**EUROPEAN COMMISSION**

**5th EURATOM FRAMEWORK PROGRAMME 1998-2002**
**KEY ACTION : NUCLEAR FISSION**

# HTR-E

## High-Temperature Reactor Components and Systems

**CONTRACT N°**
**FIKI-CT-2001-00177**

# ANALYSIS OF THE HELIUM ENVIRONMENTS IN FORMER HTRS


Hans F. Niessen

Forschungszentrum Jülich GmbH

GERMANY



Dissemination level : RE
Document N°: HTR -E- 02/12-D-6 -1 -6
Status : Final
Deliverable D 44

# Modifications

| Revision | Date | Modifications |
|---|---|---|
| A | November 2002 | First issue |
| | | |
| | | |
| | | |
| | | |
| | | |
| | | |
| | | |
| | | |
| | | |
| | | |
| | | |

| | Name | Signature | Date |
|---|---|---|---|
| Author | H. F. Nießen | H.F. Nießen | |
| | | | |
| | | | |
| Reviewed by | W. Scherer | W. Scherer | 13. 12. 2002 |
| | | | |
| | | | |
| Approved by | K. Kugeler | Kugeler | |
| | | | |
| | | | |

**Summary**

This report contains state of the art information on the helium purification of different HTRs in the UK the Dragon project, in Germany (AVR test reactor, THTR-300 and HTR Modul), and in Asia (HTTR and HTR-10).

The described systems are in principal similar. Two stages are applied:

1. Removal of chemical impurities to limit the corrosion of the core.
2. Removal and decay of radioactive gases

In the first stage as a “by-product” tritium is removed. The second stage has become smaller during the history because of advantages in fuel elements. Differences in detail are mostly connected to changes of the technology over the years (e.g. catalysts, compressors). Auxiliary duties of the gas purification system, which are connected to the fuel type and the fuel handling, result in design differences.

For the Dragon reactor the impurity content over several operation periods is given. The dust experiments at the AVR are discussed.

While the above mentioned reactors use the reactor heat indirect via heat exchangers, for HTR-E the direct cycle is foreseen. It might be necessary, that because of this, the helium purification system as an additional task has not only to fulfill the corrosion requirements of the core but also those of the gas turbine materials. Therefore one contribution to the report discusses the interaction of helium impurities with Ni-base alloys.

## Contents

1 Tasks of the helium purification sytem ..... 1
2 Helium purification in the Dragon Reactor ..... 2
3 The German former HTR's AVR and THTR ..... 3
3.1 AVR ..... 3
3.2 THTR ..... 3
4 Asian HTRs ..... 4
4.1 High Temperature Engineering Reactor (HTTR) ..... 4
4.2 High Temperature gas-cooled reactor test module (HTR-10) ..... 4
5 Helium Purification System of HTR Module ..... 5
6 Review of the gas/Ni-base alloys interaction in HTR impure helium ..... 6
7 List of attached Contributions ..... 7

# 1 TASKS OF THE HELIUM PURIFICATION SYTEM

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 of the system are:

1. Reduction of corrosive impurities in the primary coolant
    - Removal of gaseous contaminants from the primary coolant to maintain design values, especially oxidising gases and their reaction products with carbon in particular e.g $H_2O$, $CO$, $CO_2$, $H_2$, $CH_4$
2. Reduction of gaseous radioactive impurities in the primary coolant
    - Removal of tritium and its compounds with oxygen or carbon
    - Removal of inert gases, e.g. nitrogen
    - Removal and decay of radioactive noble gases
3. Support of operation procedures like:
    - Helium supply and storage
    - Evacuation of the primary circuit
    - Fuel handling (pebble bed type)
    - Storage of radioactive gaseous materials
    - Start-up purification of the primary system before initial start-up and after inspections and maintenance
    - Purification of newly delivered helium

The fulfilment of these tasks and in what way they are reached is analysed for different HTR-plants. Additionally the design philosophy and operational experience are given.

It could be necessary to extend the requirements of the Helium Purification System for modern direct cycle HTRs to have a larger choice in high temperature metallic materials (Ni-base alloys) for the turbine. Therefore the interaction of helium impurities with Ni-base alloys are reviewed.

## 2 HELIUM PURIFICATION IN THE DRAGON REACTOR

(Attachment 1)

The purification requirements are given for normal operation and during start-up period. The purification process is done by adsorption on nitrogen-cooled charcoal beds for impurity gases and gaseous fission products as well. Fission products escaping from the fuel boxes were continuously scavenged and transported to a trapping system by a small flow of helium, drawn from the primary cooling circuit via the interior of the graphite fuel element cans.

The helium purification consists of the following stages:

- A flow of scavenge gas to the top of each fuel rod.
- Scavenging away gaseous and volatile fission products appearing in the annular gap between the graphite fuel boxes and graphite outer fuel cans.
- Trapping out fission products likely to cause corrosion or blockage of subsequent parts of the system, using a trap situated in the bottom of each fuel rod.
- Gas stream containing the products from each fuel element into a common purge gas manifold, via equipment for equalising and sampling streams from the individual fuel elements, and detecting and locating faulty or unseated fuel elements.
- Condensing out volatile fission products and cooling the purge stream to a temperature not exceeding 100°C.
- Trapping out any remaining iodine and bromine fission product.
- Delaying the fission product gases xenon and krypton in water-cooled charcoal beds.
- Oxidising impurity gases such as CO, $H_2$ and $CH_4$ to $H_2O$ and $CO_2$.
- Removing water vapour and carbon dioxide in two beds of 4A molecular sieve, at ambient temperature.
- Further delaying the residual xenon and krypton in a liquid nitrogen cooled charcoal bed for sufficient time to eliminate activity due to all nuclides except $^{85}Kr$.
- Trapping the remaining stable xenon and krypton, together with $^{85}Kr$, and the residual non-oxidisable impurity gases such as $N_2$, Ar, etc, in a regenerable charcoal trap.
- Discharging the contents of the freezer unit and the regenerable charcoal trap at intervals to a system of dirty gas receivers where they will be sampled and monitored for activity. Then discharging them in a controlled fashion to the atmosphere via the stack.
- Warming up the cleaned helium and returning it to the reactor by means of purge gas circulators.

The impurity levels for the design, steady state operation, and after maintenance are given for the periods up to 1970. The purification system worked well.

# 3 THE GERMAN FORMER HTRS AVR AND THTR

(Attachment 2)

## 3.1 AVR

The helium purification of the AVR consists of the following steps:

- **Dust filter**
- **Oxidizing** of corrosive gas components like $H_2$, CO, $CH_4$
- **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** in a silica gel bed the final drying takes place. $H_2O$ and $CO_2$ are removed
- **LN cooler** The incoming gas is precooled with purified helium and afterwards with boiling liquid nitrogen to a temperature of –175°C.
- **Activated charcoal** to absorb and decay noble gas impurities krypton and xenon

Activities of the primary coolant are shown. The purification was in the low-temperature part oversized because of fuel element improvement, while a separator for larger amounts of water after a water ingress was missed.
Experimental results for dust concentration and the particle size are given.

## 3.2 THTR

The helium purification and the design philosophy is given. The tasks for the system are:

- limit of the water content in the circuit at a given maximum leakage of the steam generator
- minimise carbon deposition on the steam generator

The elements used are similar to those already described:
**Removing dust** to protect the following units of the gas purification
**Partial condensing** of water
**Oxidation** of chemical impurities in a copper oxide bed
**Absorption** at molecular sieves for water and carbon dioxide
**Activated charcoal absorber beds** (located in a cold box) as intermediate storage to give enough decay time for the radioactive noble gases

# 4 ASIAN HTRS

(Attachment 3)

## 4.1 High Temperature Engineering Reactor (HTTR)

The Japanese HTTR and its purification system is described. The different stages of helium purification system are similar to the other ones. The experimental results of the quantity of impurity eliminated by the purification train of the commissioning test are given. An interesting result of the tests is unsatisfactory removal of $H_2O$ in the molecular sieve. It has to be mentioned, that in the HTTR the same molecular sieve is used for $CO_2$ and $H_2O$. All other components of the helium purification are proved satisfactory. Operation difficulties were also recognised in the gas sampling system(gas analysis not purification).

## 4.2 High Temperature gas-cooled reactor test module (HTR-10)

The Chinese HTR-10 is described. The helium purification system of the HTR-10 is based on the HTR-modul. The HTR-10 is in the commissioning phase. Therefor besides design data no operational data are available.

# 5 HELIUM PURIFICATION SYSTEM OF THE HTR MODULE

(Attachment 4)

The helium auxiliary systems of an HTR Module power plant (twin plant) supply the primary circuits are shown. Each module has its own purification train. A third is in reserve is in common for both and can be switched on, if the reactors own system is not available. In common for both reactors is the low temperature supply and a water separation for accident cases (AVR experience).

Requirements for the operation but also safety aspects are shown.

The tasks for the system are:

- Purification of the primary coolant to remove entrained dustlike and gaseous impurities. Anticipated values for normal operation are for $H_20$ =0.1 vpm, for CO =2 vpm, for $N_2$ =1 vpm, for $H_2$ 55 vpm and for $CH_4$ =1 vpm on entry into the helium purification system. After accidents, short term maximum values can result as follows: $H_2O$ 3 vpm, $H_2$ 70 vpm, CO 100 vpm, $CH_4$ 5 vpm and $N_2$ 100 vpm, the purified helium is returned to the primary circuit with impurities $\leq$0.1 vpm,
- tritium removal,
- removal of other radioactive impurities from the helium, in particular before transfer to the purified gas store (Xe, Kr, Ar),
- start-up purification of the primary circuits prior to initial commissioning and after inspections and maintenance work,
- purification of newly delivered helium and
- removal of water after a water ingress accident.

A process description of the helium purification and the description of the auxiliary systems of the helium purification plant are given like:

Regeneration for molecular sieves, regeneration for low temperature absorbers, pressure relief for helium auxiliary systems, fuel handling, and gaseous waste storage, water extraction (post accident), water extraction (normal operation), helium supply and storage, evacuation for helium auxiliary systems.

Operation of the helium purification including normal operation, commissioning, purification operation, pressure control and accident operation are described. Important components of the helium purification are shown, e.g. copper oxide bed, molecular sieve, low temperature absorber, post accident cooler, other heat exchangers, coolers, heaters and blowers.

# 6 REVIEW OF THE GAS/NI-BASE ALLOYS INTERACTION IN HTR IMPURE HELIUM

(Attachment 5)

The interaction of the impurities $H_2O$, $H_2$, CO and $CO_2$ with the metallic surface are discussed. Because of the low concentrations of the impurities kinetic effects are more likely than thermodynamic equilibrium. Three parameters were identified:

- Oxygen activity given by the $\frac{P_{H_2O}}{\sqrt{P_{H_2}}}$-ratio
- Carbon activity given by $\frac{P_{CO}}{\sqrt{P_{O_2}}}$ -ratio
- $P_{CO}$, which controls the reaction between chromium oxide and chromium carbide.
- With these parameters stability diagrams can be build up, which allow the prediction of the behaviour of an alloy. To allow the formation of a protective layer three requirements for the helium impurities are necessary:

  Partial pressure of CO, $\frac{P_{H_2O}}{\sqrt{P_{H_2}}}$-ratio and $\frac{P_{H_2O}}{\sqrt{P_{CH_4}}}$ -ratio must exceed critical values, which increase with the temperature and decrease with Cr and Mo content of the alloy

  The effects of minor oxidizable elements e.g. Al, Si Ti can modify the behaviour of a Ni-base alloy significantly but is not predictable today.

# 7 LIST OF ATTACHED CONTRIBUTIONS

Attachment 1: Helium purification in the Dragon Reactor NNC (7) contribution
Reference HTR-E 02/09 D-6-1-5

Attachment 2: The German former HTR's AVR and THTR FZJ (5) contribution
Reference HTR-E 02/09 D-6-1-4

Attachment 3: Asian HTRs FRAN-ANP (1) contribution
Reference HTR-E 02/09 D-6-1-1

Attachment 4: Helium Purification System of HTR Module FANP GmbH (15) contribution
Reference HTR-E 02/06 D-6-1-3

Attachment 5: Review of the gas/Ni-base alloys interaction in HTR impure helium
FRAN-ANP (1) contribution
Reference HTR-E 02/10 D-6-1-2

All these references are available on SINTER web site forum HTR-E.