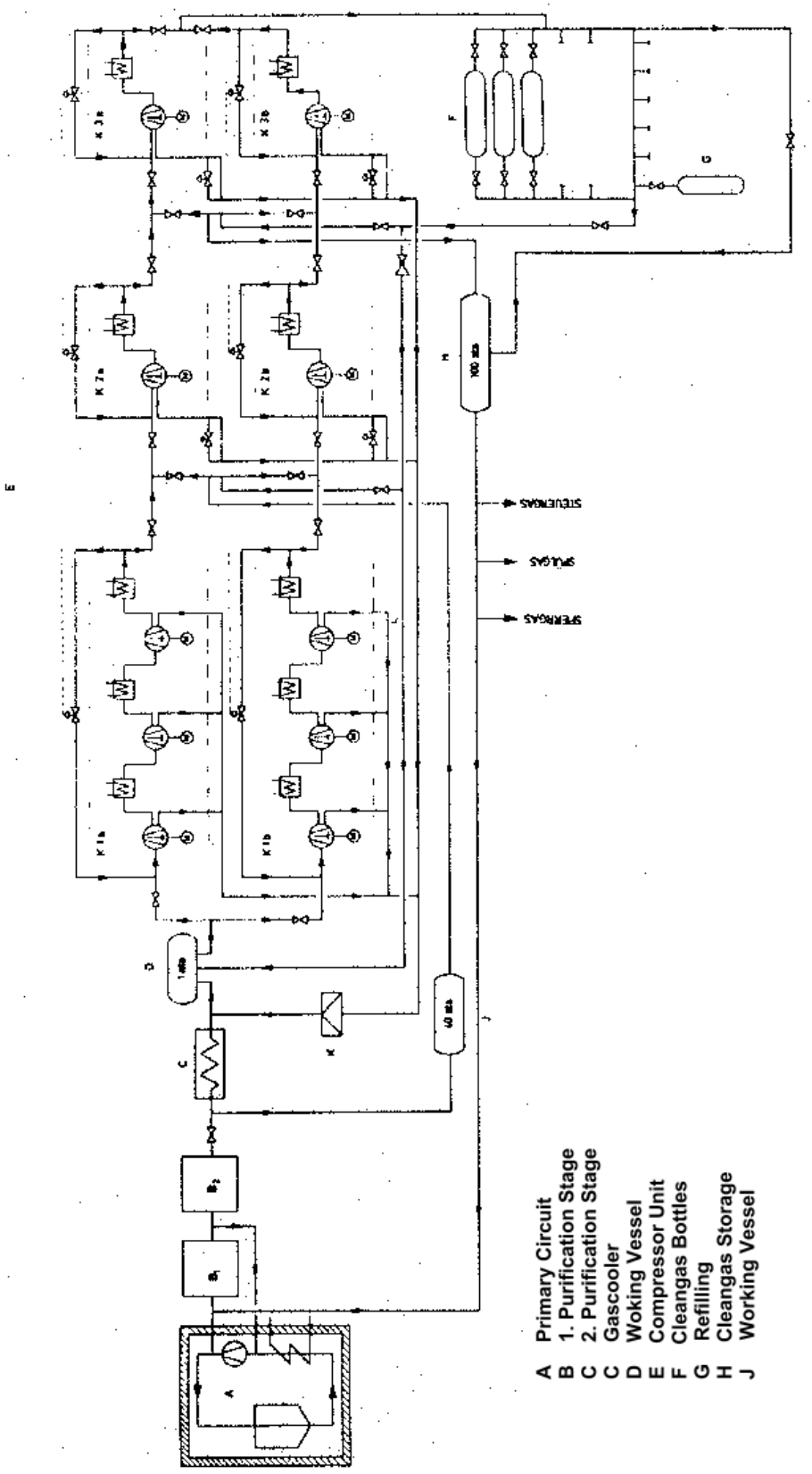



Figure 6 Cleangas Compressor Unit & Storage

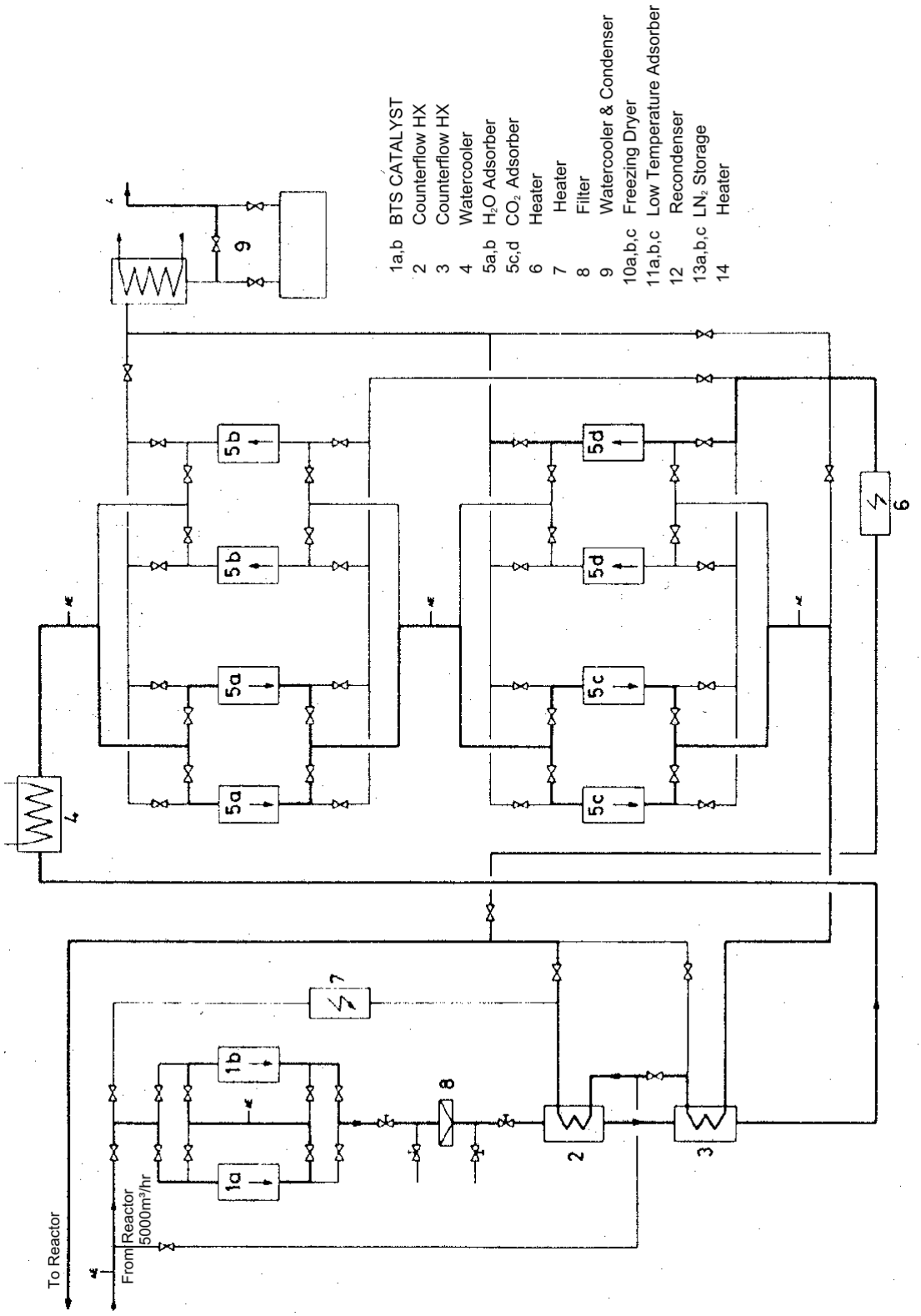



Figure 7 THTR Helium Purification 1st stage

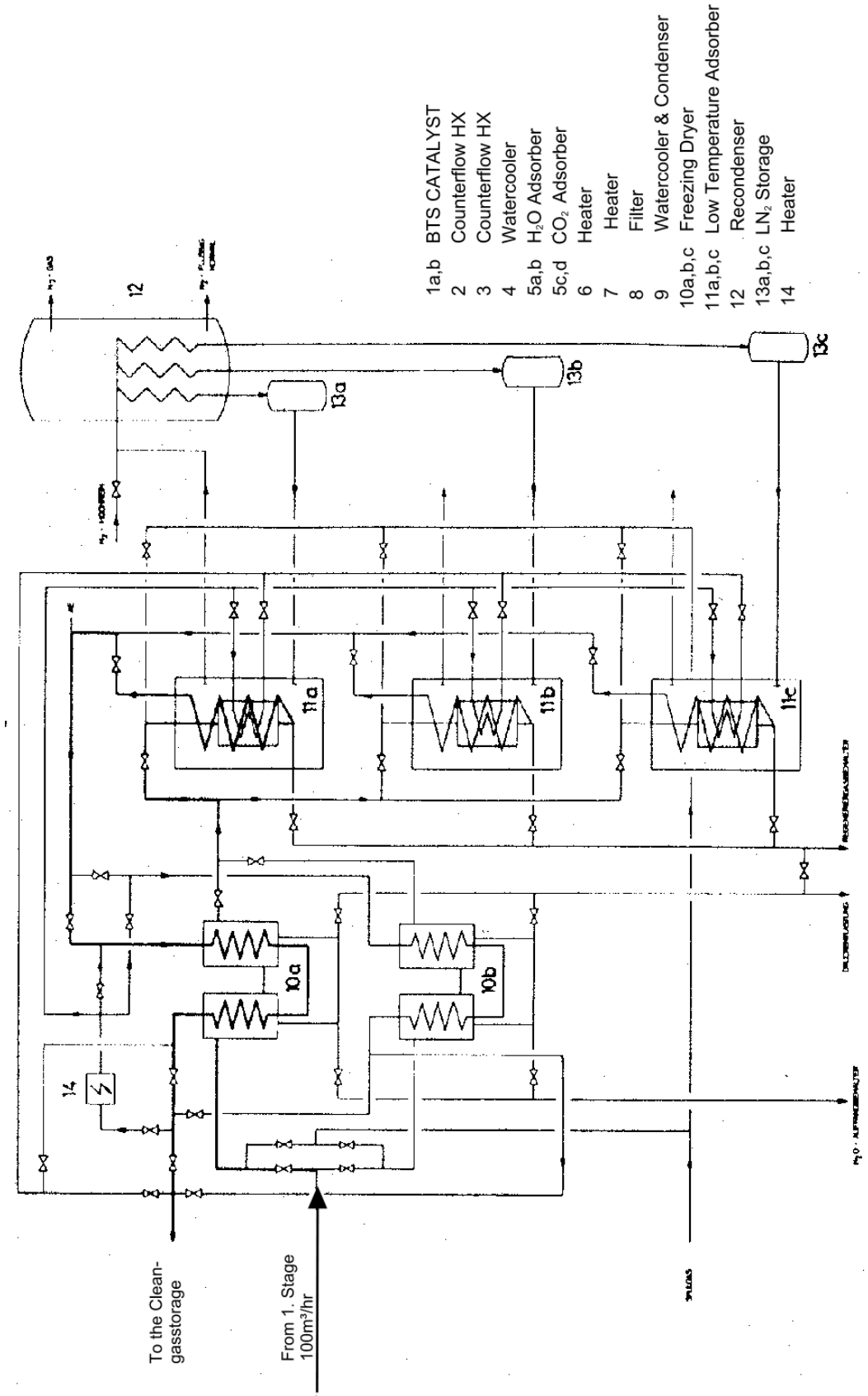



Figure 8 THTR Helium Purification 2nd stage

## 5 LIST OF FIGURES



## 6 LIST OF TABLES



## 7 LIST OF REFERENCES

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