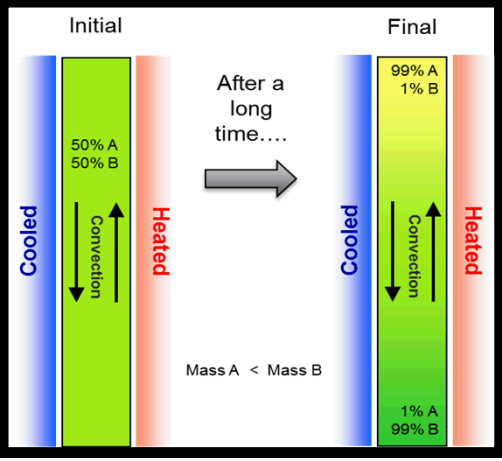
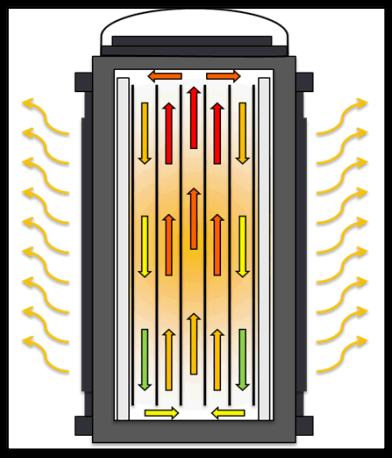
Exceptional service in the national interest

SAND2015-9200PE





Potential for Gas Separation by Thermal Diffusion in the High-Burnup Demonstration Cask

Charles Bryan, Sandia National Laboratories

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000. SAND NO. 2015-XXXXX

High Burn-Up Demonstration Project:
Gas Separation via Thermal Diffusion?

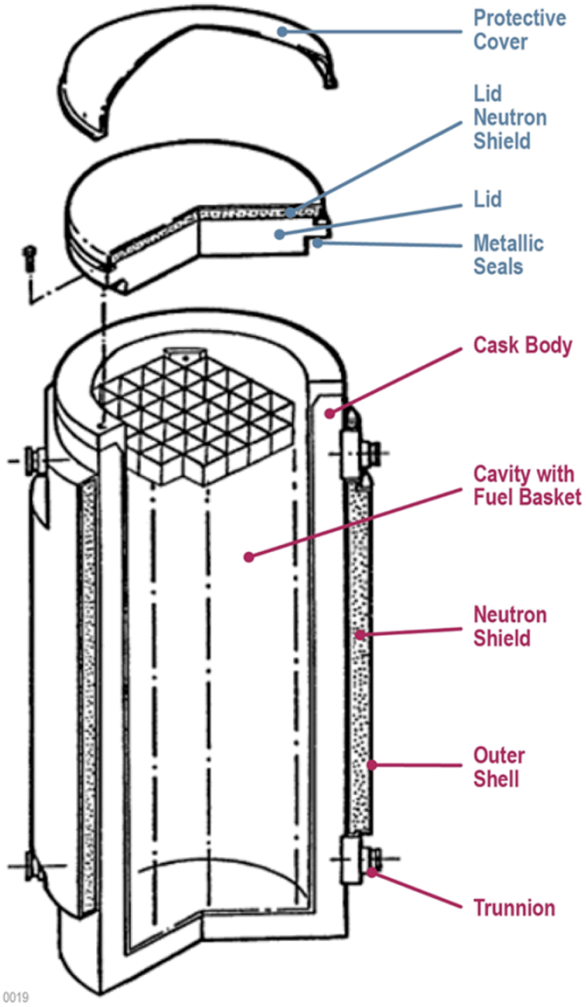
Sandia National Laboratories

- Demo cask (TN-32) filled with helium
- Periodic gas sampling for Krypton as an indicator for cladding failure
- Gas samples will be taken from top of the canister.
- Can thermal diffusion result in vertical compositional gradients within the canister?
- Can He and Kr be sufficiently separated by this process (He at the top) to inhibit detection of Kr?

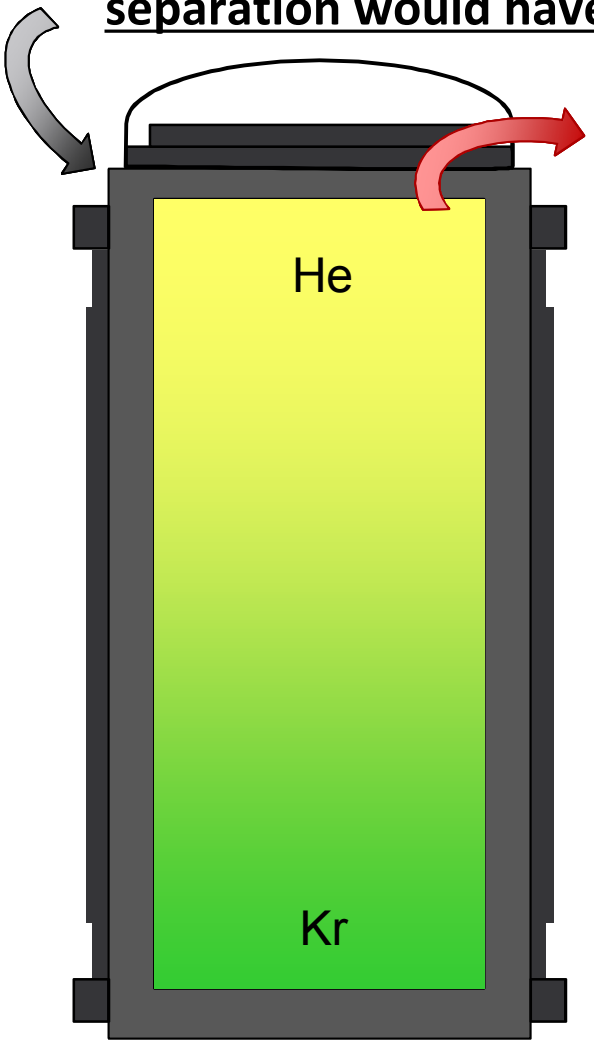
It is not necessary to quantify the amount of Kr present; it is only necessary to detect it. How much separation would be required to limit detection? What is the gas volume sampled, and what are detection limits? What are expected amounts of Kr present? *Assume a large degree of separation is necessary to affect detection.*

Relevance to the High-Burnup Demo Cask?

TN-32 cask:



To inhibit detection of Kr, high degrees of vertical separation would have to occur

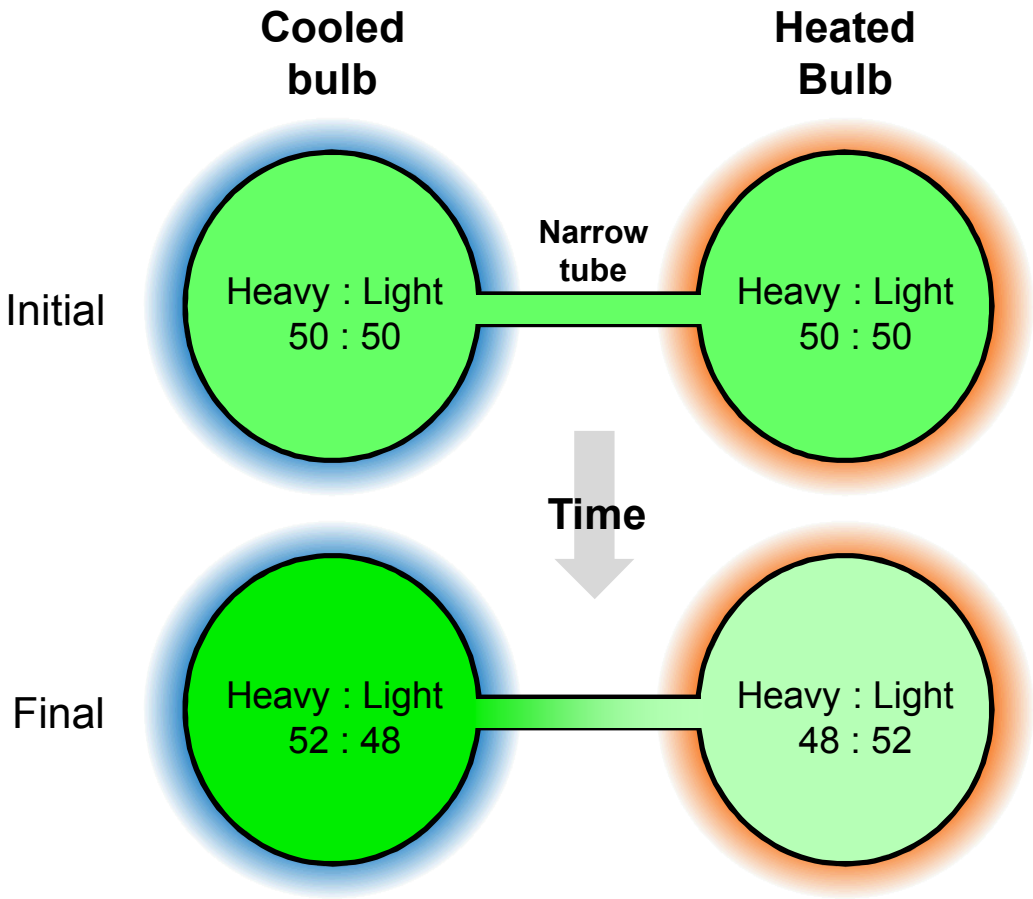


Samples will be taken from top of the canister. Can sufficient gas separation occur to inhibit Kr detection?

Thermal Diffusion:

Static System

In a 2-component static gas system with a temperature gradient, thermal (Soret) diffusion will result in a small amount of separation between the two gases. The lighter component will be slightly concentrated towards the hotter end, while the heavier component will slightly concentrated towards the cooler end.



Thermal Diffusion:
Static System

General equation for thermal diffusion:

$$n'_{10} - n''_{10} = k_T ln\left({}^{T'}/_{T}\right)$$

The degree of separation is a function of the temperature difference

where:

n'_{10} is the conc. of component 1 at the low temp end

n''_{10} is the conc. of component 1 at the high temp end

$(n_{10} - n'_{10})$ is termed the separation

k_T is the thermal diffusion ratio

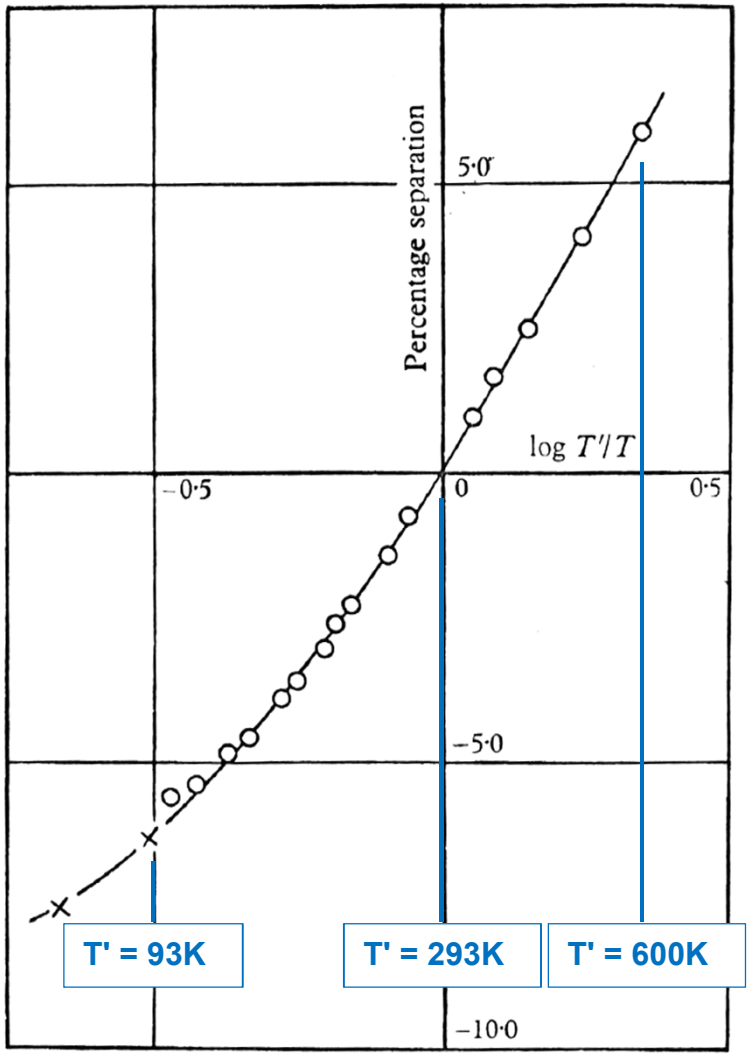
T' is the temperature at the hot end

T is the temperature at the low end

Static System:
Effect of Temperature

Relation between separation (n'_{01}- n''_{01}) and log (T'/T) for H_2-N_2 mixture (Mass ratio ~ 2:28) containing 39.8% H_2.

T = 293K



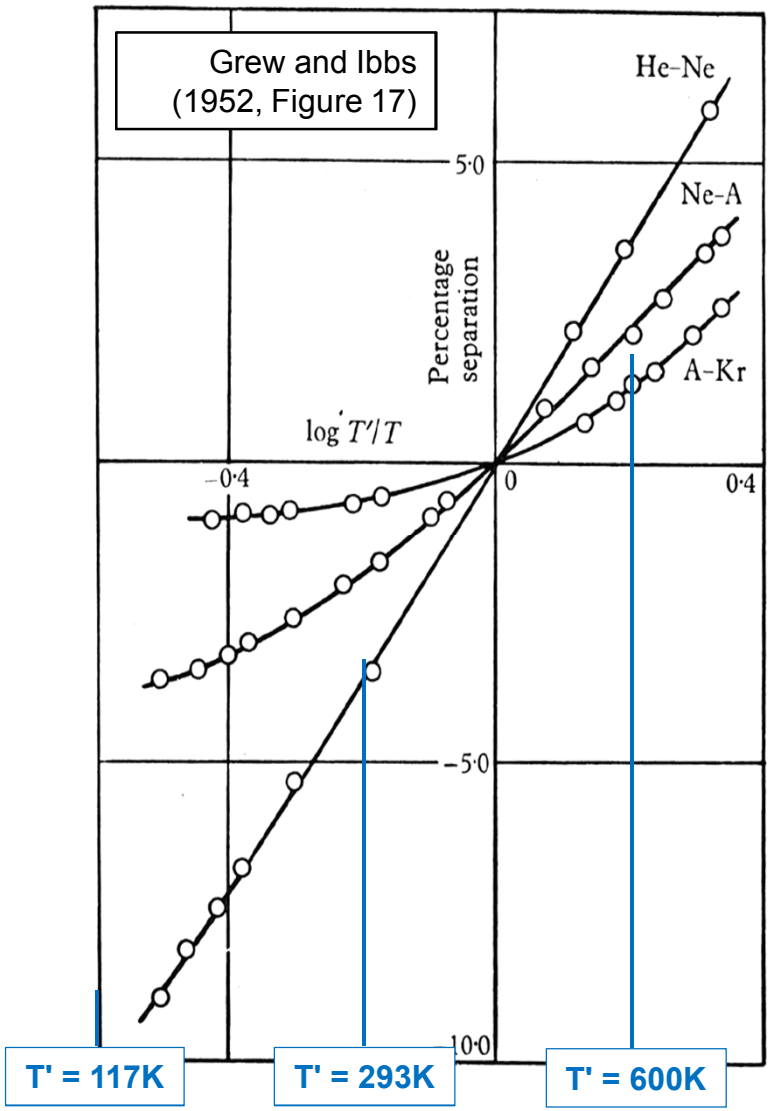
Grew and Ibbs, (1952, Figure 5)

Static System:
Effect of Temperature

Relation between separation (n'_{01}- n''_{01}) and log (T'/T) for mixtures of noble gases.

- He-Ne (53.8% He)
  (Mass ratio 4:20)
- Ne-Ar (51.2% Ne)
  (Mass ratio 20:40)
- Ar-Kr (53.5% Ar)
  (Mass ratio 40:84)

T = 293K



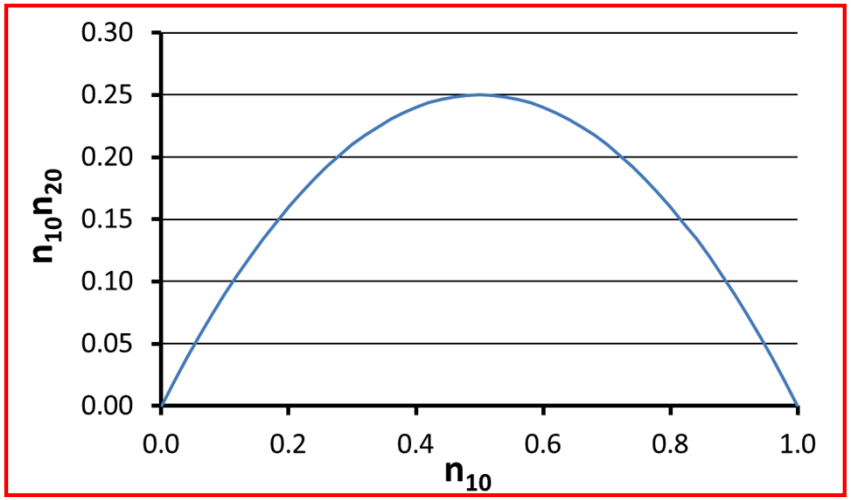
Static System:

Effect of Mass Difference and Composition

Thermal diffusion coefficient:

$$k_T = \left(I - \frac{a}{2}\right) \frac{a_1 n_{10} - a_2 n_{20}}{b_1 n_{10}^2 + b_2 n_{20}^2 + b_{12} n_{10} n_{20}} n_{10} n_{20}$$

The degree of separation is a function of the relative proportion of component 1 and 2, and is maximum when $n_{10} = n_{20}$.



where:

- n_{10} and n_{20} are the bulk concentrations of components 1 and 2, respectively
- $\left(I - \frac{a}{2}\right)$ describes the extent to which the gas atoms/molecules behave as rigid elastic spheres
- a_1, a_2, b_1, b_2, b_{12} are functions of the molecular diameters and masses

The larger the mass difference, the greater the degree of separation. The larger the size difference, the greater the separation

Should conditions be favorable, ^{4}He and ^{85}Kr might be expected to separate measurably.

Static System:

Effect of Mass Difference and Composition

Separation as a function of composition for mixtures of Kr with:

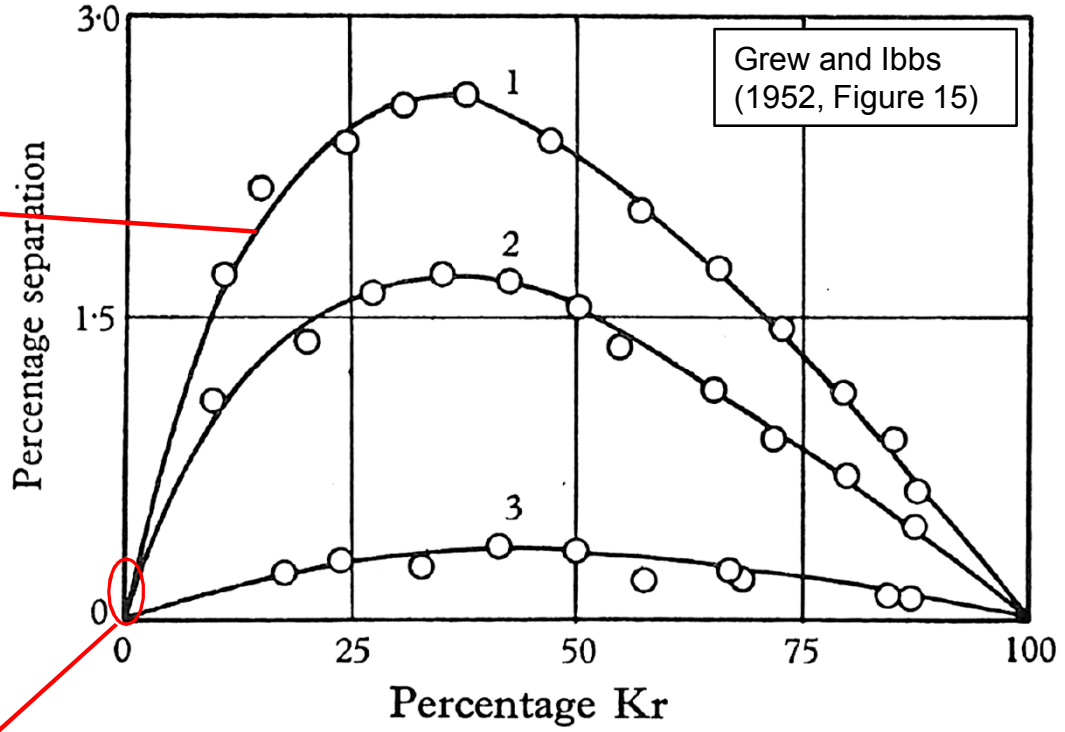
(1) **He (MR = 4:84)**

(2) Neon (MR = 20:84)

(3) Argon (MR = 40:84)

T = 288 K

T' = 373 K



In the high-burnup demo, very low Kr concentrations will inhibit any thermal separation

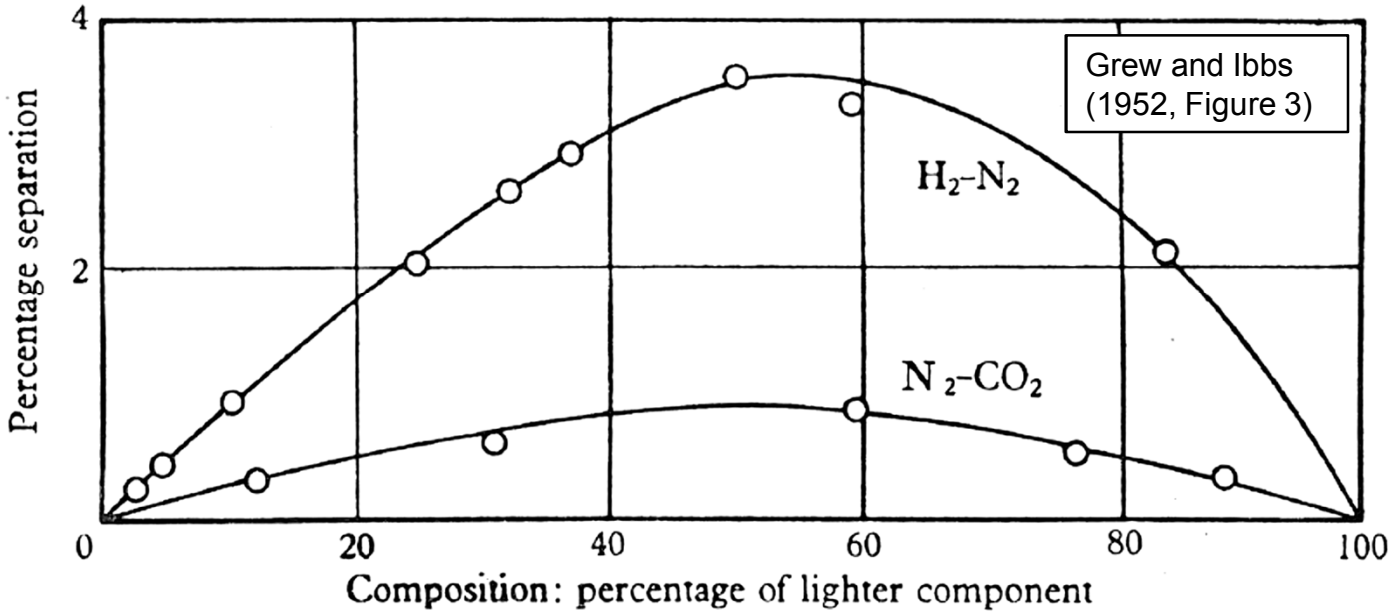
Static System:

Effect of Mass Difference and Composition

Relation between separation ($N'-N_0$) and composition for H_2-N_2 mixtures and for N_2-CO_2 mixtures.

T = 283 K

T' = 450 K



High Burnup Demo Cask as a Static System

Sandia National Laboratories

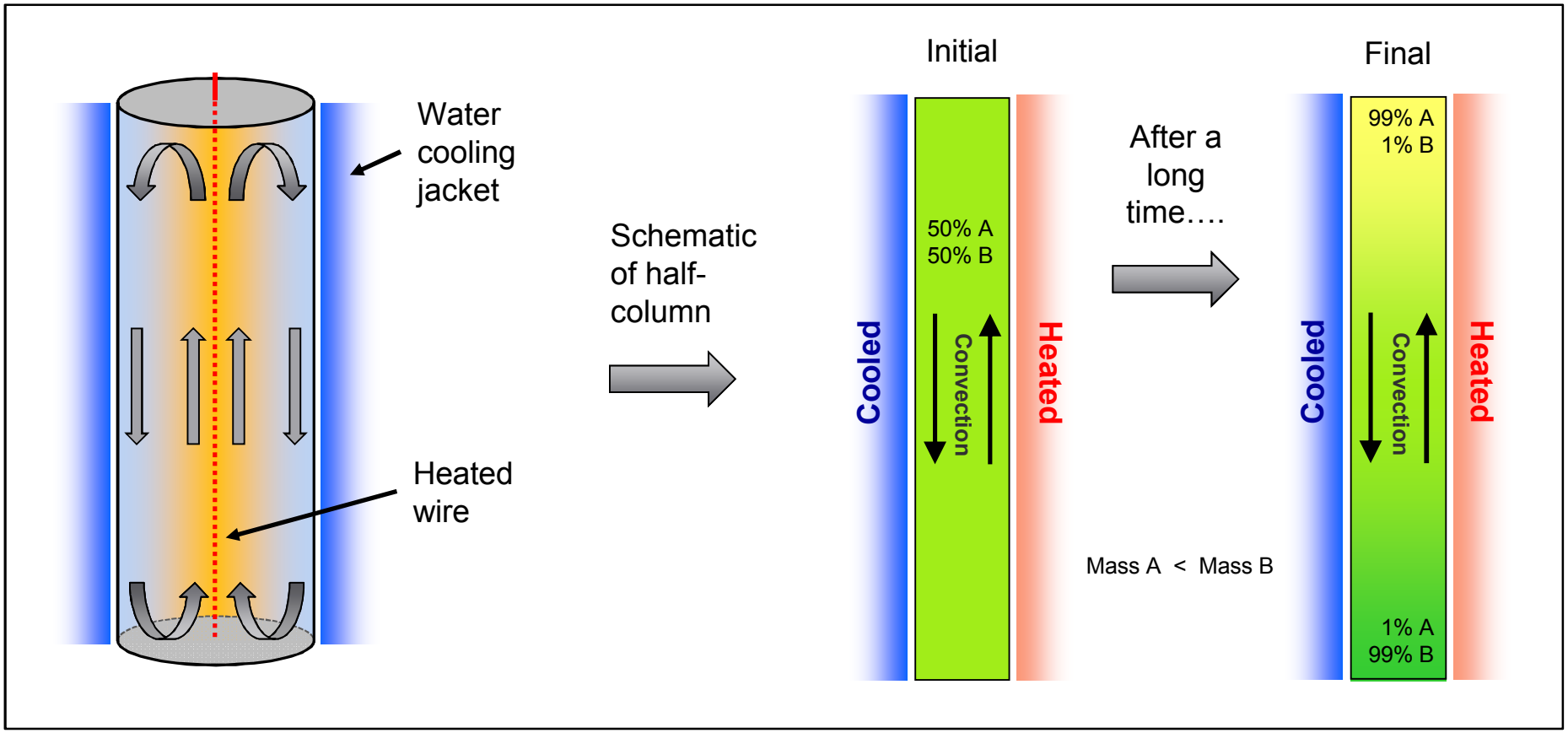
Static system?

- **If the cask were a static system, significant separation of gas components could not occur. Very small Kr concentrations inhibit separation, and temperature gradients are low, and would not yield a high degree of separation. Moreover, temperature gradients are not vertical (the canister cools by heat loss from the sides), so vertical separation would not occur. Kr detection in a static system would not be impaired.**

- **The cask is not likely to be a static system**, unless internal structures within the cask partition into regions and inhibit convection. This does not appear to be the case. What is the effect of thermal diffusion in a convecting system?

Convecting System:

Example of a Clusius-Dickel column

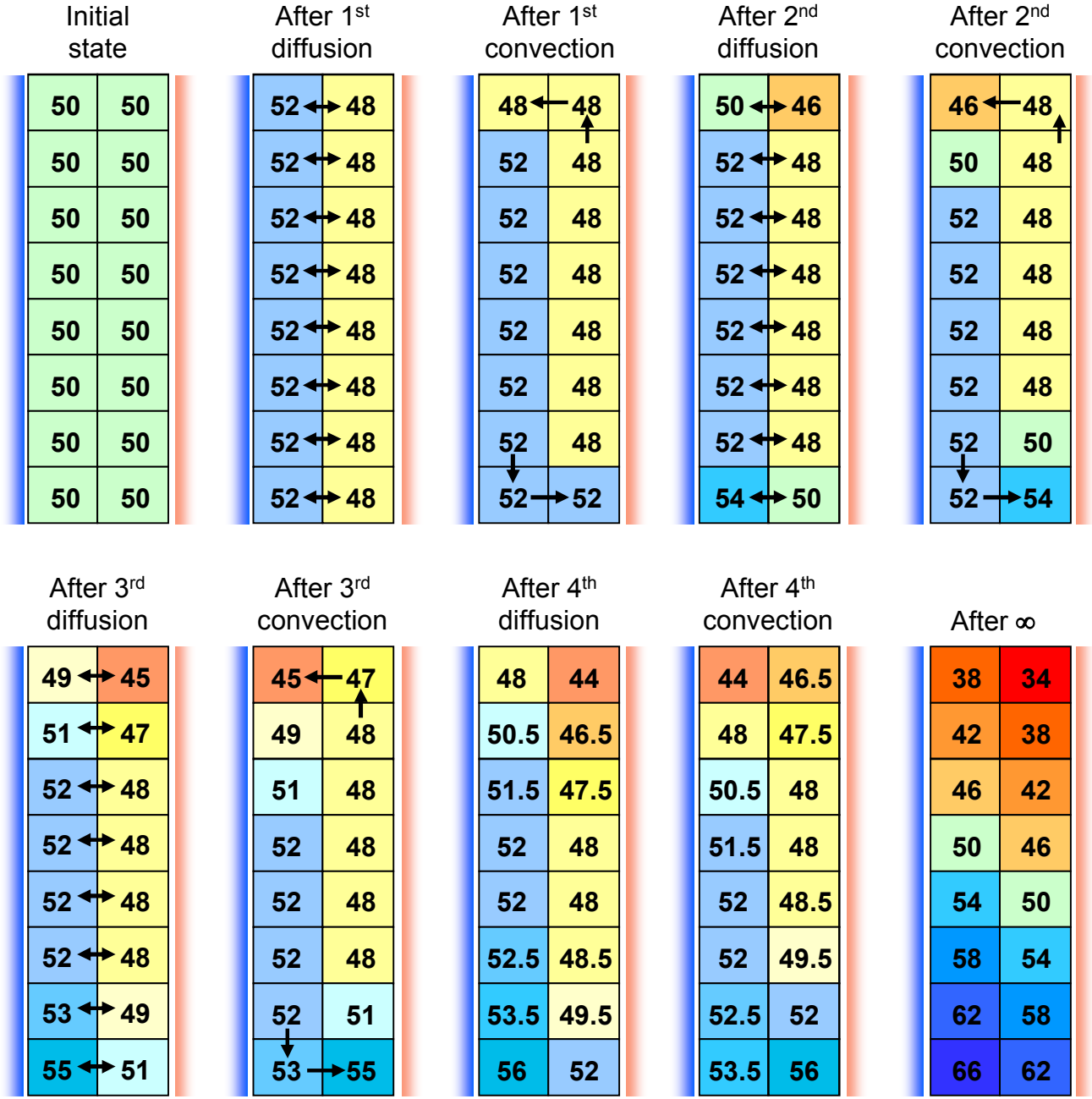


Mechanism of separation:

Combined effects of diffusion and convection.

The numbers reflect the concentration of the heavier phase in the mixture.

In this example, the final enrichment is a function of the number of horizontal rows into which the column is discretized. In the real case, there is no discretization, and near-purity can be achieved at the ends of the column.

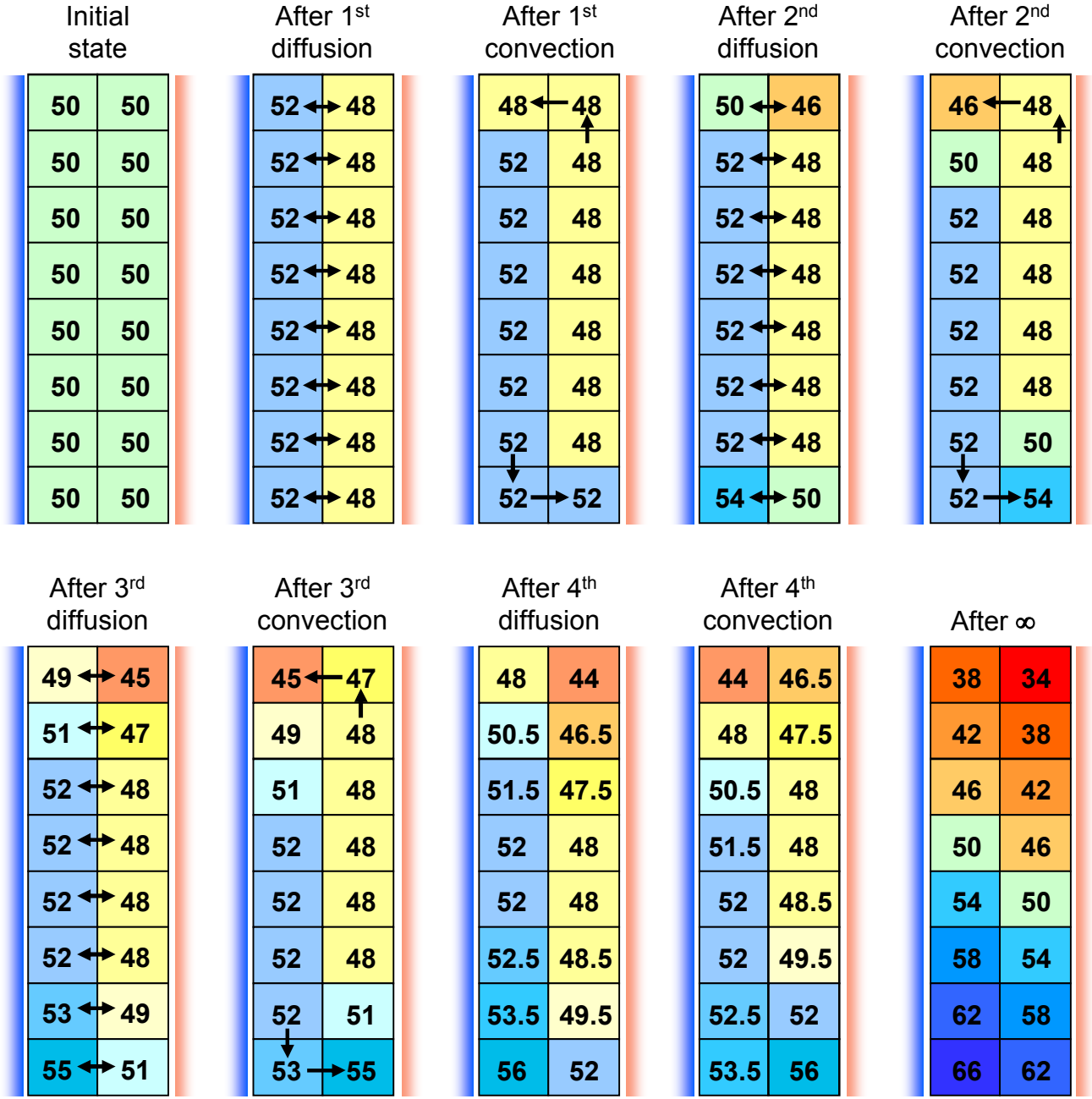


Mechanism of separation:

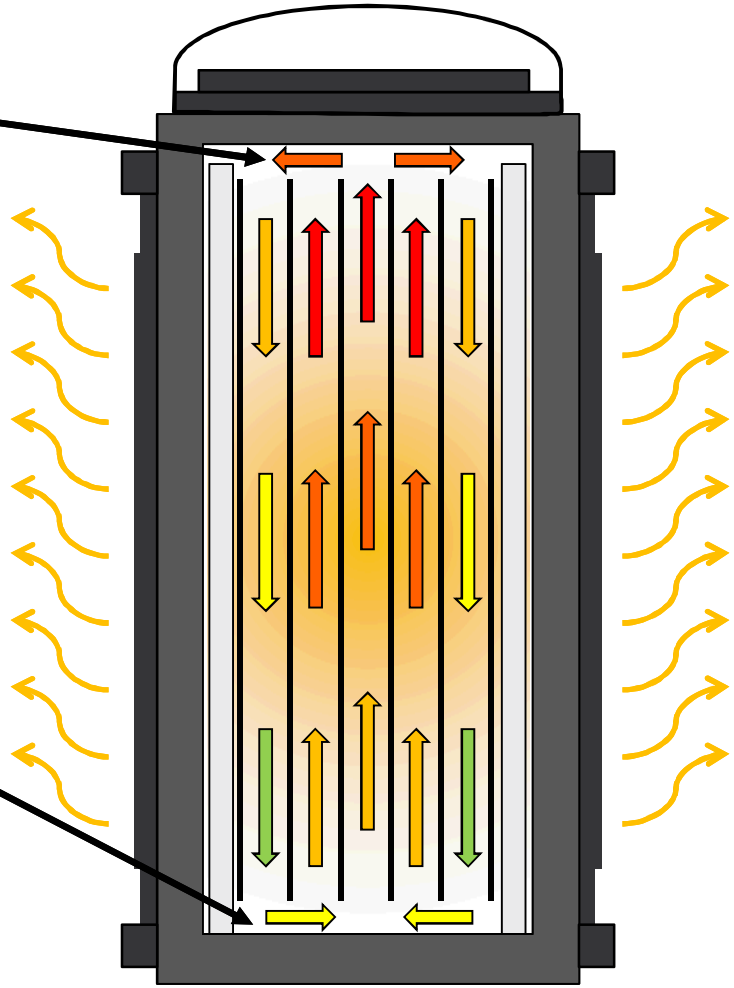
Combined effects of diffusion and convection.

The numbers reflect the concentration of the heavier phase in the mixture.

In this example, the final enrichment is a function of the number of horizontal rows into which the column is discretized. In the real case, there is no discretization, and near-purity can be achieved at the ends of the column.



High-Burnup Demo Cask as a Convecting System



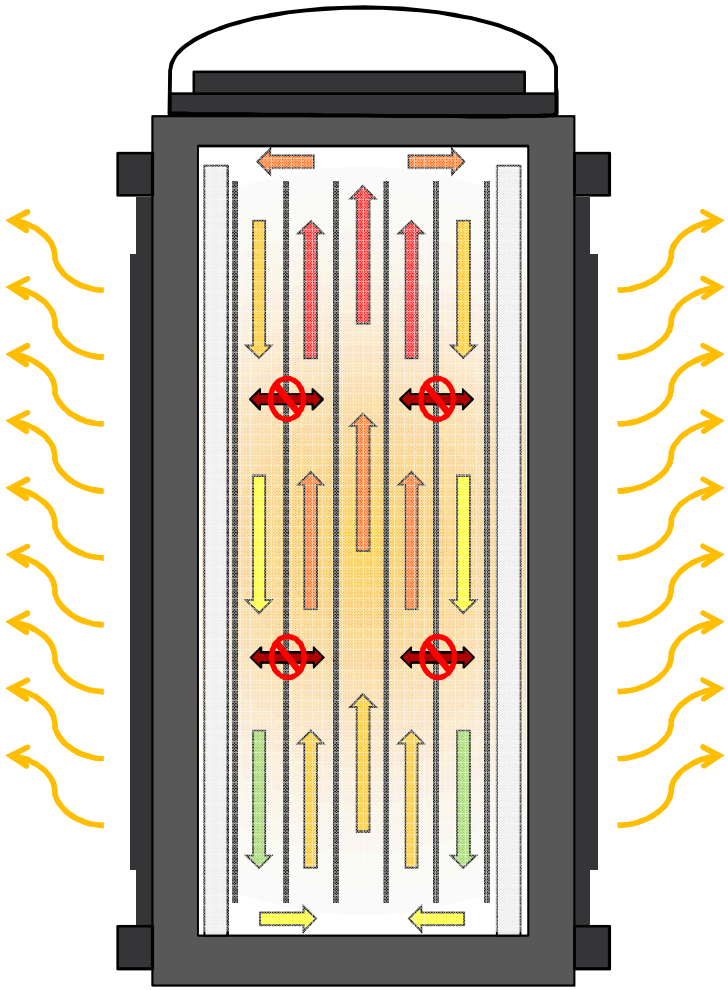
Convecting System?

Looks a little like a Clusius-Dickel column

Will vertical separation of He and Kr occur?

High-Burnup Demo Cask as a Convecting System

Convecting System?



Will vertical separation of He and Kr occur?

Unlikely.

High degrees of separation require the combined effects of:

- convection
- thermal diffusion

Diffusive exchange between the rising and falling limbs of the convection cell **cannot occur**—there are no perforations in the basket partitions. Convection alone will only mix the system.

Conclusions

Factors favoring gas separation by thermal diffusion:

- Large mass difference between He and Kr
- Convecting system, stable for a very, very long time

Factors limiting separation by thermal diffusion:

- Very small concentrations of Kr relative to He inhibit separation
- Relatively small temperature gradients
- Basket partitions do not allow diffusion between rising and falling limbs of the convecting system.
- Complex internal structure may promote turbulence and mixing

Gas separation, if it occurs, will be small—the gas sampling location should not affect ability to detect Kr (assuming that Kr is present in amounts significantly above the detection limit).

Acknowledgements

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.