Argonne
NATIONAL LABORATORY

... for a brighter future

U.S. Department of Energy

THE UNIVERSITY OF CHICAGO

A U.S. Department of Energy laboratory managed by The University of Chicago

Criteria Derived for Geologic Disposal Concepts

Roald A. Wigeland
Manager, Engineering Analysis Department
Nuclear Engineering Division

Contributions from Edgar E. Morris and Theodore H. Bauer, Nuclear Engineering Division

OECD/NEA 9th Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation

September 28, 2006

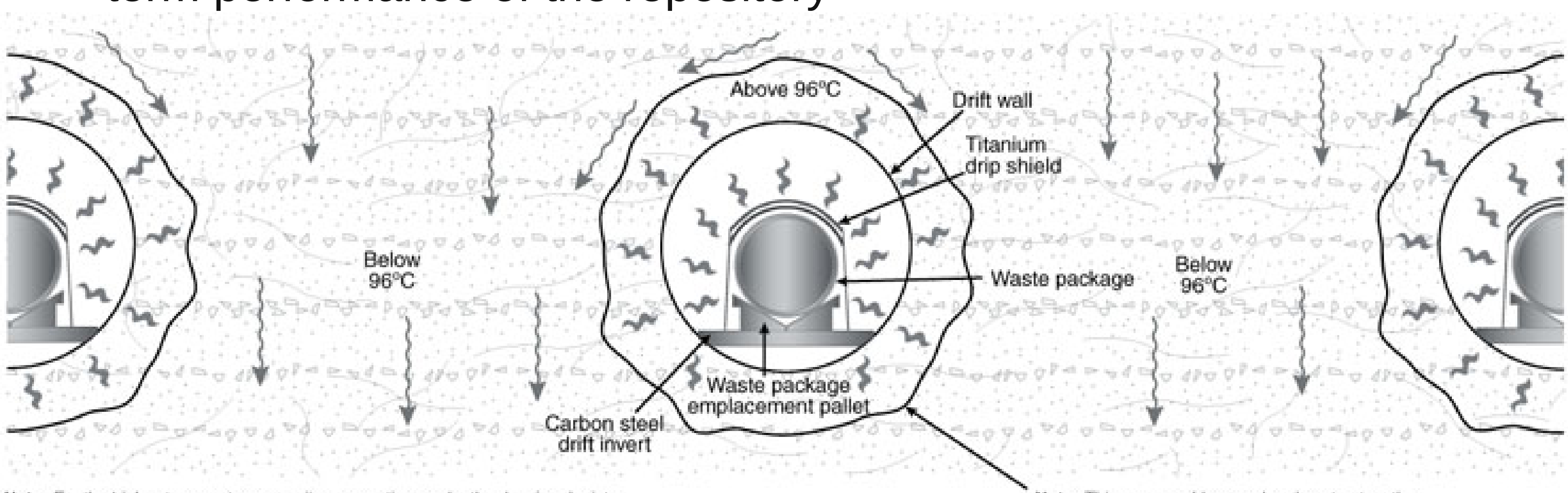
Nimes, France

Geologic Disposal of Hazardous Nuclear Waste

- In the United States, the geologic disposal of spent nuclear fuel and high-level waste is planned for the Yucca Mountain repository
- Disposal of these hazardous nuclear wastes may be constrained by at least three major factors
  - Peak dose rate for repository releases to satisfy regulatory limits
  - Temperature limits for parts of the repository system to provide greater assurance on long-term performance predictions
  - Volume of the waste materials
- For the current planned disposal, the loading of the Yucca Mountain repository is constrained by the temperature limits due to the high decay heat from the spent nuclear fuel
  - Estimates of peak dose rate have been calculated, but there are no regulations in place at this time to judge the acceptability of the planned waste disposal
  - There are spaces between the waste packages in order to limit the linear heat load in the emplacement drifts (tunnels), so waste package volume is not an issue

Current Yucca Mountain Thermal Limitations

- Current temperature limits for the repository design include:
  - Peak temperature below the local boiling point (96 ^{o}C) at all times midway between adjacent drifts
    - *To ensure adequate drainage of water at all times*
  - Peak temperature of the drift wall below 200 ^{o}C at all time
    - *Structural integrity of the repository*
  - Limits are still evolving, in response to concerns related to the long-term performance of the repository



Note: For the higher-temperature repository operating mode, the drawing depicts a period of time after the repository was closed (no further ventilation) for as long as a few thousand years. After a few thousand years, when the waste package heat had decreased considerably, the above-boiling condition would no longer exist and the emplacement heat would be similar to that in the lower-temperature repository operating mode.

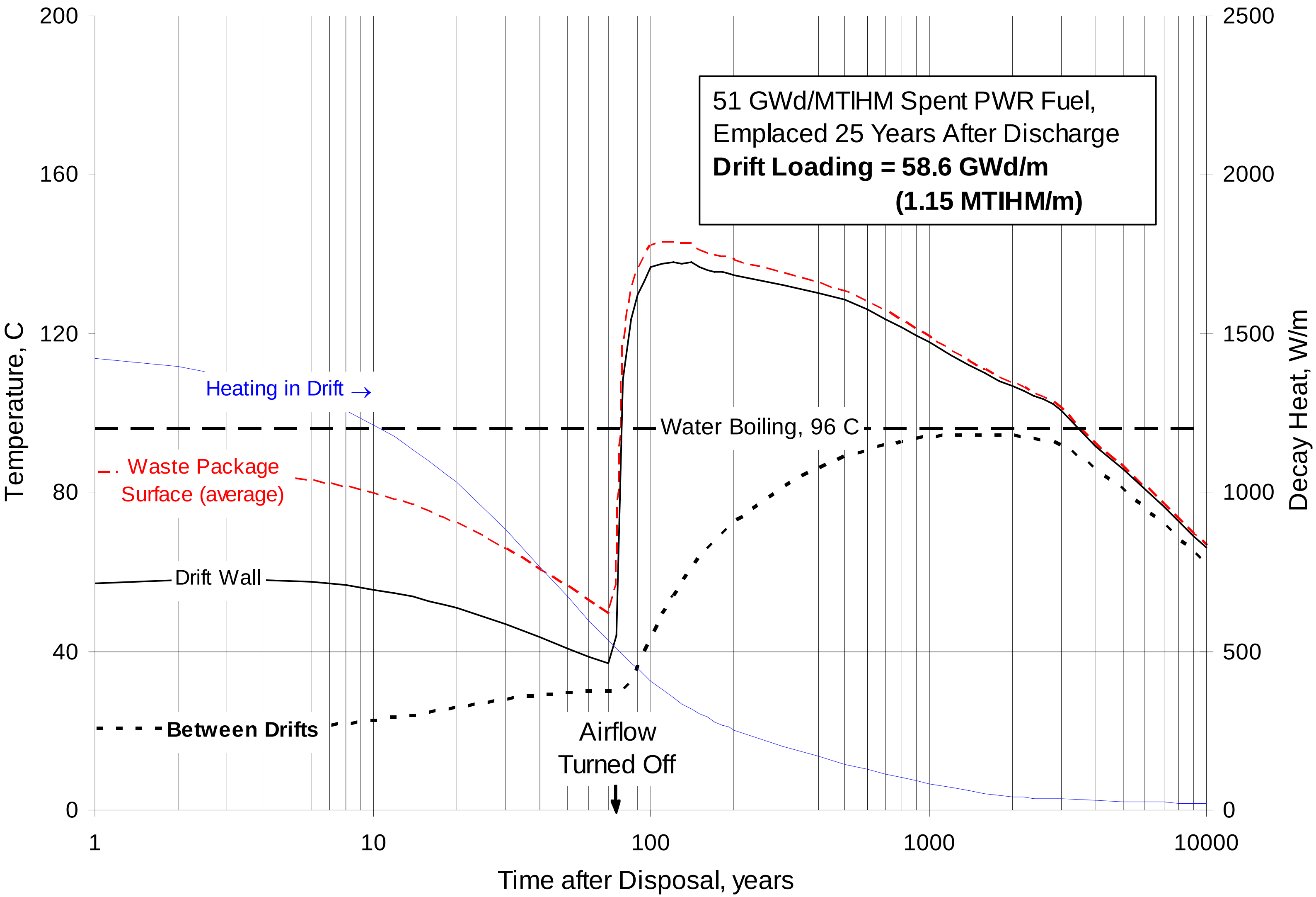
Higher-temperature repository operating mode

Note: This area would expand and contract as the temperature rose and fell over time.

Thermal Characteristics Limiting Repository Usage

- To determine the characteristics of spent nuclear fuel that limit geologic repository usage by exceeding thermal limits, a study examining direct disposal of spent PWR fuel was performed
  - Yucca Mountain is used as the example for a geologic repository
  - 51 GWd/MTIHM discharge burnup
  - Placement in the repository 25 years after discharge
- The response of the repository is modeled as follows
  - A drift near the center of the repository is used since the highest temperatures would be expected in this region
    - *allows a small section of a 3-D model to be used*
  - Forced ventilation for 75 years
- Maximum drift loading is 1.15 MTIHM / m, representing material that produced 58.6 GWd of energy being stored per meter
  - Initial linear heat load is 1.45 kW / m
  - No margin is provided for uncertainties; used as reference point

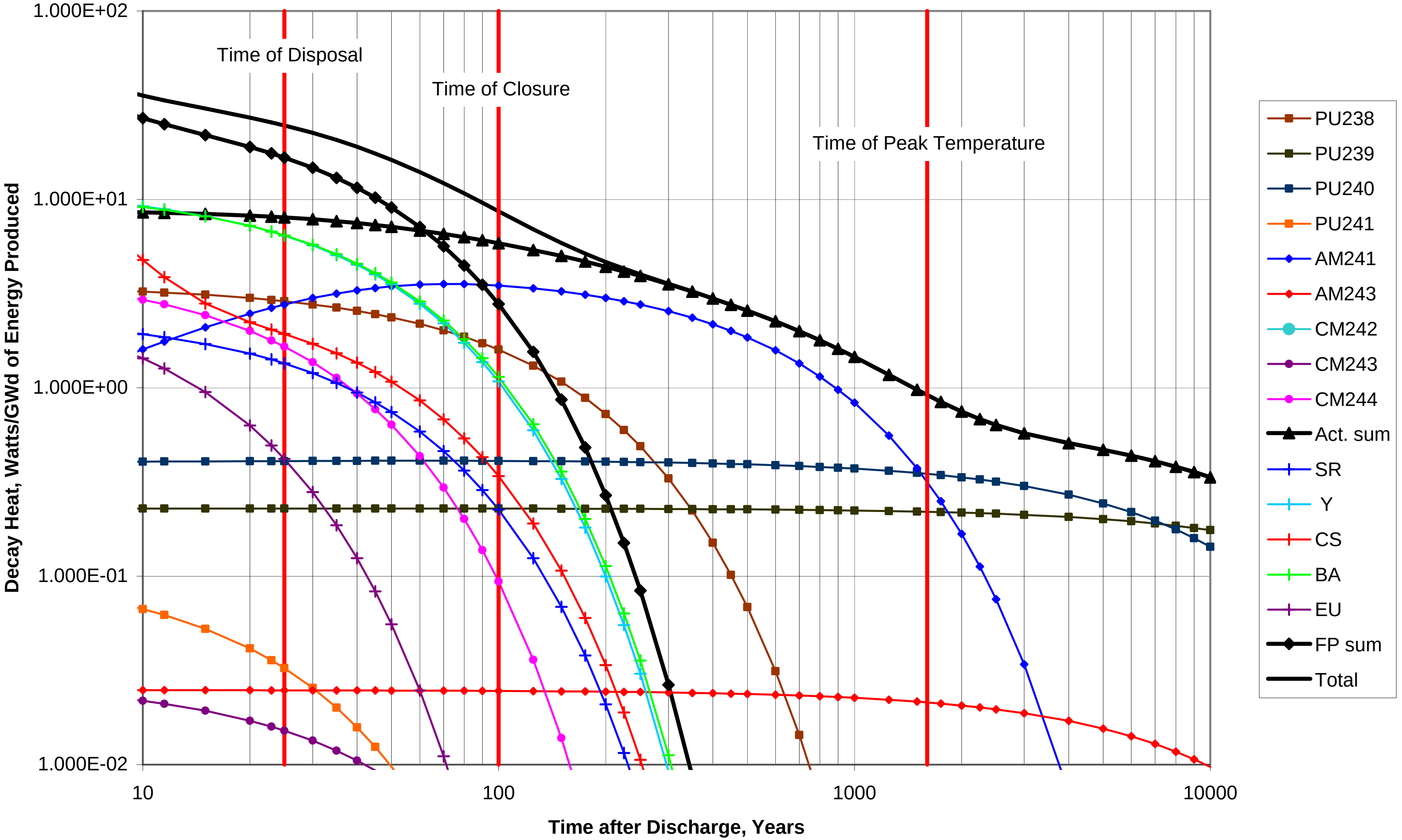
Repository Response to Direct Disposal of Spent Fuel



Repository Loading Limitation for Direct Disposal

- As the figure shows, the loading of the repository is constrained by the temperature limit of 96 ^{o}C for the location midway between adjacent drifts
  - allows water to drain through the repository at all times
- Peak temperature is reached at about 1500 years after disposal. The temperature midway between adjacent drifts only rises substantially after the repository is closed
  - most of the heat to raise the rock temperature is the integrated decay heat from the time of repository closure, 75 years after waste placement (100 years after reactor discharge), to the time of peak temperature at about 1600 years
  - forced ventilation reduces the importance of shorter-lived isotopes
- Examination of the spent fuel decay heat identifies the cause of this decay heat as plutonium and americium
  - Am-241, from the decay of Pu-241
  - Pu-238, existing at discharge, and other plutonium isotopes

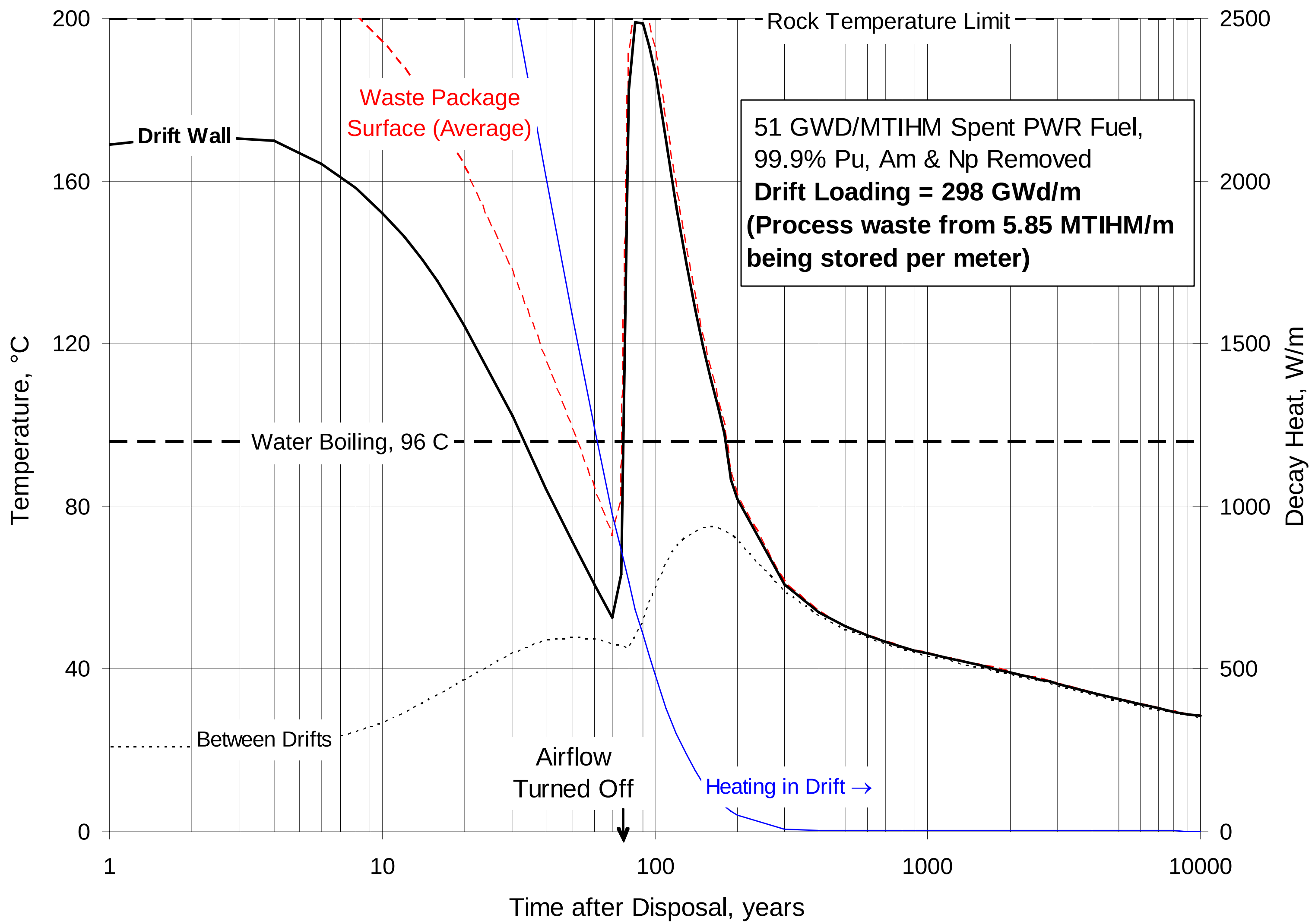
Spent PWR Fuel Decay Heat



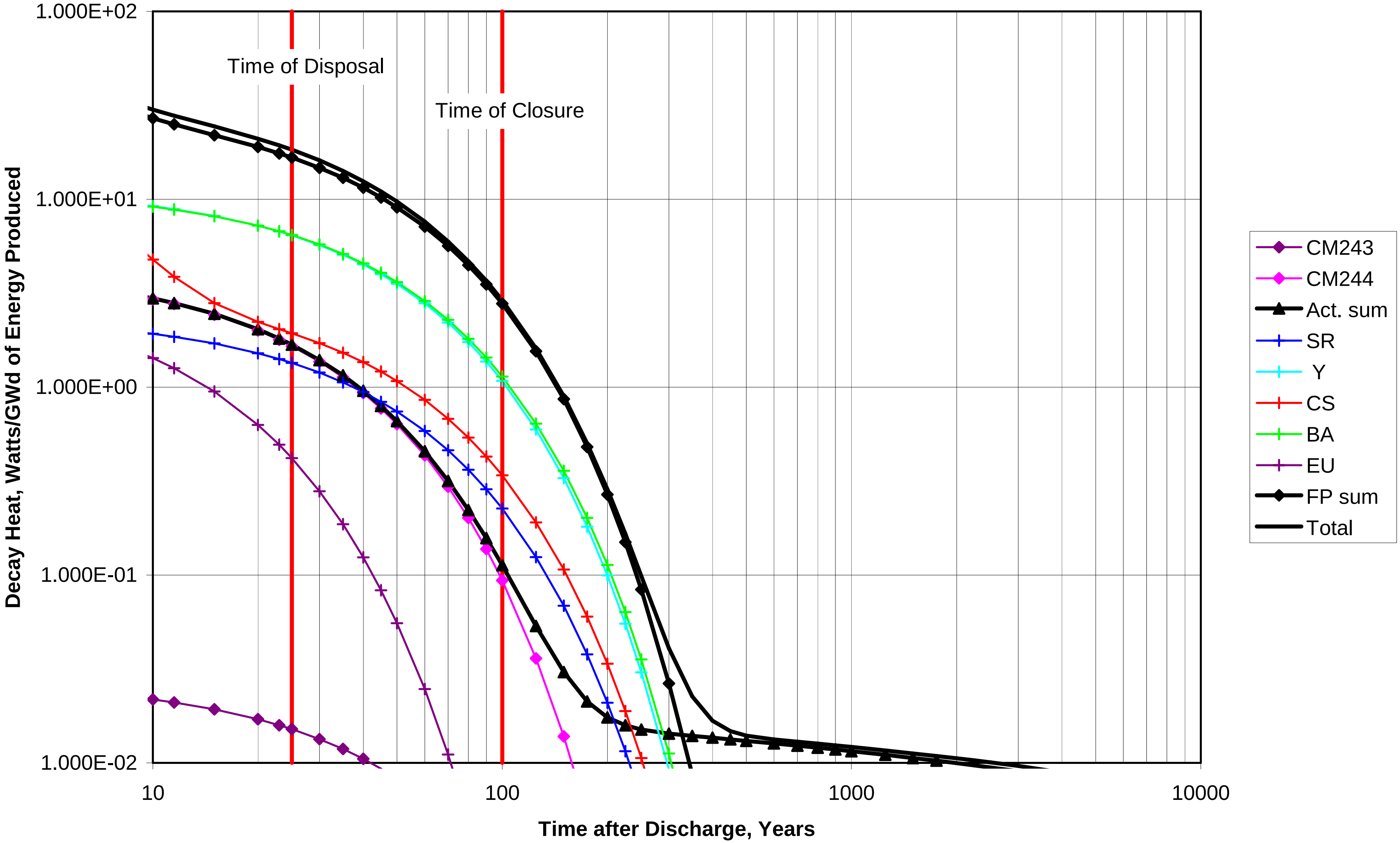
Chemical Elements to Remove from Spent Nuclear Fuel

- With the removal of plutonium and americium, it is possible to more densely load the repository with the resulting process waste
  - lower decay heat from the process waste
  - uranium is also removed to reduce the volume of the process waste
- With these elements removed from the process waste, the limiting temperature occurs much earlier, shortly after the repository is closed
  - ventilation still removes most of the decay heat prior to closure
- Examination of the decay heat for the remaining process waste identifies the cause of this decay heat as barium and yttrium, decay products of cesium and strontium
  - relatively short-lived fission products
  - can be removed from the process waste and placed in separate storage for decay instead of being placed in the repository
- Additional removal of cesium and strontium allows for much larger increases in loading density

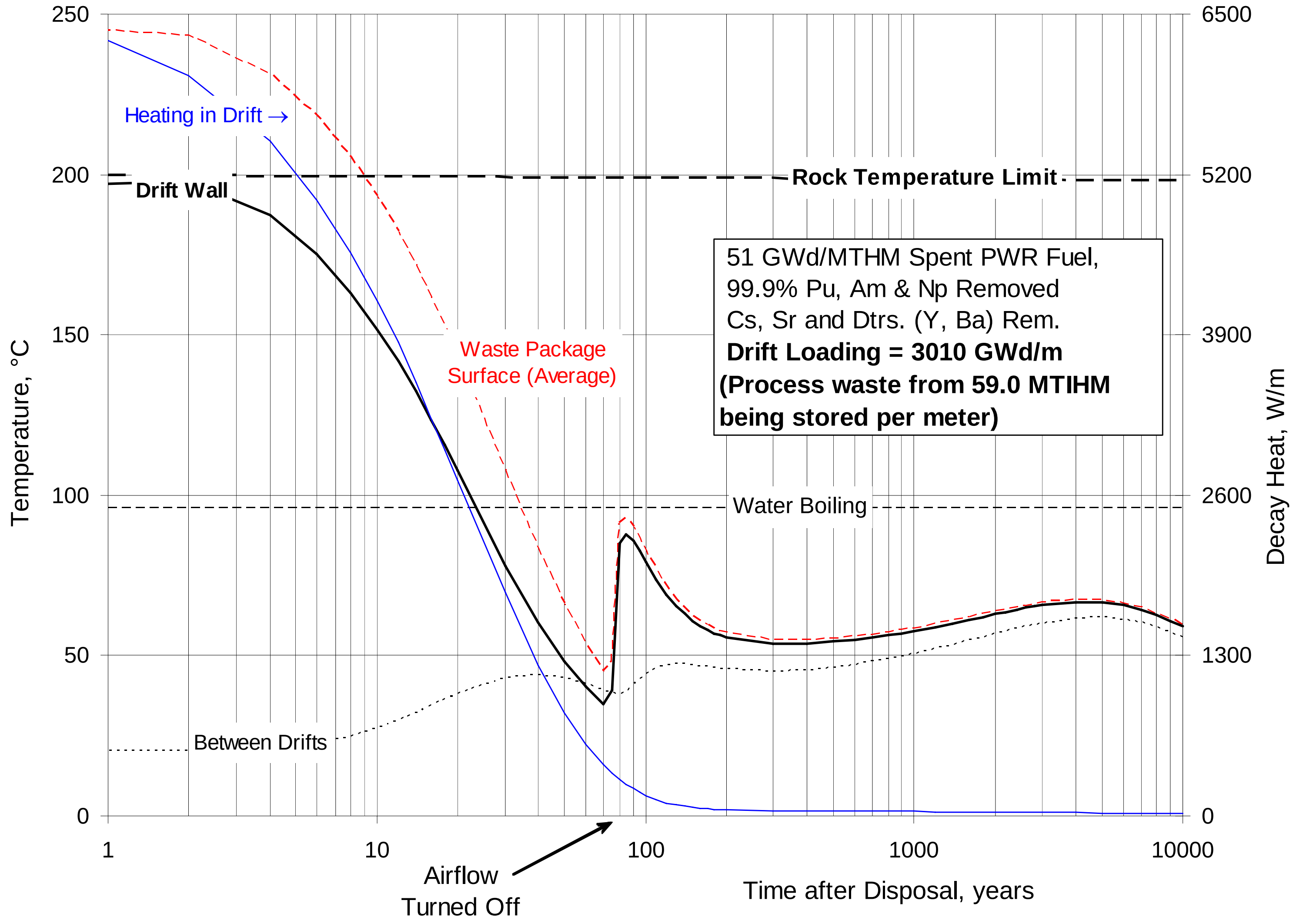
Disposal of Process Waste, Pu & Am Removed



Process Waste Decay Heat w/o Pu & Am

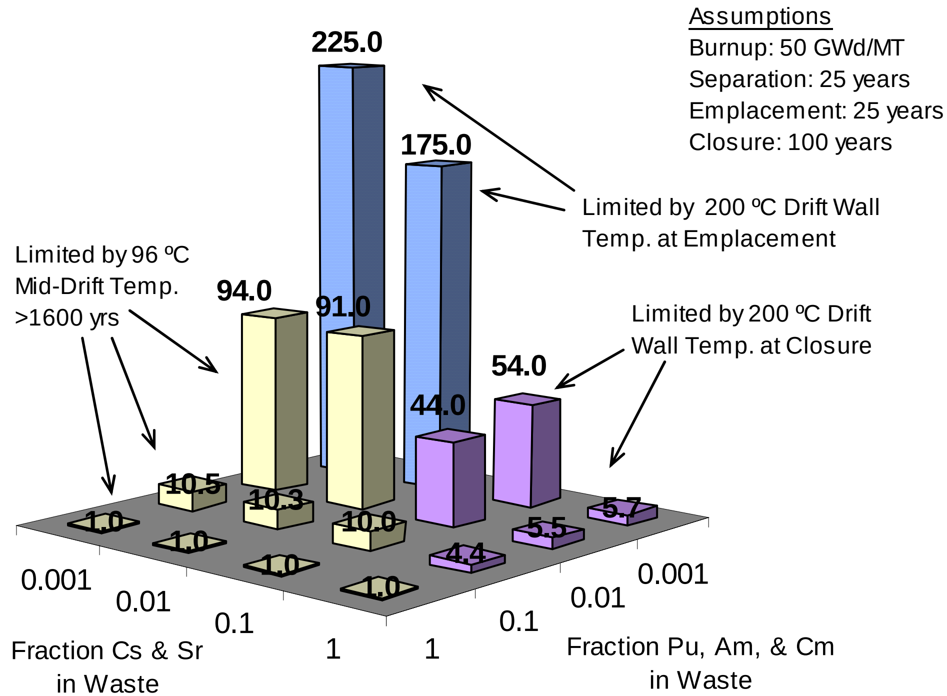


Disposal of Process Waste, Pu, Am, Cs, & Sr Removed



Potential Increase in Utilization of Repository Space

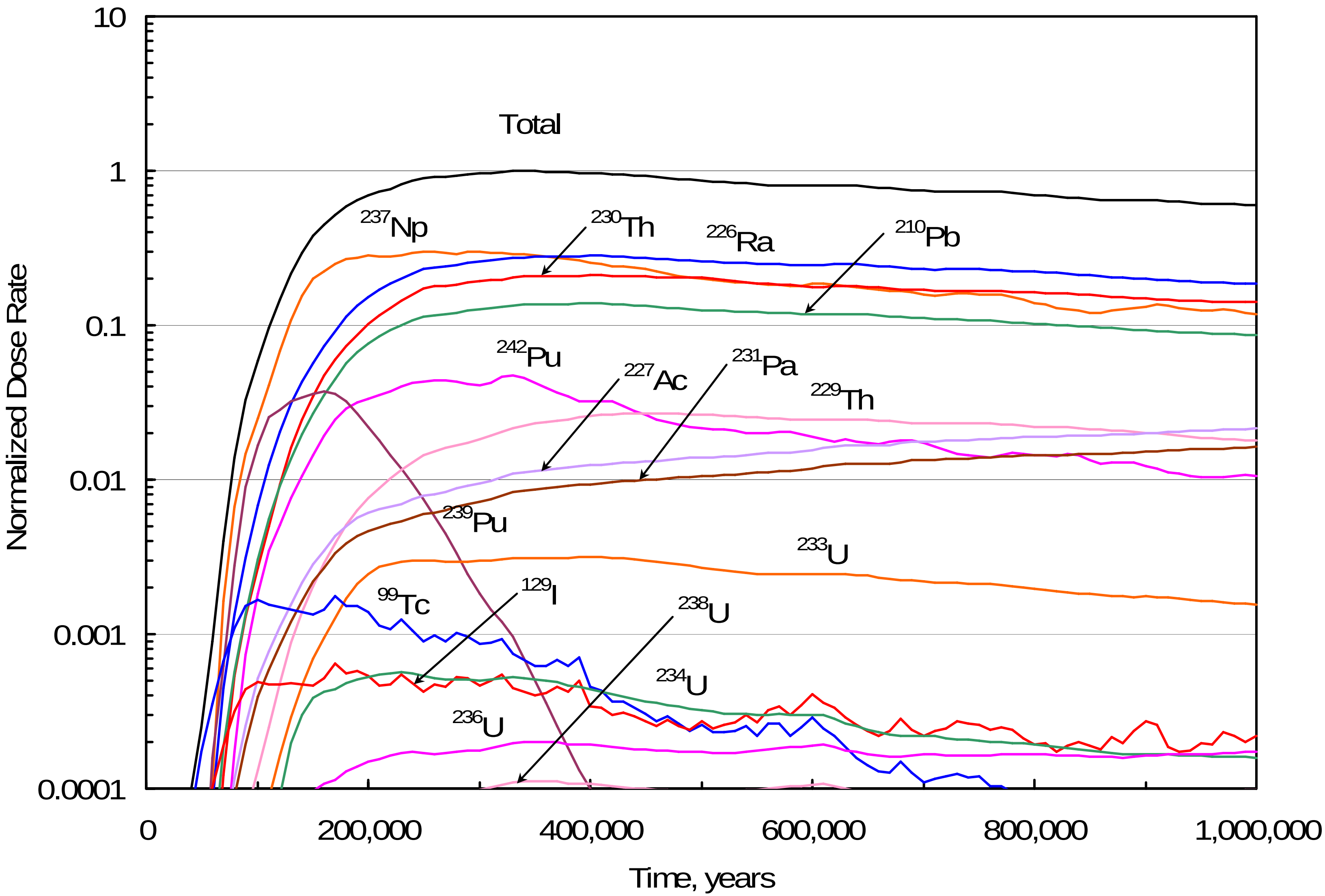
- With the processing of spent PWR fuel to remove the elements responsible for the decay heat that cause temperature limits to be reached, large gains in utilization of repository space are possible
  - The amount of gain is related to separation efficiency
  - Only considers thermal performance, not dose rate
- Pu, Am, Cs, Sr, & Cm are the dominant elements
  - The recovered elements must be treated
    - *separate storage of Cs & Sr for 200-300 years*
- Recycling of Pu, Am, & Cm for transmutation and/or fission
  - Irradiation in reactors
- No direct disposal of any spent fuel



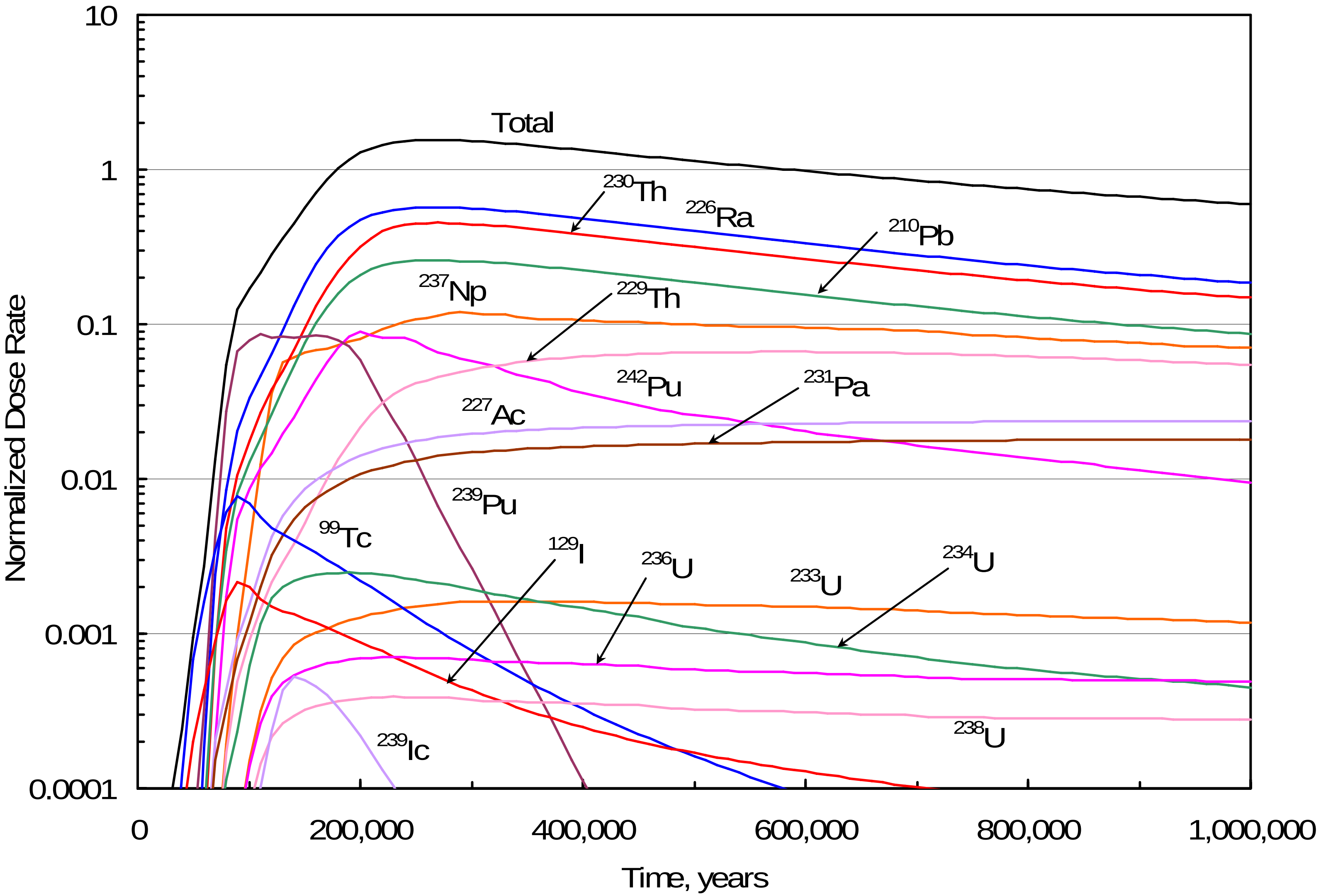
Impact of Processing and Loading Increase on Dose Rate

- The only function of a geologic repository is to isolate the hazardous materials from the public and the environment
  - Actinides are responsible for the peak dose rate in a repository like Yucca Mountain
  - Removal of actinides will decrease the peak dose rate
  - Increasing the loading of the repository will increase the peak dose rate
    - *Inventory of elements not separated from the waste is greatly increased, such as technetium and iodine*
    - *Residual actinide inventory depends on the separation efficiency*
- Peak dose rate is also affected by the waste form
  - Direct disposal of spent fuel with intact cladding
  - Process waste in a vitrified waste form such as borosilicate glass
    - *Chemical conditions as the waste forms degrade affect waste form degradation and the dissolution and mobility of hazardous materials*
- Task is to identify the processing required to lower the dose rate from spent PWR fuel
  - Results normalized to peak dose rate for direct disposal of spent fuel

Direct Disposal of Spent PWR Fuel



Disposal of Spent PWR Fuel Inventory in Glass



Dose Rate Contributors

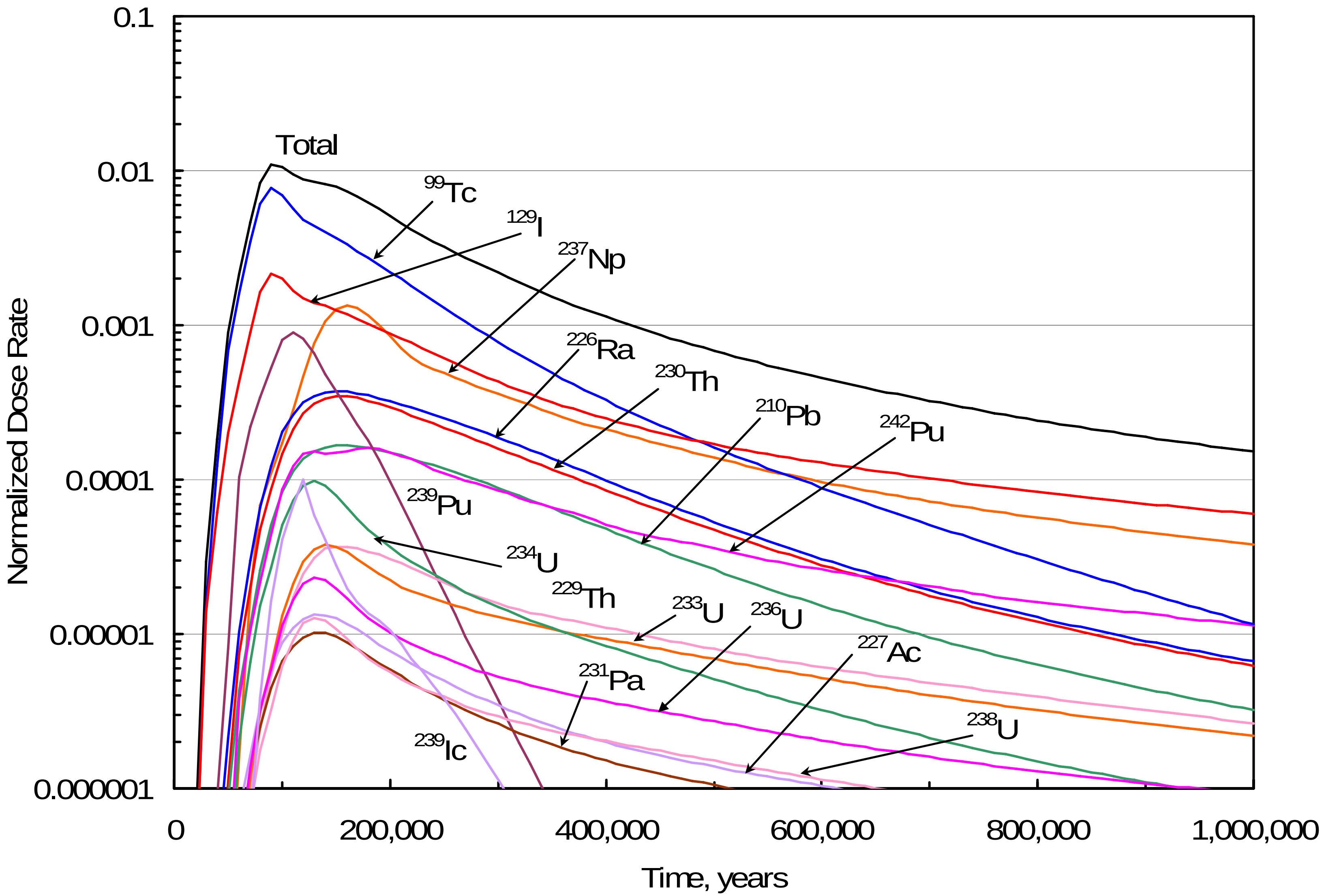
- The main contributors to the dose rate for releases from the repository are actinide isotopes
  - Np-237
    - *Np-237 is present in spent PWR fuel, but additional amounts are created by the decay of Pu-241 and higher actinides*
  - Th-230, Ra-226, and Pb-210
    - *created by the decay of uranium, plutonium, and higher actinides*
    - *238U → 234Th → 234Pa → 234U →* ***230Th*** *→* ***226Ra*** *→ 222Rn → 218Po → 214Pb → 214Bi → 214Po →* ***210Pb*** *→ 210Bi → 210Po → 206Pb*
- The difficulty in reducing the dose rate is that each of the dominant isotopes has more than one parent isotope
  - removal of individual chemical elements is not very effective
  - only group separation of the actinide elements provides large reductions

Dose Rate Reduction from Processing Spent Fuel

Element(s) Removed	Normalized Peak Dose Rate	Peak Dose Rate Reduction Factor*
None	1.550	0.65
Uranium	0.852	1.17
Plutonium	0.835	1.20
Americium	1.473	0.68
Neptunium	1.525	0.66
Uranium &Plutonium	0.199	5.03
Uranium & Americium	0.765	1.31
Uranium & Neptunium	0.823	1.22
Plutonium & Americium	0.749	1.33
Plutonium & Neptunium	0.806	1.24
Americium & Neptunium	1.395	0.72
Uranium, Plutonium & Americium	0.092	10.9
Uranium, Plutonium & Neptunium	0.163	6.13
Uranium, Americium & Neptunium	0.700	1.43
Plutonium, Americium & Neptunium	0.666	1.50
All Actinides	0.011	91.0

*The reduction factor is for the peak dose rate in each case compared to the peak dose rate for the reference case of direct disposal of PWR spent fuel assemblies. Reduction factors less than 1.0 indicate that the peak dose rate is higher than the reference case.

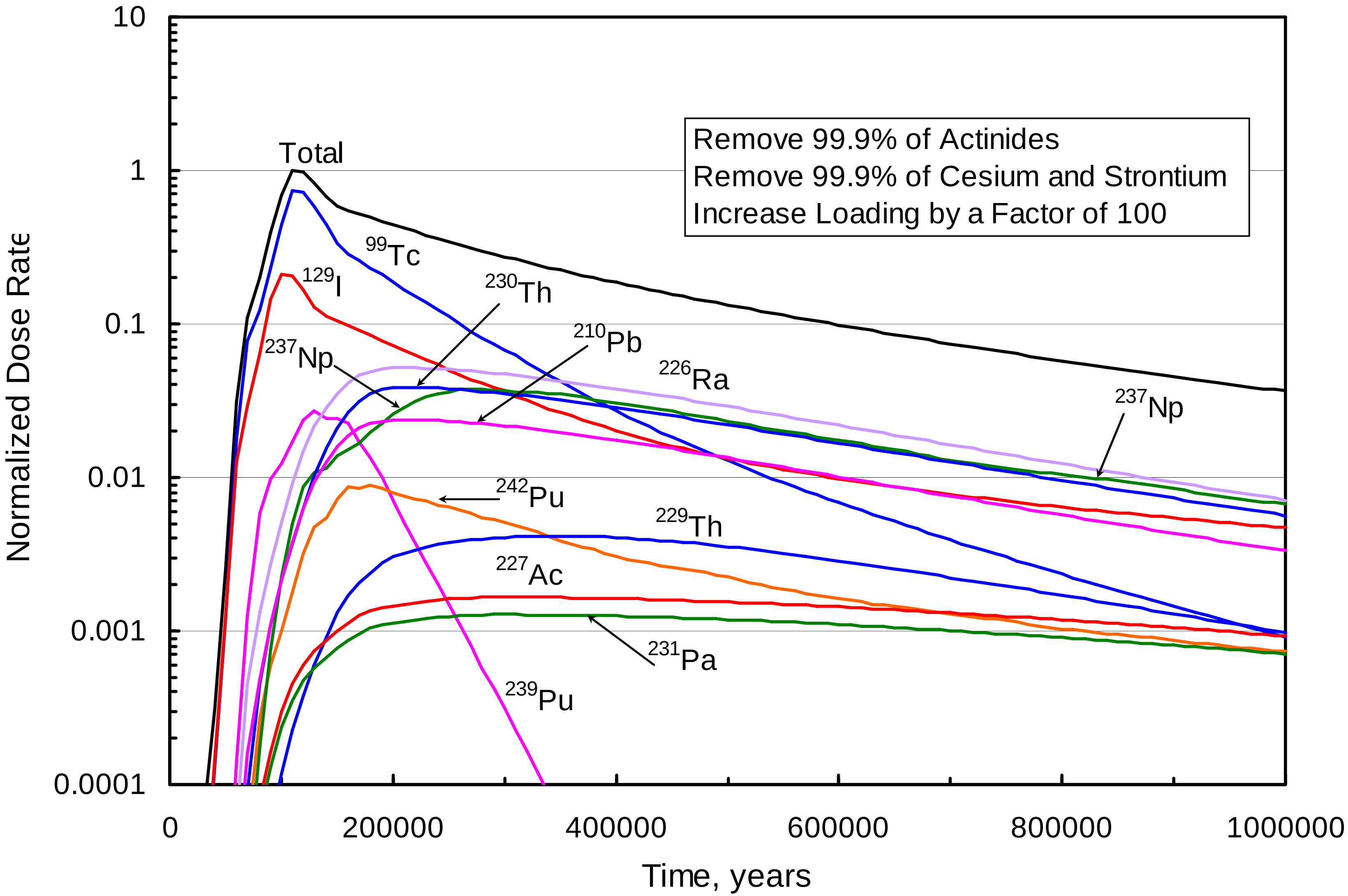
Process Waste with 99.9% Actinide Removal



Waste Management Options

- The use of a geologic repository like Yucca Mountain can be greatly increased by processing spent nuclear fuel to remove all actinides along with Cs & Sr and by recycling the transuranic elements
  - Large increases in the utilization of space by a factor of about 100 can be achieved on a peak dose rate and thermal basis, or
  - Large reductions in peak dose rate (about a factor of 100) are possible with 99.9% removal of all actinide elements
- Three basic options
  - Can be used to greatly lower the peak dose rate associated with a repository intended for a specific amount of spent fuel
  - Can be used to greatly increase the amount of waste placed in the repository without increasing the estimated peak dose rate as compared to the direct disposal of spent fuel
  - Can be used to both reduce peak dose rate (but less than the maximum reduction) and increase the amount of waste (but less than the maximum increase)
- The need for additional repository space or a second repository can be significantly delayed if there is no direct disposal of spent fuel

Example of Increased Repository Utilization



Waste Volume Considerations

- The third major factor in determining repository usage is waste volume
  - Waste forms have chemical loading and temperature limits
    - *to ensure that the waste form can be produced*
    - *to ensure long-term stability of the waste form*
  - At this time, the ability to produce waste forms that would allow the potential densification of the waste products is unknown
- However, waste volume can be addressed by altering repository design
  - If the waste form loading is lower than desired, the waste form will also have lower decay heat generation than would be allowed
    - *Drift spacing can be reduced to allow more drifts in a given repository area (about a factor of 3 for Yucca Mountain)*
    - *Depending on the heat load, multilevel placement can also be considered*
- The overall conclusion is that waste volume should not be a primary determinant for repository usage, and less than optimal waste form loading can be accommodated by increasing waste package loading per unit area, consistent with the applicable peak dose rate and thermal limits

Conclusions

- The use of space in a geologic repository like Yucca Mountain can be greatly improved by processing spent nuclear fuel and recycling the transuranic elements
- The amount of improvement is controlled by the separation efficiencies for spent fuel processing
  - Increasing the drift loading by a factor of about 100 can be achieved while satisfying thermal limits with essentially no change in the estimated peak dose rate as compared to direct disposal of spent fuel
  - waste form volume appears to be a secondary issue, with less than optimal waste form loading compensated by repository design changes while still satisfying peak dose rate and thermal limits
- Continuous recycling of the recovered transuranics is essential
  - no direct disposal of spent fuel
  - transuranics remain in the fuel cycle
- The need for additional repository space or a second repository can be delayed for at least a century, probably much longer