R&D Works on HTR Spent Fuel Treatment in Germany

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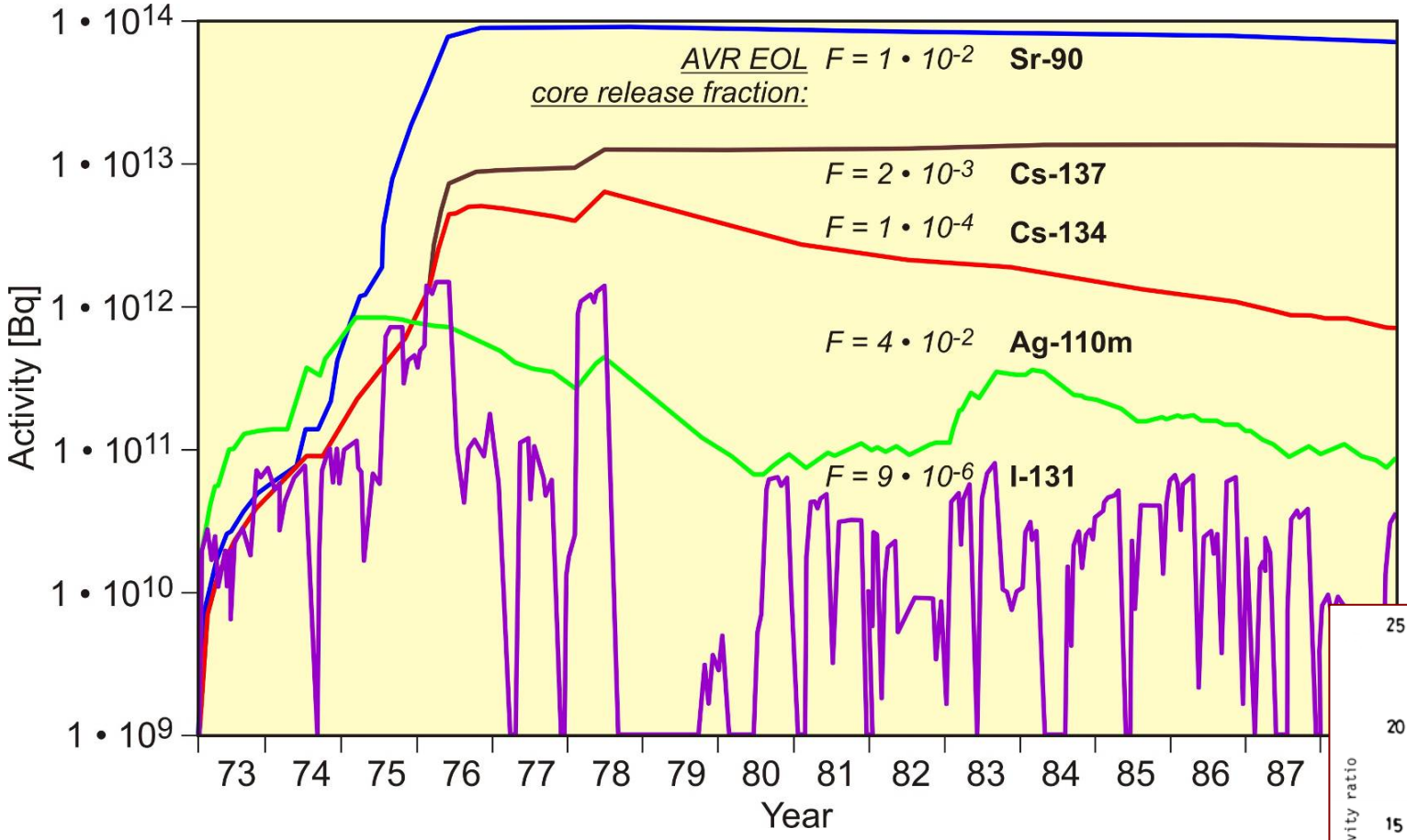
Contents

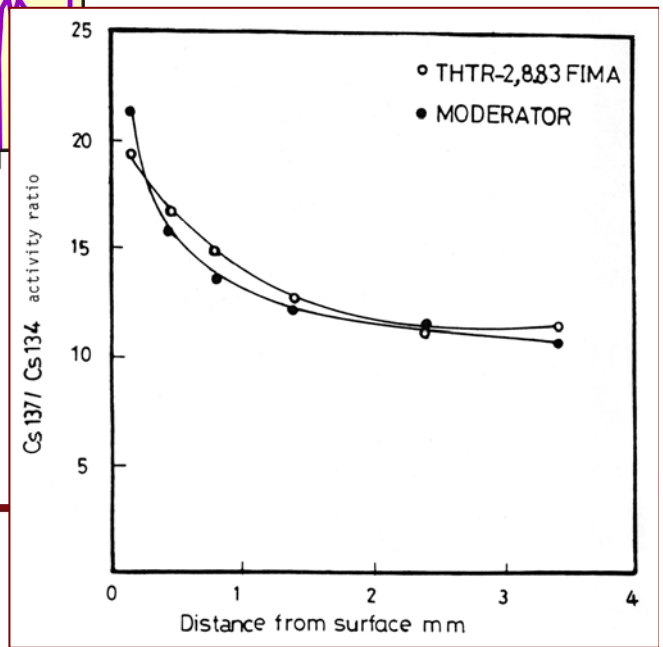
- Introduction
- Activity release into AVR primary circuit and heavy metal inventories of spent fuel spheres
- Activity release from spent fuel containers
- Reprocessing of spent fuel spheres
- Long-term fuel behavior under final repository conditions
- Conclusions

Types of fuel elements and particles in AVR

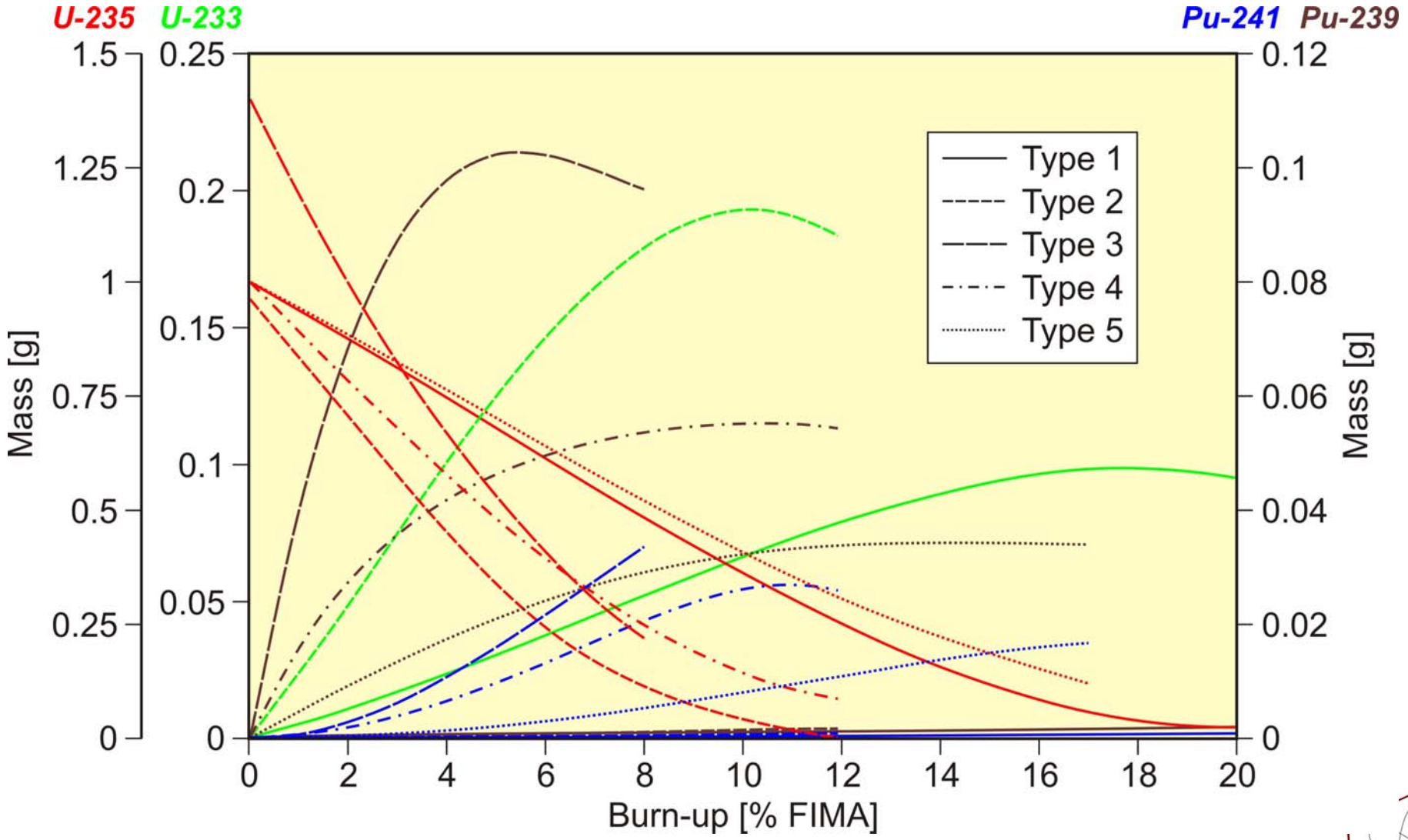
Fuel design:	
HEU mixed carbide	87,600
HEU mixed oxide	129,400
HEU fissile/fertile	20,300
LEU oxide	53,400
Coating design:	
BISO	202,900
TRISO	74,300
Mixed (fissile TRISO, fertile BISO	13,500
Fuel element design:	
Shell type	37,700
Moulded type	253,000
A3-3 matrix material	135,300
A3-27 matrix material	117,700

Activity release into AVR primary circuit





Conversion of AVR core from HEU to LEU



Fissile material inventories measured

	GO-1		GLE-3	GLE-4	
	79/12	**90/28**	**89/22**	**86/20**	**92/03**
Operation time [efpd]	3854	4700	2386	1618	1794
Burnup [% FIMA]	18.5	18.7	9.6	10.6	11.6
Decrease U-235 enrich. [%]	93 → 9-10		9.8 → 1.7	16.7 → ~7	
Nuclide	**Inventories at end-of-life [mg]**				
U-233	95.5	96.2			
U-234	22.41	23.39	4.21	6.30	6.06
U-235	38.2	30.7	157.	368.1	352.6
U-236	146.3	140.6	139.2	107.1	113.8
U-238	59.2	52.1	8790.	4822.	4806.
Σ	361.5	343.0	9090.	5303.	5278.

Contents in total AVR fuel [kg]						
U-233	**U-235**	**U-238**	**Th-232**	**Pu-238**	**Pu-241**	**Pu-total**
19.1	57.	424.	1262.	2.75	0.95	6.81

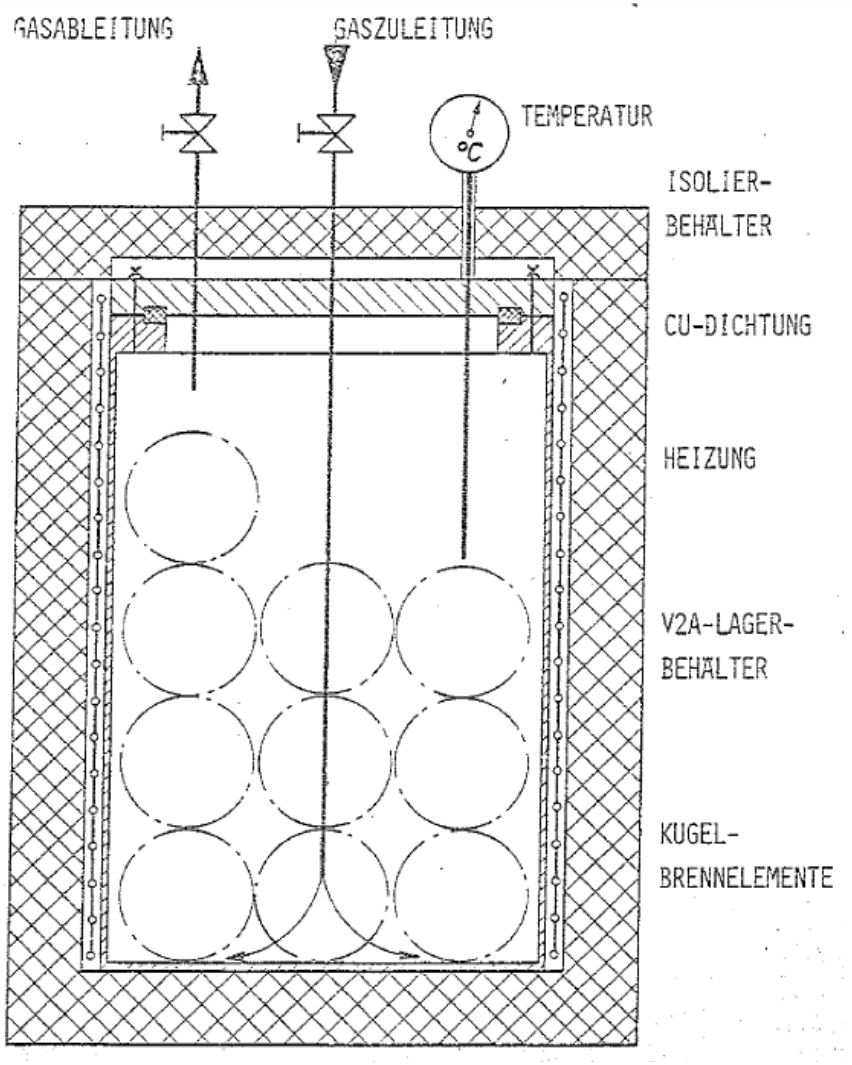
Falta 1993

Radionuclides of interest for interim storage

- H-3 (12 yrs halflife)
  Fission product, B and Li impurities in graphite, He-3
  ~2 GBq (50 mCi) per fuel sphere
  existing as HT, HTO (95%), T_2
- Kr-85 (10.8 yrs halflife)
  Fission product, released from U_{free}
  ~11 GBq (0.3 Ci) per fuel sphere
- C-14 (5730 yrs halflife)
  Natural production in atmosphere at rate 10^6 GBq/yr (27,000 Ci/yr)
  Production from nuclear weapon tests: 2.3×10^8 GBq
  Production in 1000 MWt HTR: 4.3×10^3 GBq (sources: N-14, C-13)
  1 g of C-14 corresponds to 170 GBq (4.6 Ci)
  ~15 MBq C-14 per fuel sphere

Activity measured in dry canister atmosphere

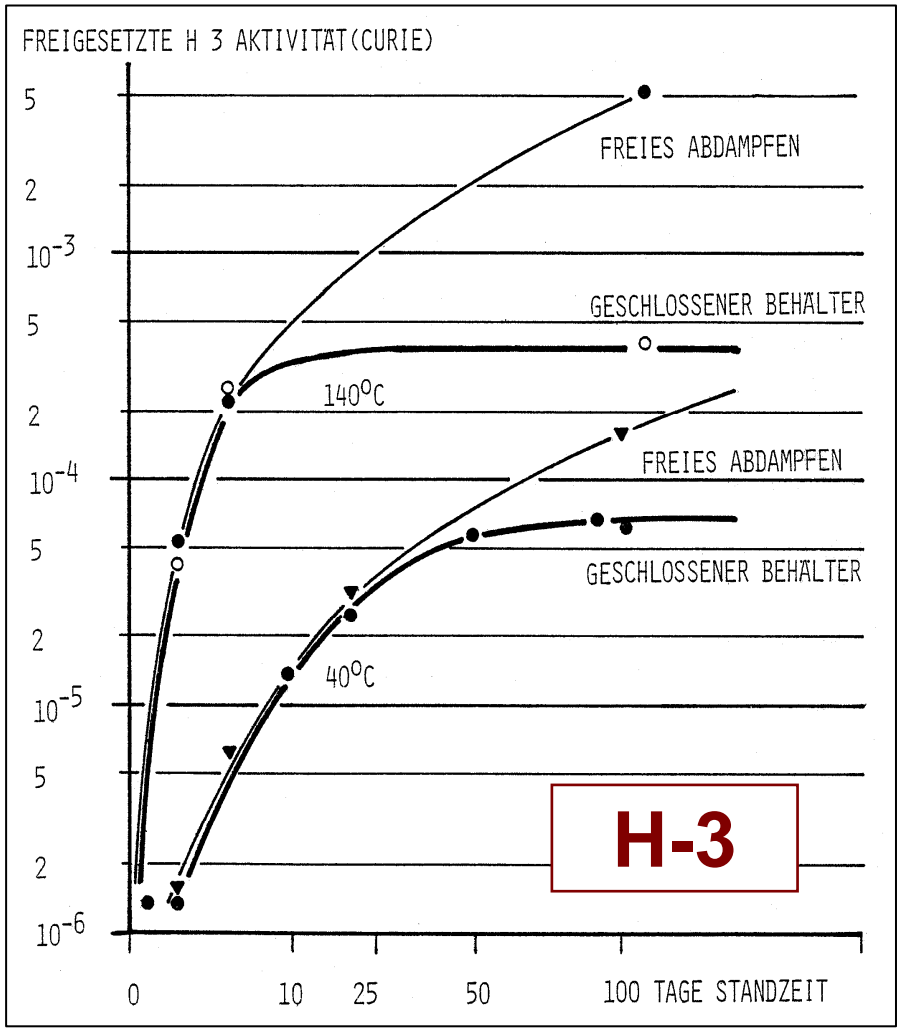
- Activity release from 20 spent fuel spheres measured on two separate canisters (heatable), one canister with oxidic fuel, one with carbidic fuel
- Also use of 5-spheres furnace for higher temperatures
- Cooling period of fuel ~2 yrs

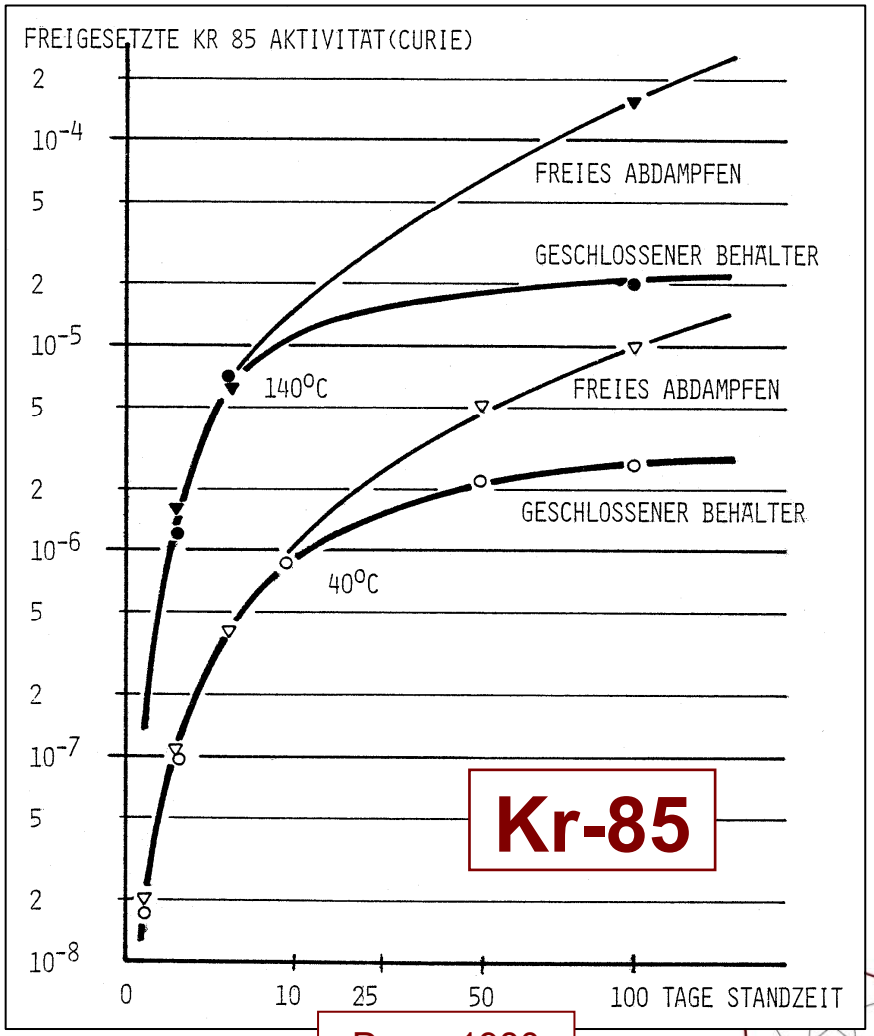


Duwe 1980

Activity release measurements

Release from 20 AVR spheres in closed/open container





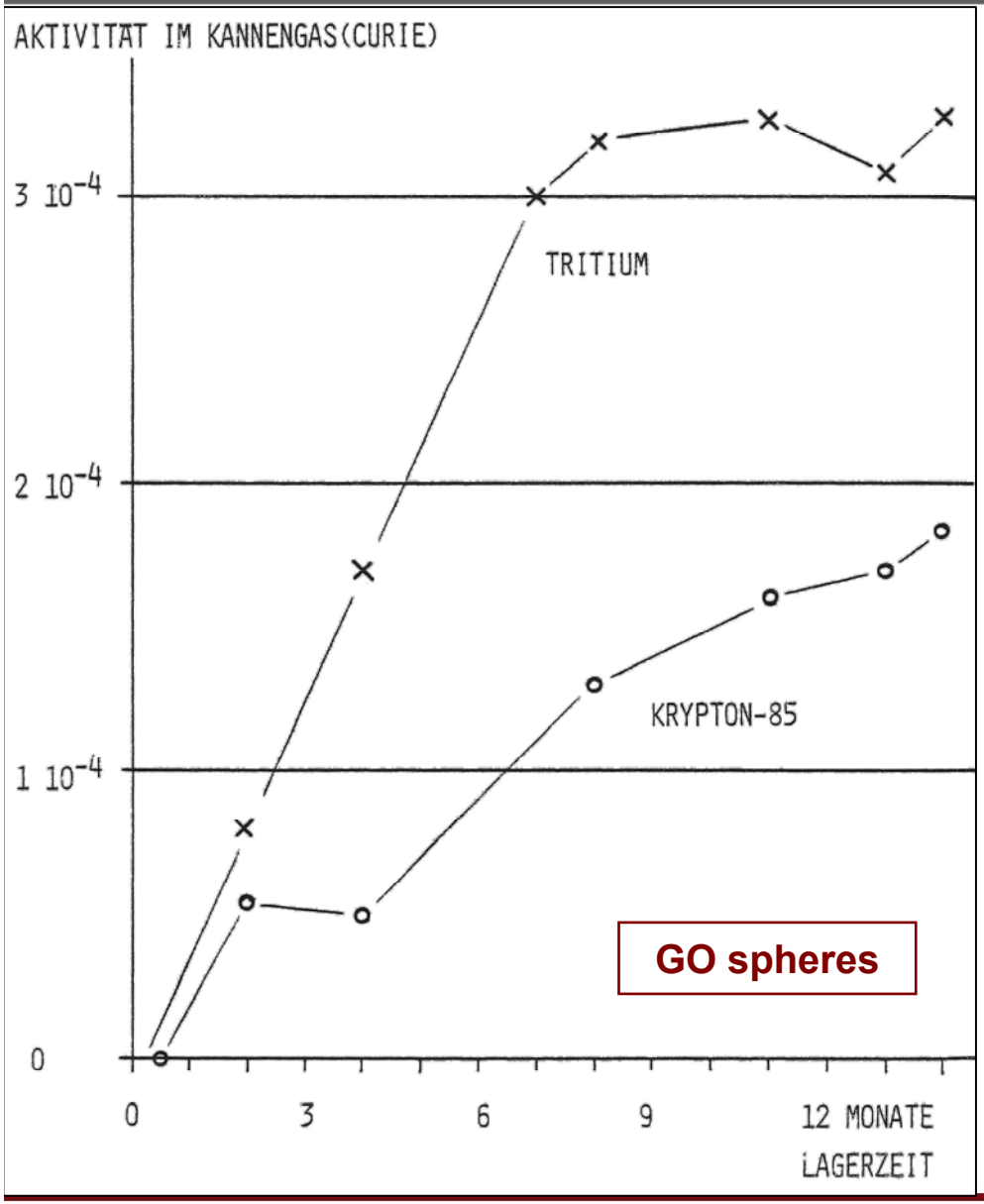
Duwe 1980

Activity measured in canister atmosphere

- Activity release from 950 spent fuel spheres measured on two separate canisters
- one canister with oxidic fuel, one with carbidic fuel
- Cooling period of fuel ~4 yrs (thermal power < 100 W per canister)
- Void volume per canister 113 liters
- 2 valves, 44 thermocouples

Duwe 1984

Activity measured in AVR-TK canister

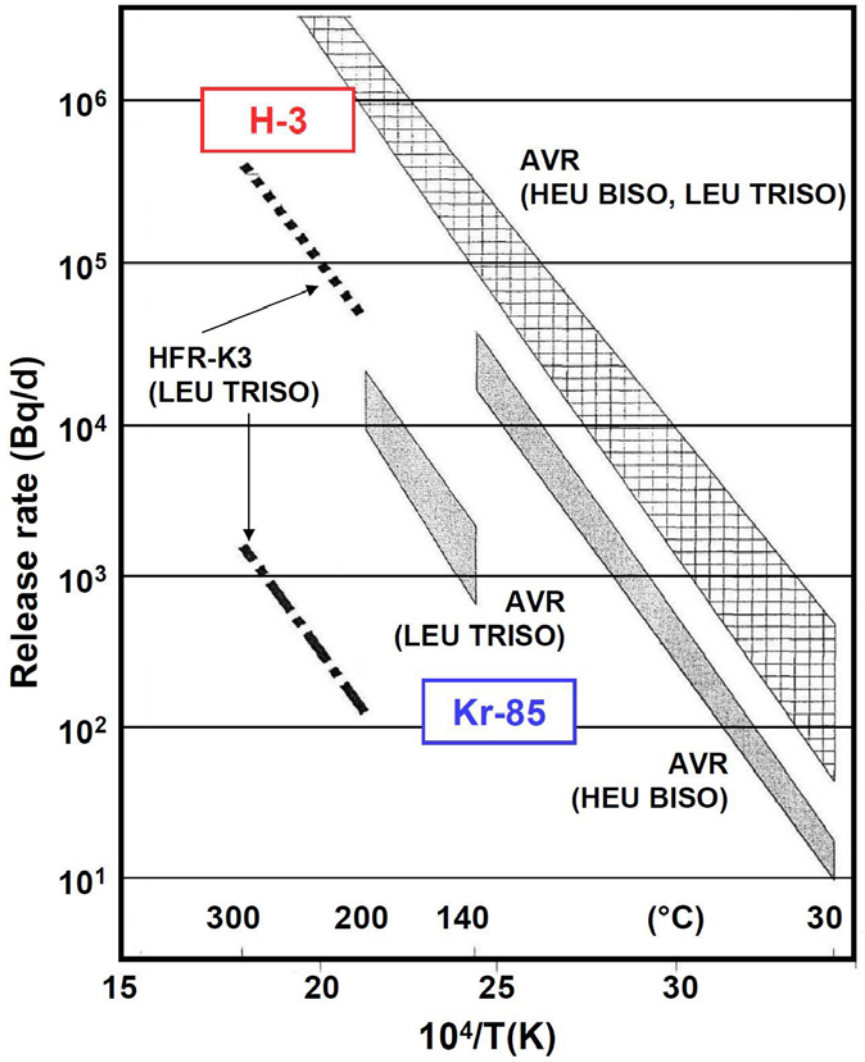


- After ~6 months, H-3 (HTO) release levels out, sorption equilibrium inside canister
- 50 times the H-3 in the canister atmosphere adsorbed in matrix
- Assuming specified leakage rate, 0.6% Kr-85 and 1.4% H-3 are released per yr from canister, compared to 6% decrease by decay

Activity release from spent HTGR spheres

Release into canister gas atmosphere

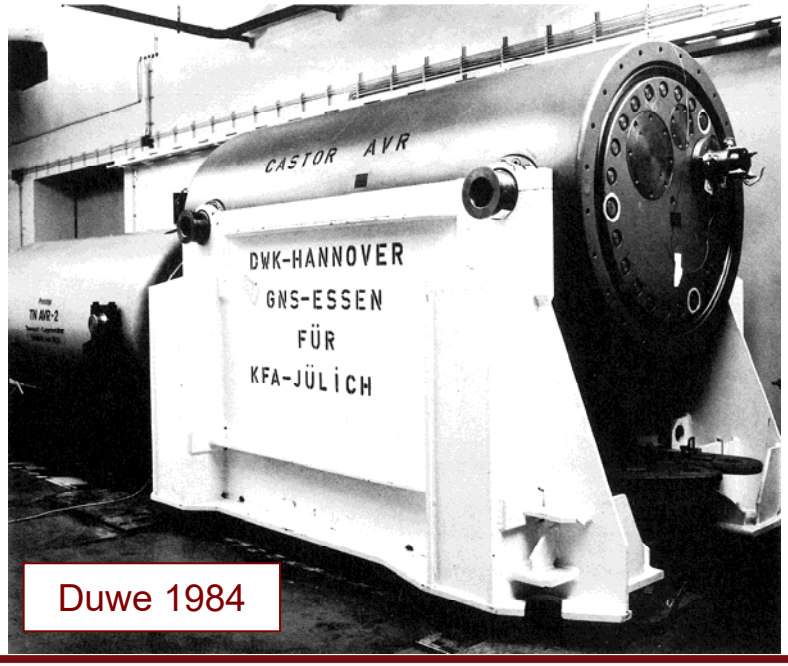
- Summary of measurements for temperature range up to 400°C
- Lower release rates for MTR-irradiated spheres vs. AVR



Duwe 1987

Testing of shielded storage and transport casks

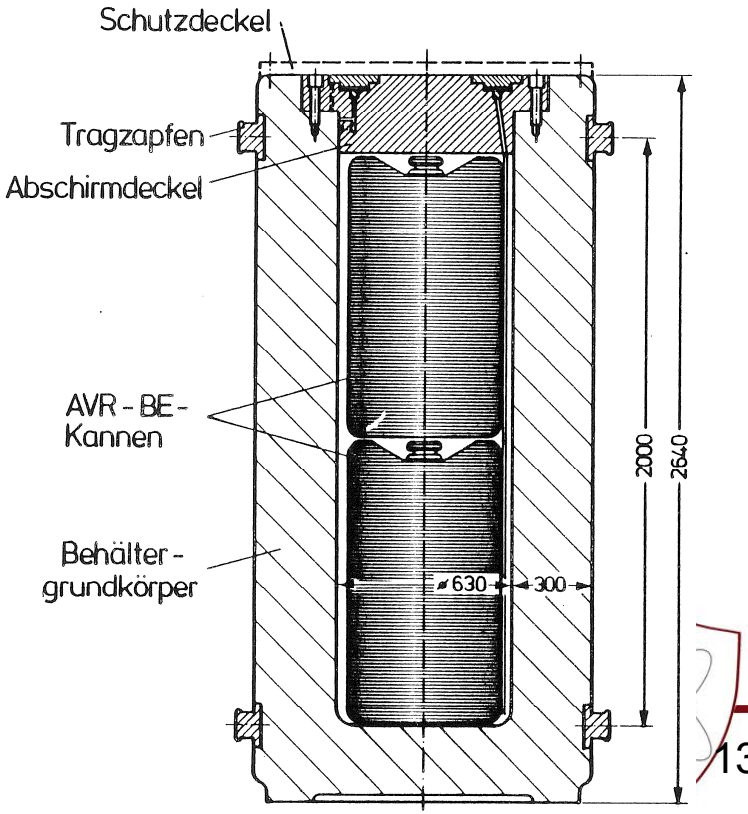
- Demonstration testing of two prototype AVR T/S casks (GNS, TN)
- Each contained two AVR-TK canisters with 950 spheres, one oxide fuel (~13% FIMA), one carbide fuel (~16% FIMA)
- Cooling period of fuel: 405 days
- Neutron and gamma dose power, temperature distribution, activity release to be measured



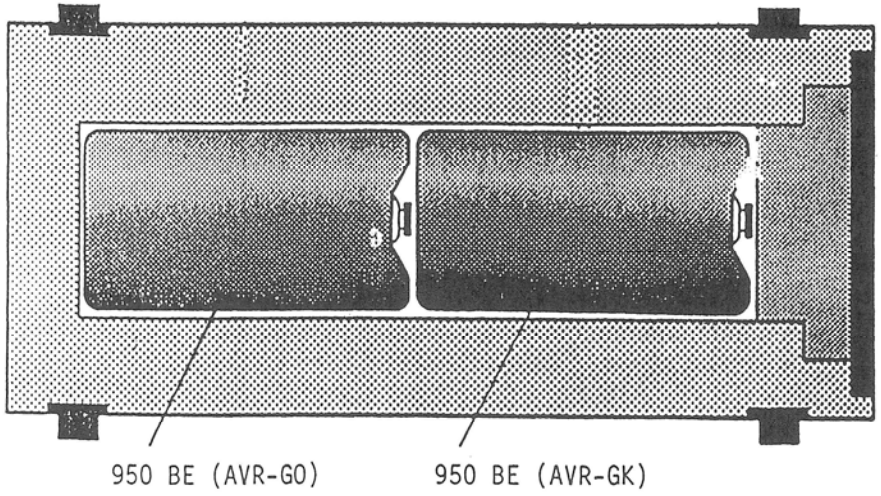
Duwe 1984

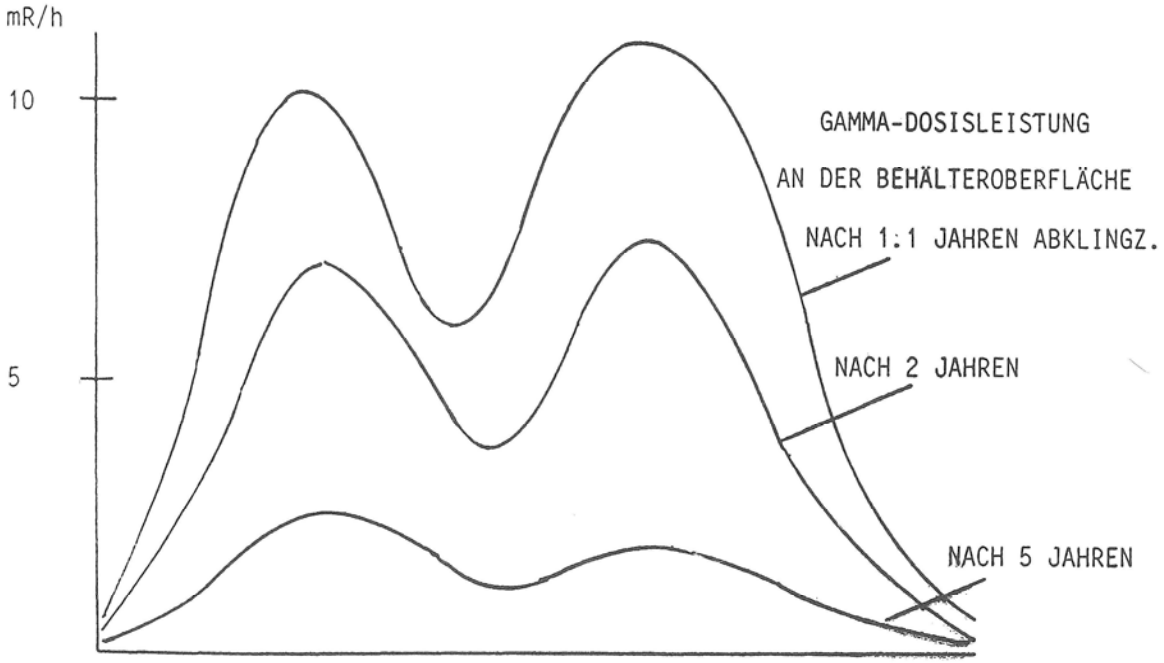
CASTOR cask

Wall thickness:
top=270
bottom=315
side= 300 mm
Weight (empty): 18.25 t



Gamma dose measurements at cask



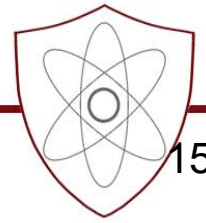


- γ-Dose at cask surface (30 cm shielding): up to 12 mR
- 5 yrs later: up to 2 mR/h
- γ-Dose for carbidic fuel higher because of higher burnup
- Contributors: Ce-144, Cs-134, later Cs-137 dominant

Duwe 1984

(3) Reprocessing of HTR fuel

- All early HTRs based on Th-U fuel cycle
  Breeding of new nuclear fuel was attractive
  Reprocessing required
- Research work at Jülich started in the 1960s
- Construction of semi-technical scale facilities
  JUPITER project in Germany, HET project in the USA
  Cooperation program for mutual benefit



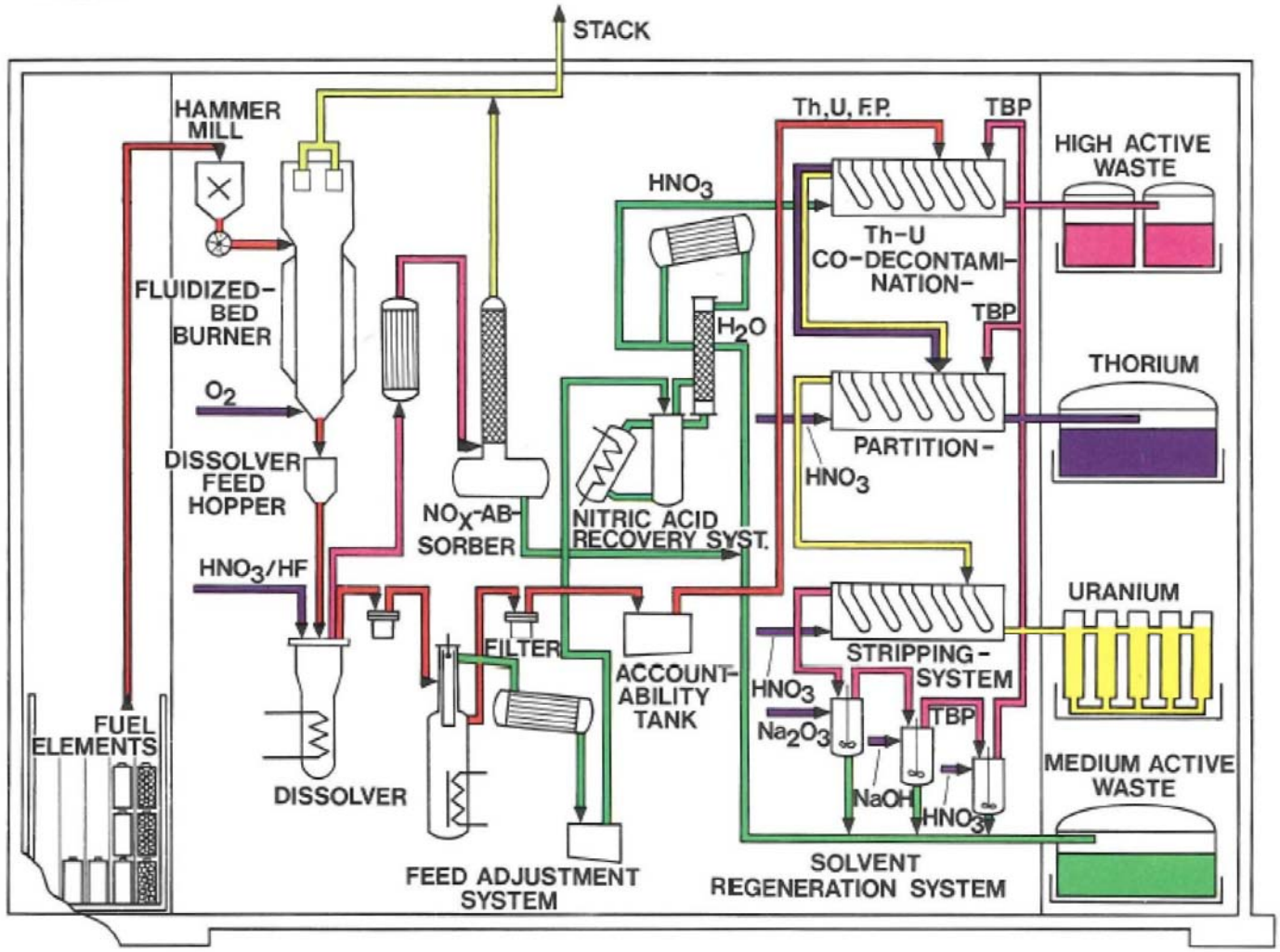
Reference process flow steps of JUPITER

- Reduction of FE to fragments in a hammer mill
  suitable for fluidized-bed burner
- Combustion with oxygen @ 800-850°C
  left-over from BISO: kernels; from TRISO: also SiC coating
- Remaining spent fuel dissolved in acid
  (13M HNO_3, 0.01M $Al(NO_3)_3$, 0.05M HF)
- Resulting nitrid acid solution with Th concentration of 1 mol/l
  filtered, stripped off from acid to be ready for solvent extraction
- Separation of U, Th, fission products by TBP extraction
  (THOREX process)

Designed for 65,000 fuel elements with oxidic fuel and 3yrs operation time

Process flow in JUPITER

JUPITER = Juelich Pilot Plant for Thorium Element Reprocessing

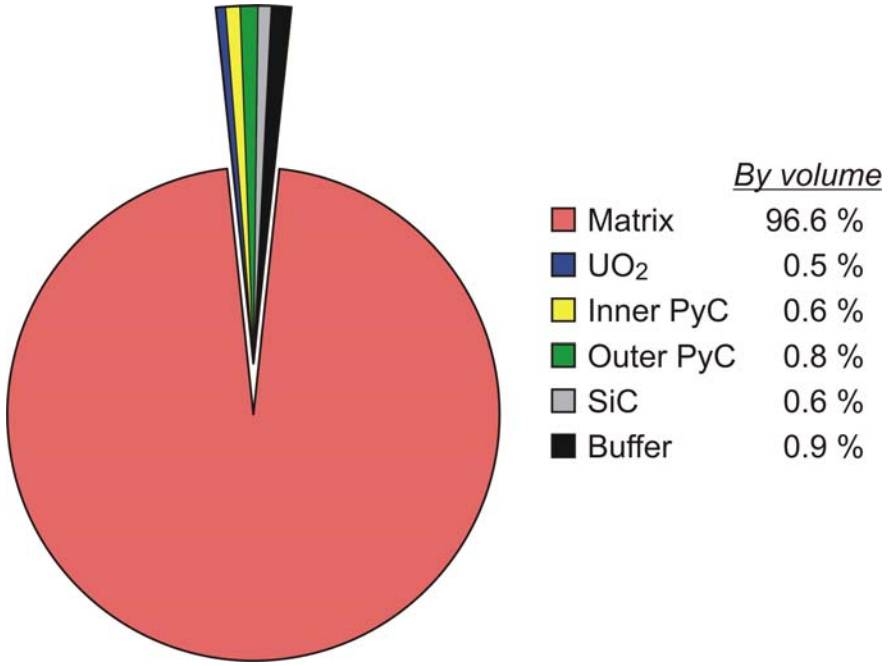


Kaiser 1978

Single steps of reprocessing spent fuel spheres

Head-end

- **Size reduction**
  - separation of graphite and coated particles
  - considered useful (different waste classification)
- **Cracking of SiC coating**



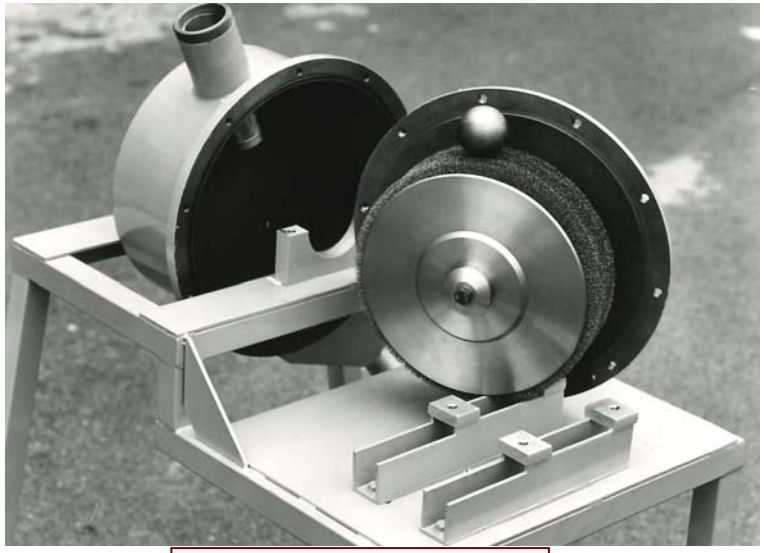
Ratio of HM to matrix:

GO: 6 to 195
THTR: 11 to 193
GLE-1: 20 to 193
GLE-3: 11 to 193
GLE-4: 7 to 193

JUPITER head-end: Incineration of graphite

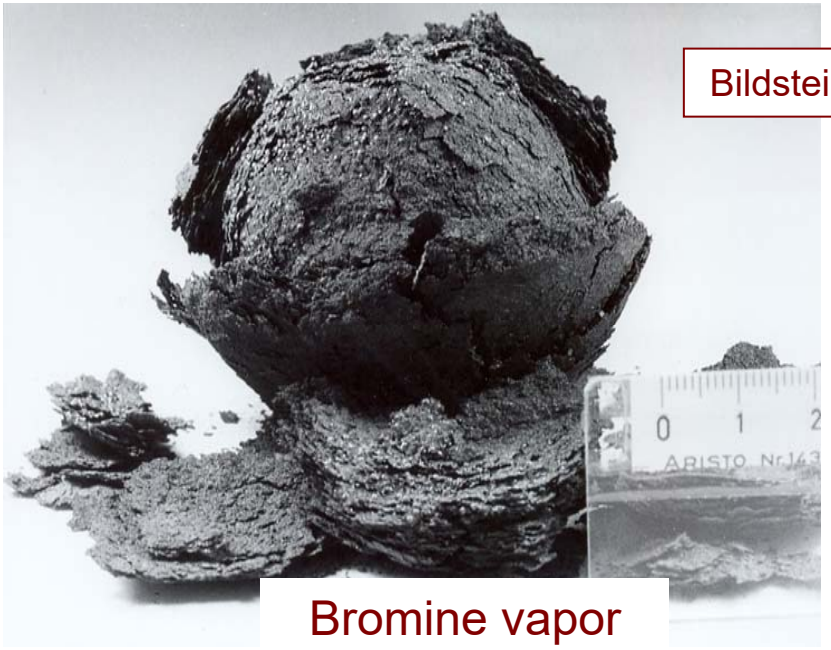
- Combustion unit installed and tested with
  800 graphite spheres
- Demonstration test at FZJ with 11,000 fresh fuel elements with $(U,Th)O_2$ TRISO particles
  Grinding and combusting at a rate of 14 kg/h
  No specific precaution for C-14 release
- In 1981, test with 1500 FE with UO_2 TRISO particles
- Head-end operated with hot-cell technology

Alternative sphere disintegration (1)



Kronschnabel 1981

Brushing device



Bildstein 1966

Bromine vapor

- Destruction of graphite structure after 1 h exposure
- No attack on coated particles (no intercalation in PyC)

Alternative sphere disintegration (2)

Selfrag High voltage discharge method

Separation matrix-particles: 0.4-1.1 kWh

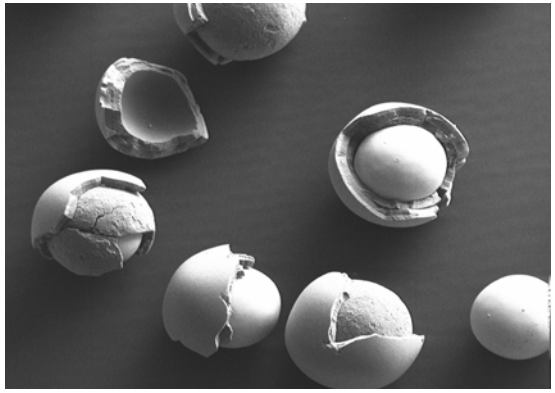
Cracking particles: 0.23 kWh

Energy from one sphere: ~540 kWh

Futterer, JRC, SELFRAC 2010

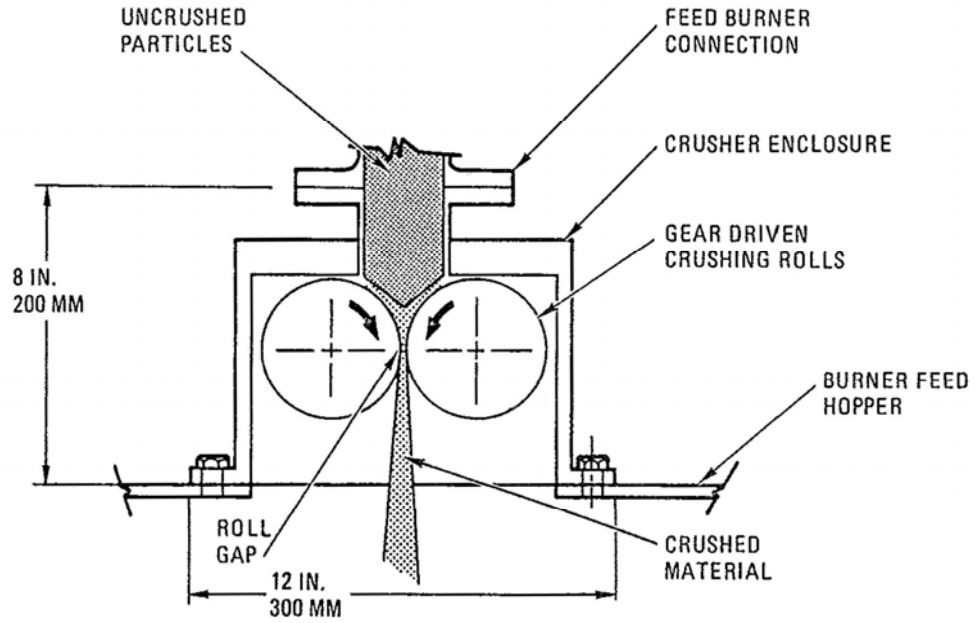
After 3 pulses

300 pulses (1 min)



Cracking of TRISO coating

- **Jet stream method**
  cp in a fluidized-bed accelerated by compressed air jet at 300 m/s and mechanically ruptured when hitting steel plate
- **Diamond disk mill**
  cp pressed into a mill gap by butterfly screw
- **Double-roll crusher**
  cp pressed between two counter-rotating rollers with defined distance and crushed



Lensa 2010

Summary of JUPITER project

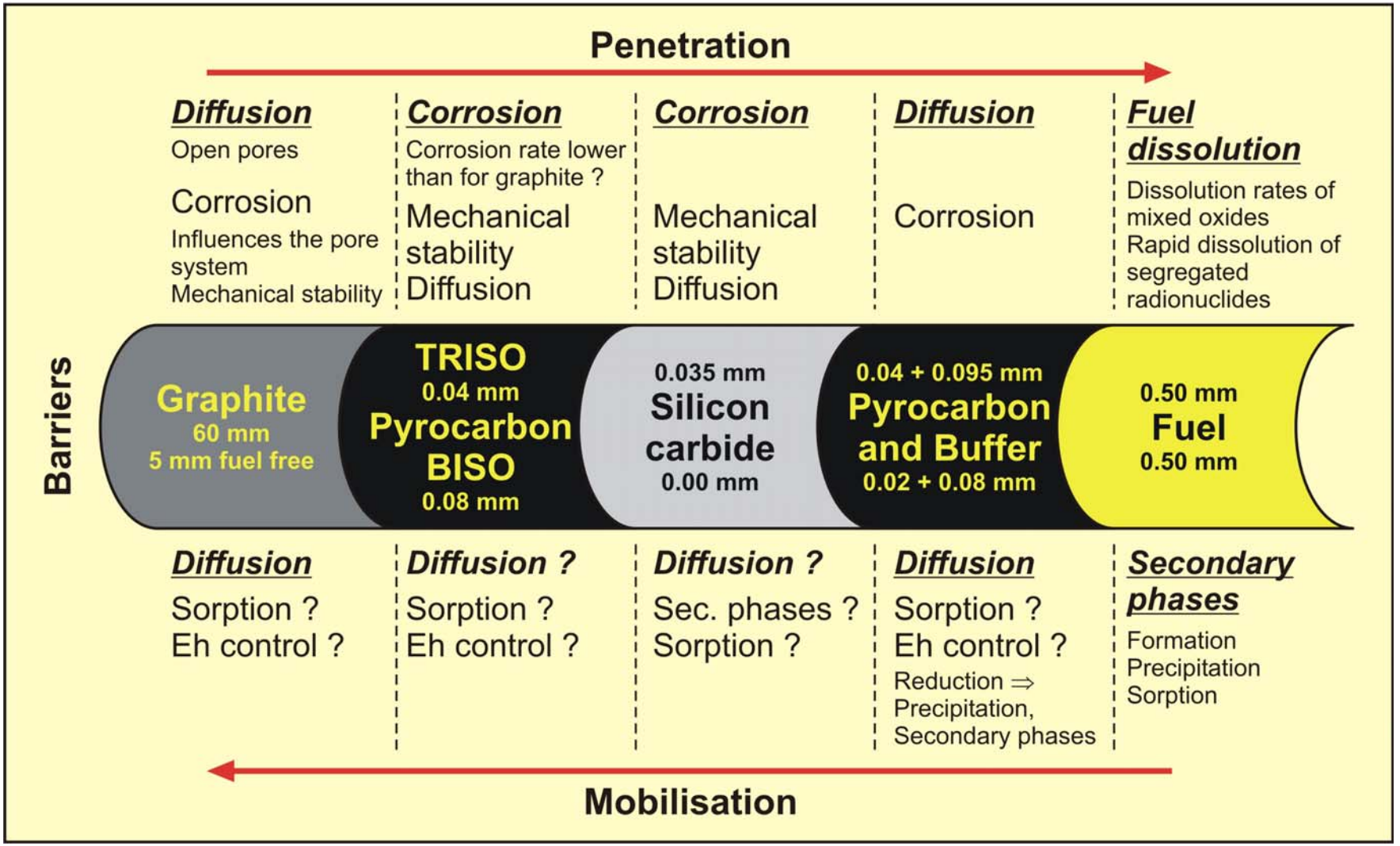
- THOREX process steps developed and demonstrated in cold state incl. material balance and operational analysis
- JUPITER plant was almost ready to start when decision was made to move from HEU to LEU fuel
- After strategy change, hot operation with spent fuel spheres never materialized
- Works continued on head-end only, now also including cracking of TRISO coated particles

Efforts finally abandoned in 1985

(4) R&D works on HTR spent fuel spheres

- Investigation of barrier functions of spent fuel during long-term storage (leaching in different environments)
- Mechanical and chemical behavior under realistic repository conditions (corrosion in salt brine, mechanical interaction with converging rock)
  to develop leach-resistant matrices (SiC)
- Treatment and disposal of contaminated graphite
- Manufacture and testing of transmutation fuel, conservation of knowhow on thorium fuels

Separate effect investigations on spent fuel



Fachinger 2006

Brine leaching tests

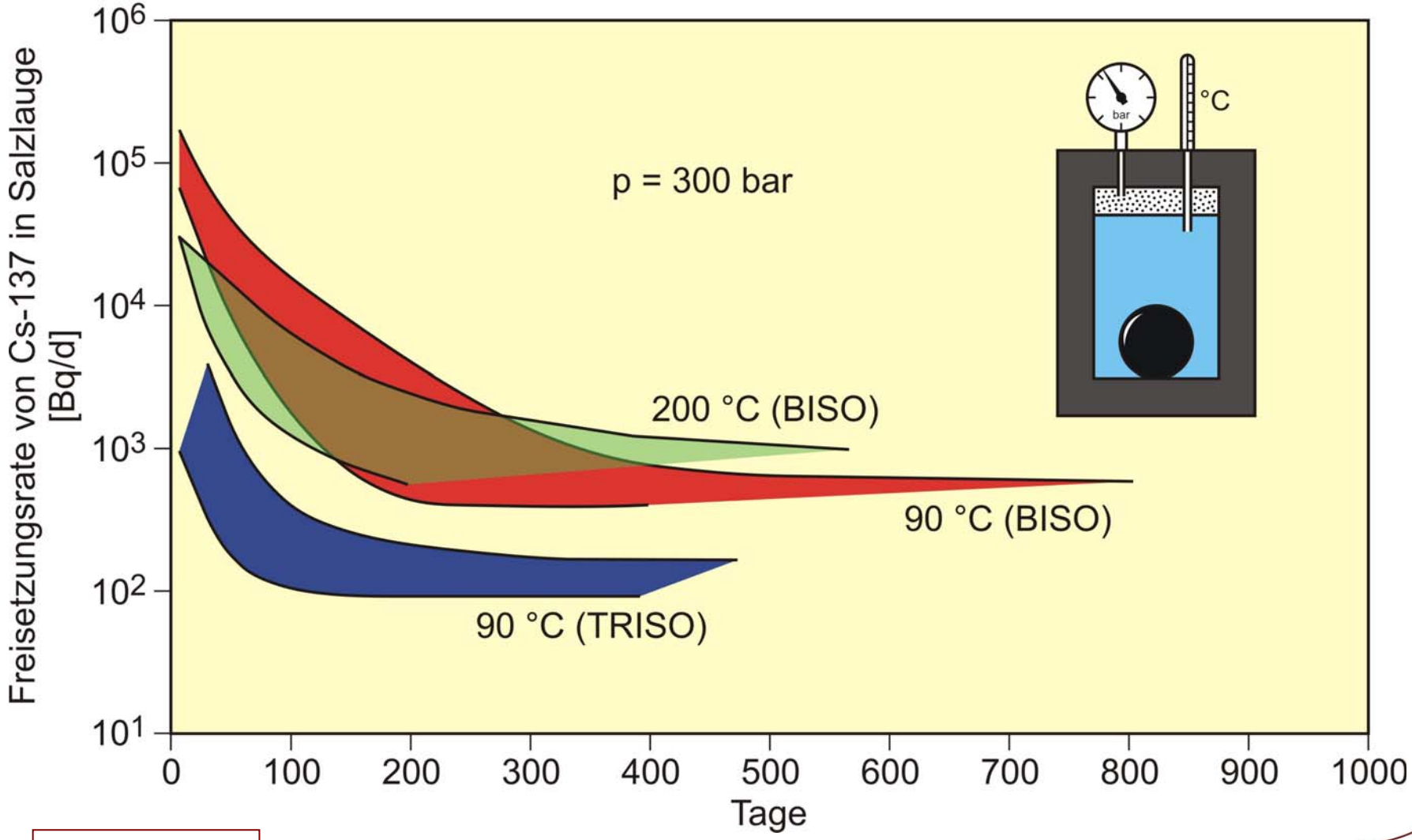
- A salt mine can typically be considered dry, but
  - lye inclusions may migrate
  - ingress of groundwater saturating with salt
- Corrosion experiments with different metal pieces in different compositions of lyes
- Nuclide mobilization experiments with spent fuel spheres in a lye over several years
  @ T up to 200°C and @ p up to 30 MPa

Composition of brines

Compound	Brine-2 [g/l]	Brine-3 [g/l]	Clay* water [g/l]
$MgCl_2\ 6H_2O$	937.08	-	3.457
$MgSO_4$	0.615	1.953	-
NaCl	4.13	309.4	12.38
KCl	1.42	-	0.12
$CaCl_2\ 2H_2O$	39.68	2.74	3.793
K_2SO_4	-	2.83	-
Na_2SO_4	-	2.63	2
$SrCl_2\ 6H_2O$	-	-	0.136
$NaHCO_3$	-	-	0.04
			*Opalinus clay water

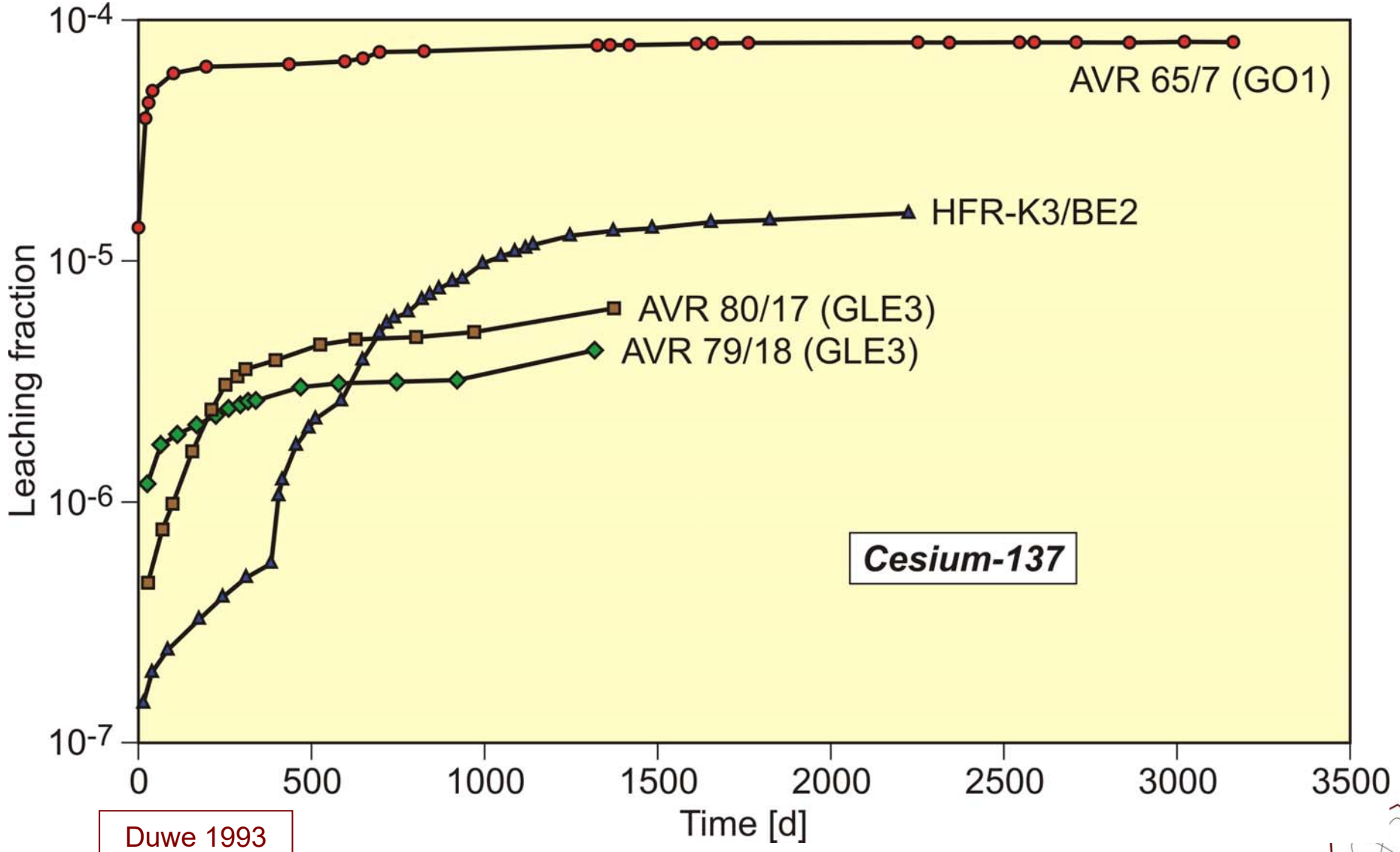
Lye typically changed every month

Cesium release rates in AVR fuel spheres



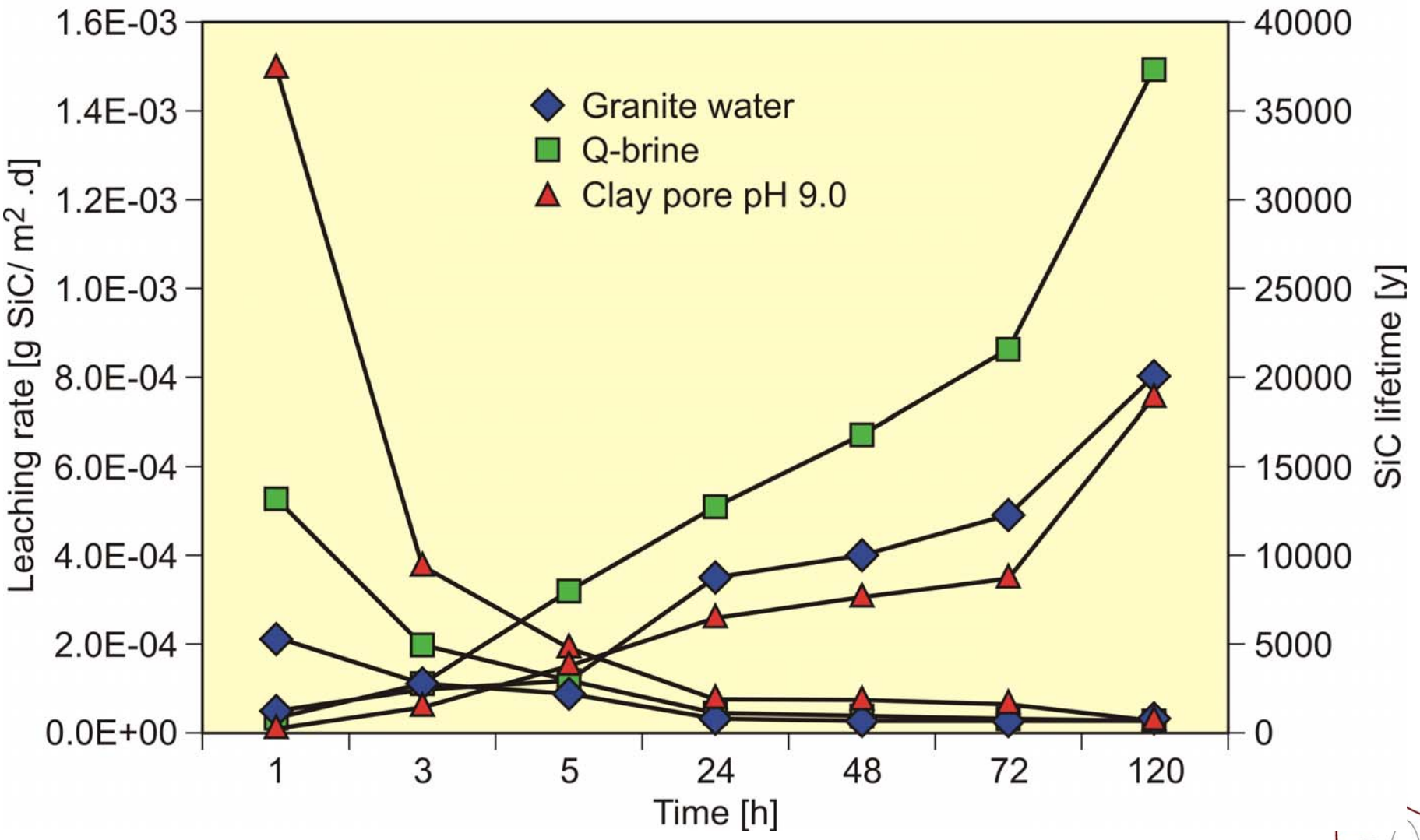
Ganser 1987

Cesium fractional release in AVR fuel spheres



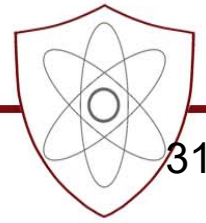
Duwe 1993

SiC leaching rates and lifetime in different media



Results of leach tests

- Initial release of cesium (10-20%) bound at surface and in porosity of matrix
- After 200-300 days, steady-state leach rate
- Higher release from BISO compared to TRISO fuel
- Cesium release rates after 4 yr ~10^3 Bq/d, fraction $F_{tot} < 10^{-4}$
- Much higher release for defective/failed coated particles
- 100% release for bare UO_2 kernels, ~10% for bare $(U,Th)O_2$ kernels after 1 yr
- No corrosion effect on intact TRISO coating



Summary and conclusions

- Much experience obtained on the characterization of spent HTGR fuel from AVR and THTR-300
- All fuel in intermediate storage before agreement is made on final repository for direct disposal
- Segregation of fuel matrix recommended, various processes demonstrated on non-irradiated fuel
- More future activities on separate effect investigations on barrier function of single fuel components desirable with focus on TRISO