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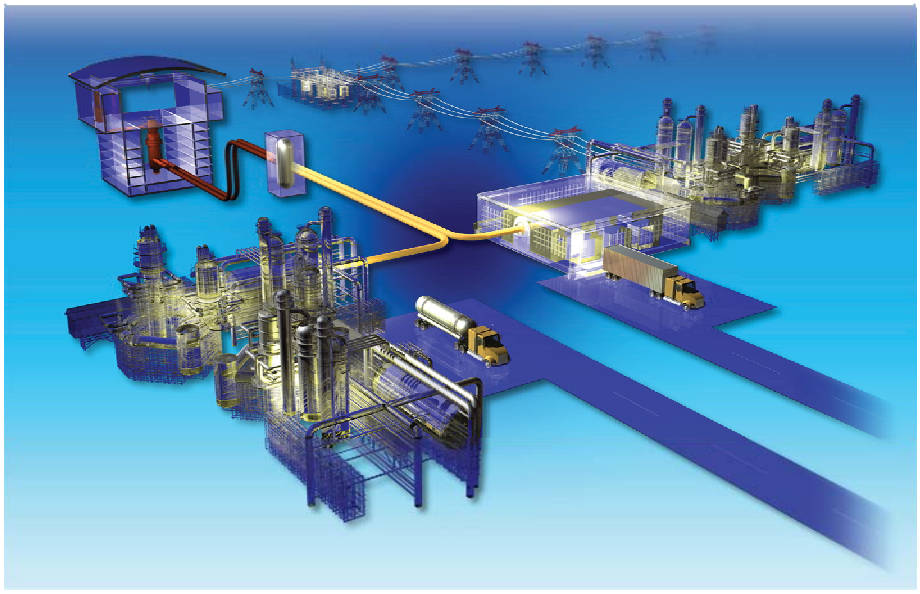
## Technical Evaluation Study

## Project No. 29147

# NGNP Component Test Capability Test Loop Helium Supply and Purification Study



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| NGNP COMPONENT TEST CAPABILITY TEST LOOP HELIUM SUPPLY AND PURIFICATION STUDY | Identifier: TEV-588<br>Revision: 0<br>Effective Date: 08/12/2009 | Page: 1 of 14 |
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## Signatures

| Signature and Typed or Printed Name | Signature Code | Date (mm/dd/yyyy) | Organization/Discipline |
|---|---|---|---|
| Lance Lauerhass | P | 8/11/09 | B210 Chemical Engineer |
| L. Lauerhass | | | |
| S Austad | A | 08-11-09 | FS&S Project Lead |
| S.L. Austad, P.E. | | | |
| Vandell Balls | A | 8/11/9 | CTC Project Engineer |
| V.J. Balls, P.E. | | | |
| Vince F. Tonc | A | 8-11-09 | CTC Project Manager |
| V.F. Tonc | | | |

**P** For Preparer of the document.

**A** For Approval: This is for non-owner approvals that may be required as directed by a given program or project. This signature may not be applicable for all uses of this form.

**C** For documented review and concurrence.

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## REVISION LOG

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| 0 | 08/12/2009 | All | Newly issued document |
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# 1. INTRODUCTION

## 1.1 Study Description and Objectives

The Next Generation Nuclear Plant (NGNP) Project will require testing to develop the technologies envisioned for the components and its subsystems. Testing ranging from metal coupon corrosion testing at the laboratory scale to engineering-scale testing of the intermediate heat exchanger (IHX) has been identified in the AREVA and WEC NGNP Technology Development Roadmapping reports[1,2] Pilot and engineering-scale level testing is proposed requiring a test facility capable of providing heat in the range of 1–30 $MW_{th}$. Conceptual design studies will establish the configuration of these gas circulation loops.

This study focuses on the purification associated with these gas circulation loops with the assumption that helium is the heat transfer media. Advantage will be taken of helium purification and supply design developments to date, reviewing alternatives and identifying issues requiring resolution as the facility design effort progresses. At this stage of the project, a specification for gas purity has not been decided. Consequently, the objective of this study is to provide a basis for future detailed design work and economic assessments.

This study is draft information only to support the August 4th & 5th Component Test Capability decision analysis meeting and will be finalized at a later date based on the alternative selected for the CTC.

## 1.2 Definitions/Glossary

The facility for component testing for NGNP is referred to generically in this study as the Component Test Capability (CTC). This facility could be provided by test facilities distributed across the country at various vendor facilities, foreign test facilities, or a new DOE test facility.

## 1.3 Acronyms

CTC – Component Test Capability

DOE – Department of Energy

IHX – Intermediate Heat Exchanger

NGNP – Next Generation Nuclear Plant

PFD – Process Flow Diagram

ppm – parts per million

PSA – Pressure Swing Adsorption

VHTR – Very High Temperature Reactor

# 2. BACKGROUND

## 2.1 Helium Purification System Purpose

Chemistry control is important for the helium coolant of high temperature gas-cooled reactors because impurities at high temperature cause corrosion of materials planned for use in heat exchangers, etc., and oxidation of the graphite used in the core (not an issue in the CTC where electric heaters replace the reactor core). In a test facility, where it is desired to mimic compositions anticipated in a nuclear reactor system, it is necessary to allow for both the removal and addition of gas components. The helium purification systems are installed in the primary and secondary helium cooling systems in order to reduce the quantity of chemical impurities. Impurities are added by injecting pure gas components available from storage.

## 2.2 Sources of Helium Impurities

Chemical impurities $O_2$, CO, $CO_2$, $H_2O$, $H_2$, $CH_4$, $N_2$, NO, $NO_2$ are anticipated in the helium loop from a number of different sources[3]. In the case where a nuclear reactor core is NOT present, impurities associated with specific sources include:

- $O_2$ and $N_2$ from loading, working operations, repair (welding, junctions while maintaining operation)
- $O_2$, $H_2O$, $H_2$, and $N_2$ due to degassing of internal metallic structures
- $O_2$, $CO_2$, $H_2O$, and $N_2$ from degassing of thermal insulators
- $NO_x$ from the reaction between $N_2$ and $O_2$ at high temperatures
- CO from decomposition of $CO_2$ at high temperatures.

In the primary loop cooling system used to cool a reactor core, additional impurities associated with specific sources include:

- CO, $CO_2$, $H_2O$, and $H_2$ due to degassing of graphite reflector
- CO, $CO_2$, $H_2O$, $H_2$, and $CH_4$ from fuel rod, reflector replacement, loading and unloading operations
- C from graphite particles in the reactor core

- Radiocontaminants originating from fission/activation products.

For reference, Table 1 provides impurity levels of helium reported for steady-state operation of several VHTRs [4].

Table 1. Level of impurities (ppm) in the helium loop of VHTRs at steady-state operation.

| | $H_2O$ | $H_2$ | CO | $CO_2$ | $CH_4$ | $O_2$ | $N_2$ |
|---|---|---|---|---|---|---|---|
| Dragon | 0.1 | 0.1 | 0.05 | 0.02 | 0.1 | 0.1 | 0.05 |
| Peach Bottom | 0.5 | 10 | 0.5 | <0.05 | 1.0 | — | 0.5 |
| Fort St. Vrain | 1 | 7 | 3 | 1 | 0.1 | — | — |
| AVR | 0.15 | 9 | 45 | 0.25 | 1 | | 22 |
| THTR | <0.01 | 0.8 | 0.4 | 0.2 | 0.1 | | 0.1 |

## 2.3 Helium Specification to Control Corrosion

Metallic materials for high temperature applications in a very high temperature reactor (VHTR) must meet high level criteria in terms of thermal stability and mechanical properties. Nickel-base alloys containing 22 wt% chromium strengthened by molybdenum, cobalt, and/or tungsten are promising candidates for the IHX. A key characteristic of such metal alloys is that at intermediate temperatures, helium impurities can promote the development of chromium-rich surface oxide scales. This chromium oxide layer protects the alloys against intensive corrosion processes involving the rapid gain or loss of carbon, referred to as carburization and decarburization. These carbon transfers cause dramatic deterioration in component mechanical properties[5].

Observing the following helium impurity specifications and temperature constraints promotes and maintains this protective chromium oxide layer:

1. Maintain a partial pressure ratio of $P_{H2O}/P_{H2} > 0.01$[6] in order to allow the formation of a protective chromium oxide layer according the reaction

   $$2/3\ Cr + H_2O = 1/3\ Cr_2O_3 + H_2. \quad (1)$$

2. Maintain $P_{CO}$ in excess of a critical value to avoid the following microclimate reaction, which results in the loss of the chromium oxide protective layer:

   $$Cr_2O_3 + 3\ C_{soln} = 3\ CO + 2\ Cr. \quad (2)$$

   This critical CO partial pressure value is a function of the material (chromium activity) and the temperature. For example, from Figure 1, $P_{CO}$ must exceed 30 μbar at 950ºC for Inconel and Haynes alloys in order to

drive reaction (2) in the reverse direction leading to formation of a chromium oxide layer. For a helium heat transfer loop operating at 9MPa = 90 bar, this CO partial pressure corresponds to a gas content of 0.33 ppm CO as shown in the following calculation:

$$CO_{\text{gas content}} = \frac{P_{CO}}{P_{\text{total}}} = \frac{30\mu\text{bar}}{90\text{bar}} \times \frac{1\text{bar}}{10^6\,\mu\text{bar}} = 0.33\times10^{-6} = 0.33\,\text{ppm}$$

3. Maintain $P_{CH4}/P_{H2O}$ lower than a critical value $(0.01)^6$ to avoid excessive carburization and reduction of oxide layers. At lower values for this ratio, the Cr formed by reaction (2) reacts with water vapor according to reaction (1) to regenerate the oxide. At higher values for this ratio, the produced metal preferentially undergoes reaction with $CH_4$ according to reaction (3), leading to extensive carburization of the substrate.

$$23\ Cr + 6\ CH_4 = C_6Cr_{23} + 12\ H_2 \qquad (3)$$

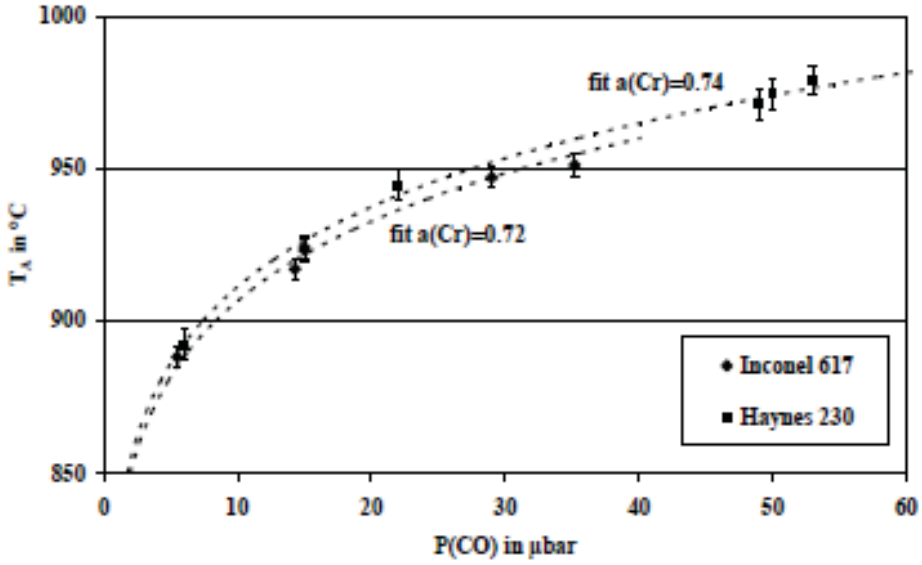


Figure 1. Critical temperature $T_a$ as a function of CO partial pressure[5]

## 2.4 Helium Loop Impurity Requirements for Testing

Target helium loop impurity levels will be driven by the most stringent requirements for tests planned in the CTC. The most complex gas mixture mentioned in the AREVA and WEC proposed tests involve the IHX. The general capability for simulating impurity levels for CO, $CO_2$, $H_2O$, $H_2$, $O_2$, $CH_4$, and $NO_2$ has been proposed by AREVA[1] for both the IHX Pilot Scale Demonstration (targeted for 1 $MW_{th}$ test loop) and the IHX Engineering Scale Demonstration (targeted for 30 $MW_{th}$ test loop). The desired impurity level requirements for

these gases have not yet been specified, which will be necessary for assessing the need and sizing for the various helium purification train unit operations described in section 4.2.

There is also a desire to include carbon dust for the Pilot Scale IHX test, to simulate particulate material expected in the primary loop, which is generated from a reactor utilizing pebble bed technology. It is recommended that for such testing, the carbon dust be limited to a dedicated test station that can be isolated from the rest of the test facility and have its own filtering system. Carbon dust would be injected immediately upstream of the test section and a filtering system would remove the carbon immediately downstream of this section.

# 3. LIMITATIONS AND ASSUMPTIONS

## 3.1 Assumptions

- Gas content is nominal 100% helium.
- Maximum loop pressure of 9 MPa. This pressure maximum selected by WEC is higher than the 8.4 MPa primary loop pressure cited in the AREVA design.
- Maximum gas temperature is 1030°C. This is the maximum temperature specified by AREVA which is more extreme than the 950°C maximum set by WEC. This maximum temperature in the heat transfer loop far exceeds the 400°C anticipated in the helium purification train located in the cold leg of the proposed loops as mentioned in section 4.1.

# 4. HELIUM PURIFICATION LOOP DESIGN

## 4.1 Purification Loop Placement

Because of the need for corrosion control, a system for maintaining target impurity levels of helium is required in both the primary and secondary cooling systems. Figure 2 shows the typical placement of the helium chemistry control system relative to main loop components. As shown in the schematic, a fraction of the main cooling stream flow (typically 5–10%) is diverted to the purification system. This occurs downstream of the circulator in the cold leg of the loop.

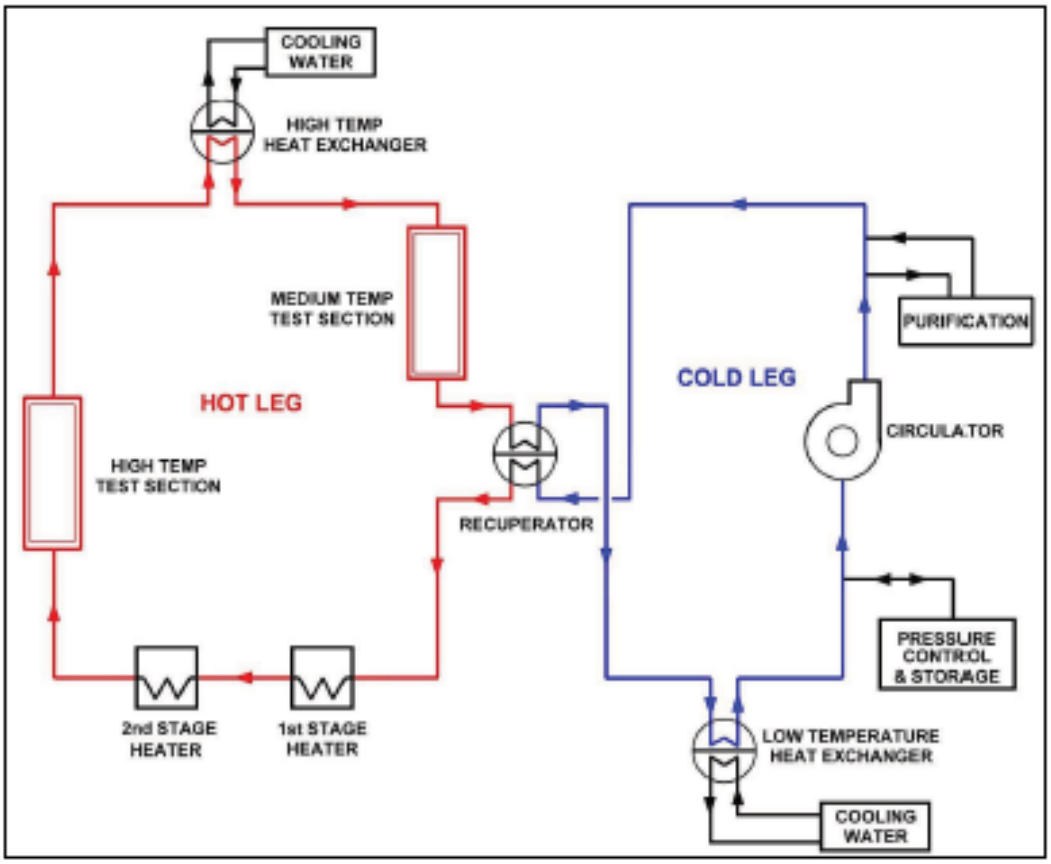


Figure 2. Typical location of He purification system relative to main loop components.

## 4.2 Purification Train Unit Operations—Cryogenic System

The typical sequencing of unit operations constituting the helium purification train is shown in Figure 3. This approach to impurity removal is common to several test facilities[7] (German EVA II and KVK, Japanese HENDEL). Figure 4 from the AREVA Pre-Conceptual Design[8] shows the operating conditions in the separation units and use of economizers for heat integration. A description of the role of each of these separation operations[3] in removing impurities is provided in the following sections.

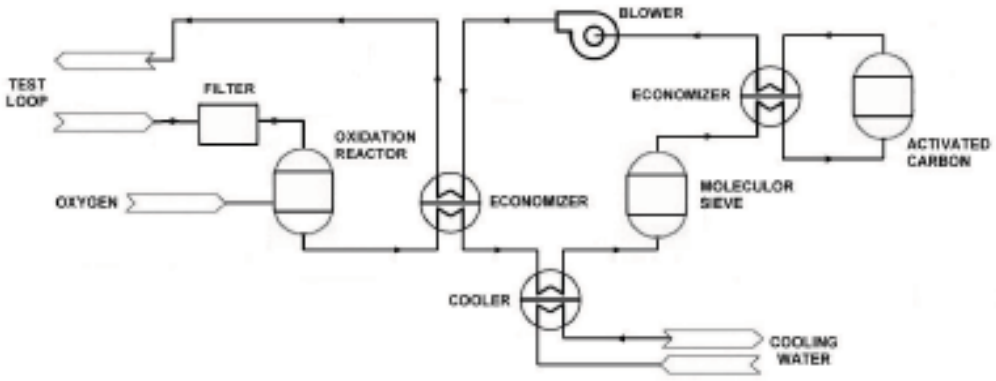


Figure 3. Major components of a typical helium purification train.

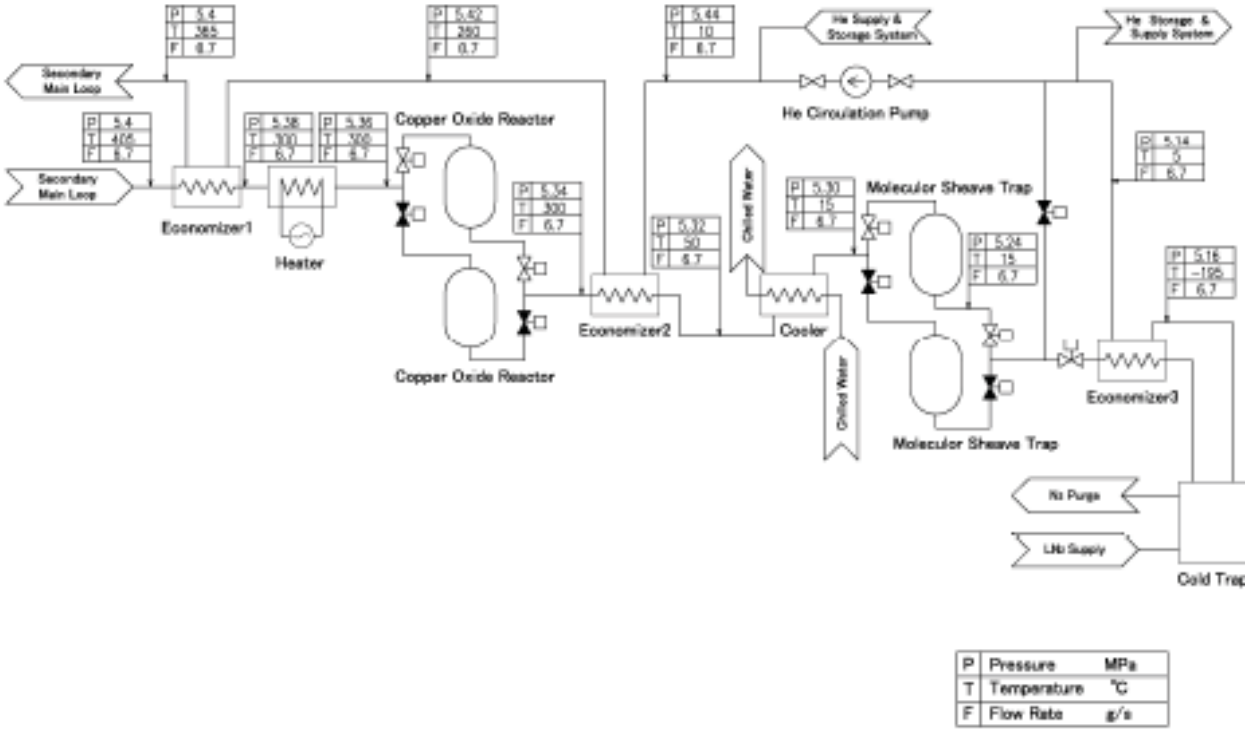


Figure 4. PFD with heat and mass balance of secondary helium purification system.

### 4.2.1 Filter

Filtration is the first cleanup step performed to ensure that any dust or particles within the system are removed in advance of downstream separation steps.

### 4.2.2 Oxidation Reactor

Since $H_2$ and CO are not readily absorbed by the downstream operations, a metallic oxide fixed bed utilizing CuO is employed to convert these components to more readily separable $H_2O$ and $CO_2$. The relevant reactions are

$$H_2 + CuO = H_2O + Cu \quad (4)$$

$$CO + CuO = CO_2 + Cu. \quad (5)$$

The column is thermally controlled using electrical heating for operation at 300°C.

### 4.2.3 Molecular Sieve

The role of the molecular sieve bed is to absorb $NO_x$, $CO_2$, $H_2O$, and $CH_4$. The adsorption of these species is expected to be able to reduce the level of these components in the gas to approximately 2 ppm.

Although not depicted in the flowsheet in Figure 4, regeneration is accomplished by heating the bed to 350°C by energizing the integral molecular sieve trap heater and driving the water to the downstream water condensate trap.

### 4.2.4 Activated Carbon Cold Trap

The activated carbon bed operates under cryogenic conditions (−180°C) to adsorb $N_2$ and $O_2$ along with the residual of the other impurities ($NO_x$, $CO_2$, $H_2O$, and $CH_4$). The adsorption of $N_2$ and $O_2$ is expected to reduce the level of these components to about 2 ppm. For the other impurities, it is assumed that a concentration of 1 ppm is reached at the end of the bed.

For the regeneration step, helium is removed from the bed and a vacuum pump lowers the pressure down to 10 kPa in the columns. Heating of the column, which can be done in 2 steps in order to limit thermal constraints, can then begin. First, liquid nitrogen is removed and air at room temperature is sent into the external chamber until the temperature in the column reaches 0°C. The air is then heated using a resistance heater in order to increase the column temperature up to 150°C for the regeneration of the adsorbent.

### 4.3 Alternative Purification Systems

A 2006 INL Conceptual Design Study[9] proposed a pressure swing adsorption (PSA) system rather than the cryogenic separator utilized in the AREVA design discussed above. In this approach, the separation train ends with the molecular sieve units of the AREVA design where the same pressure cycling of the molecular sieve columns between helium loop operating pressure (max 9 MPa) and 10 kPa vacuum occurs. Depending upon the required gas impurities in helium and their specifications established in the future for proposed pilot and engineering-scale tests, it may be possible to eliminate the need for the cryogenic separator unit.

## 5. PURIFICATION LOOP COSTS

Developing the costs for a helium storage system and a helium purification loop system for composition control is premature at this time because of the lack of design specifications. Key design uncertainty exists in the following areas:

- Heat transfer loop configuration and size
- Specific components that must be present as impurities in the helium heat transfer media to meet the objectives of planned pilot and engineering-scale tests
- Acceptable composition range for control of these component impurities

Once the design specifications become developed through on-going and future studies in these areas, a design and the associated costs for helium storage and heat transfer media composition control can be developed.

## 6. FUTURE SUPPLY OF HELIUM[10]

As a decay product of uranium and thorium isotopes located in the earth's mantle, helium has slowly evolved over the 4.5 billion years of earth's existence. Where geological formations exist capable of trapping gases, helium has collected over the eons along with natural gas. Untrapped helium seeps out of the earth's mantle and drifts into the atmosphere where it is present at about 5 parts per million. This helium, along with any let into the atmosphere by users, drifts up and is eventually lost to Earth.

Since helium is a separation by-product of natural gas production, its largest sources in the United States are the natural gas fields in southwest Kansas and the panhandles of Texas and Oklahoma which contain unusually high percentages of helium, from 0.3% to 2.7%. However, these gas reserves are being depleted so quickly that this domestic source of helium is expected to be nearly depleted by 2015.

Beyond the United States, helium is marketed in Australia and Algeria. Russia has the world's largest reserves of natural gas, where helium certainly exists, making Russia the world's potentially major helium source in 30 years.

This depletion of United States helium reserves and reliance on Russia for future supplies raises concerns regarding the availability and cost of helium in the future. Given the envisioned future reliance of potentially hundreds of NGNP reactors on helium as a heat transfer medium, it is recommended that an assessment be conducted of the severity of this helium supply/cost issue and the need to incorporate alternatives to helium in the NGNP technology development plan.

# 7. CONCLUSIONS AND RECOMMENDATION

As the design of the CTC progresses, further study is needed to examine the following issues identified in this study related to the helium supply and purification loop composition control:

- Establish purity requirements and cost implications for the various tests planned in the CTC in order to guide the range of tests that will be accommodated by the facility.
- Evaluate potential elimination of the cold trap in the AREVA purification train with reliance on PSA to achieve required helium purity levels.
- Perform analysis of purification system sizing of components and system flow requirements to support the selected heat transfer loop(s) configuration.
- Study the trade-offs between the cost of helium purchased at various purity levels vs. the cost of using the CTC helium purification system to reach target impurity levels.

# 8. REFERENCES

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# 9. APPENDIXES

None