ENGINEERED BARRIERS FOR THE DISPOSAL OF NUCLEAR FUEL WASTE

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HUMAN RESOURCES DEVELOPMENT
LINKAGE PROGRAM

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REGULATORY REQUIREMENTS FOR HIGH-LEVEL WASTE DISPOSAL

IAEA - **Safety Principles and Technical Criteria for the Underground Disposal of High-Level Radioactive Wastes: IAEA Safety Series No. 99 (IAEA 1989)**

CANADA - **Regulatory Documents:**
R-72 (AECB 1987),
R-104 (AECB 1987)

KEY IAEA TECHNICAL CRITERIA FOR ENGINEERED BARRIERS (IAEA 1989)

- The long-term safety of high-level radioactive waste disposal shall be based on the multibarrier concept and shall be addressed on the basis of the disposal system as a whole.
- Substantially complete isolation for an initial period of time
- Repository operation and closure should preserve the post-sealing safety functions of the host rock
- Waste should be emplaced such that fissile material remains in a subcritical configuration

REGULATORY REQUIREMENTS RELEVANT TO ENGINEERED BARRIERS FOR GEOLOGIC DISPOSAL OF NUCLEAR FUEL WASTE

1. No dependence on intervention in the post-closure period should be required
2. A quality assurance program must be in place at all stages
3. Multiple (engineered plus natural) barriers must be used
4. The disposal system must not be compromised by provisions for
    a. pre-closure measurements
    b. post-closure retrieval
    c. post-closure measurements

REQUIREMENTS FOR DETERMINING THE ACCEPTABILITY OF A DISPOSAL CONCEPT

1. **CRITERIA** that define what is acceptably safe
2. **METHODOLOGY** to evaluate the performance of a proposed disposal system against the safety criteria
3. **TECHNOLOGY** to site, design, build, operate, decommission and close a disposal facility that satisfies the safety criteria
4. **CONFIDENCE** that an acceptable site exists, that, together with a suitably designed facility, would meet the safety criteria

ENGINEERED BARRIERS - GUIDING PRINCIPLES FOR R & D PROGRAM

- compatible with disposal at 500 - 1000-m depth in plutonic rock
- design technically feasible with available technology, or reasonably achievable developments
- flexible design approach to provide a range of options
- engineered barriers performance assessed in terms of the overall disposal system

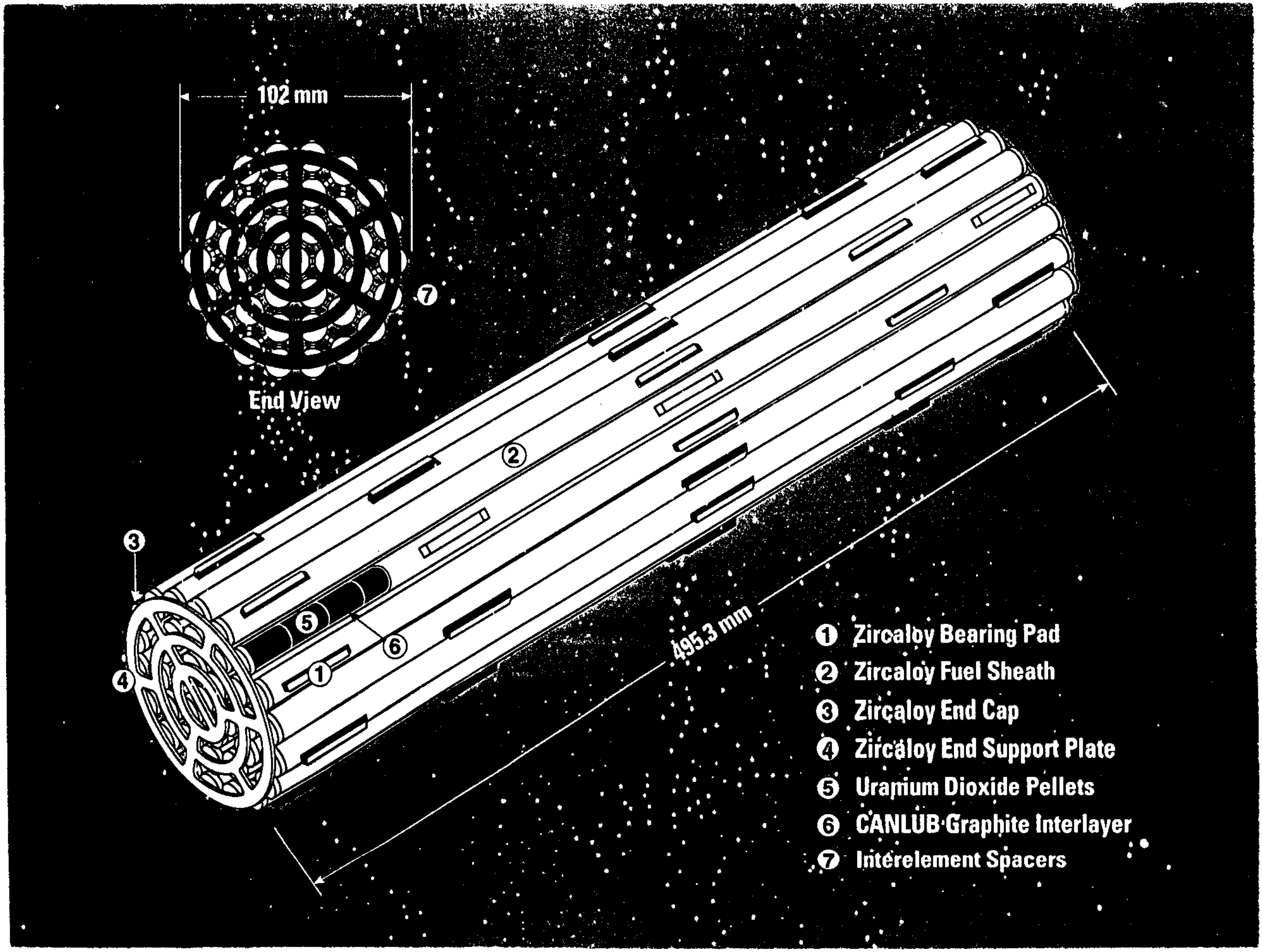
ENGINEERED BARRIERS R & D

OBJECTIVES

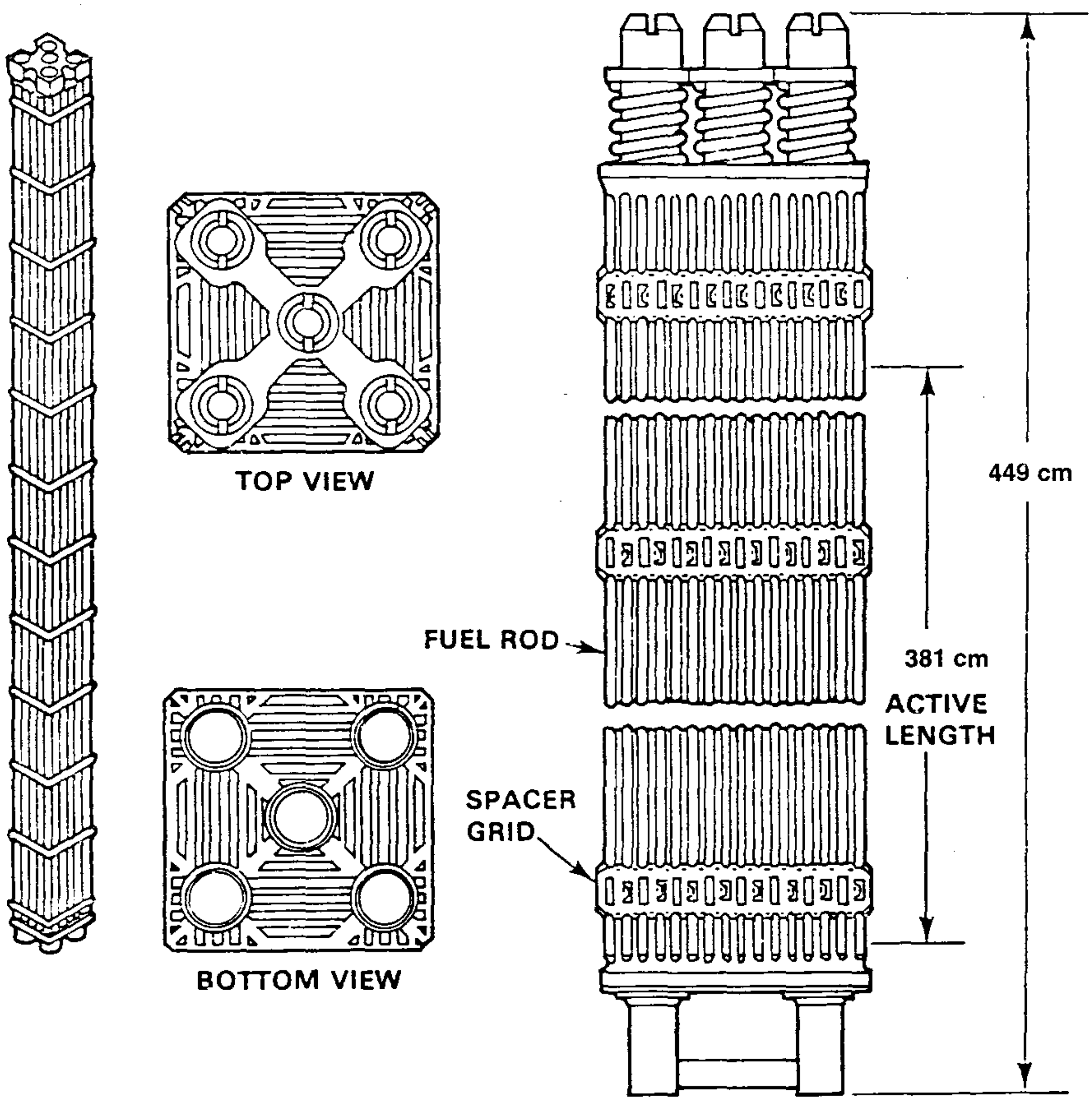
- evaluate the performance of a used fuel as a waste form
- develop and assess processes and waste forms for immobilizing wastes from fuel re-processing
- develop containers to isolate the waste for an appropriate period
- develop materials and designs to effectively seal a disposal vault
- develop models to describe the rate of release and transport of radionuclides to the geosphere
- develop the base of understanding to defend the models and to define the limits of acceptable performance of the engineered barriers

WASTE FORMS

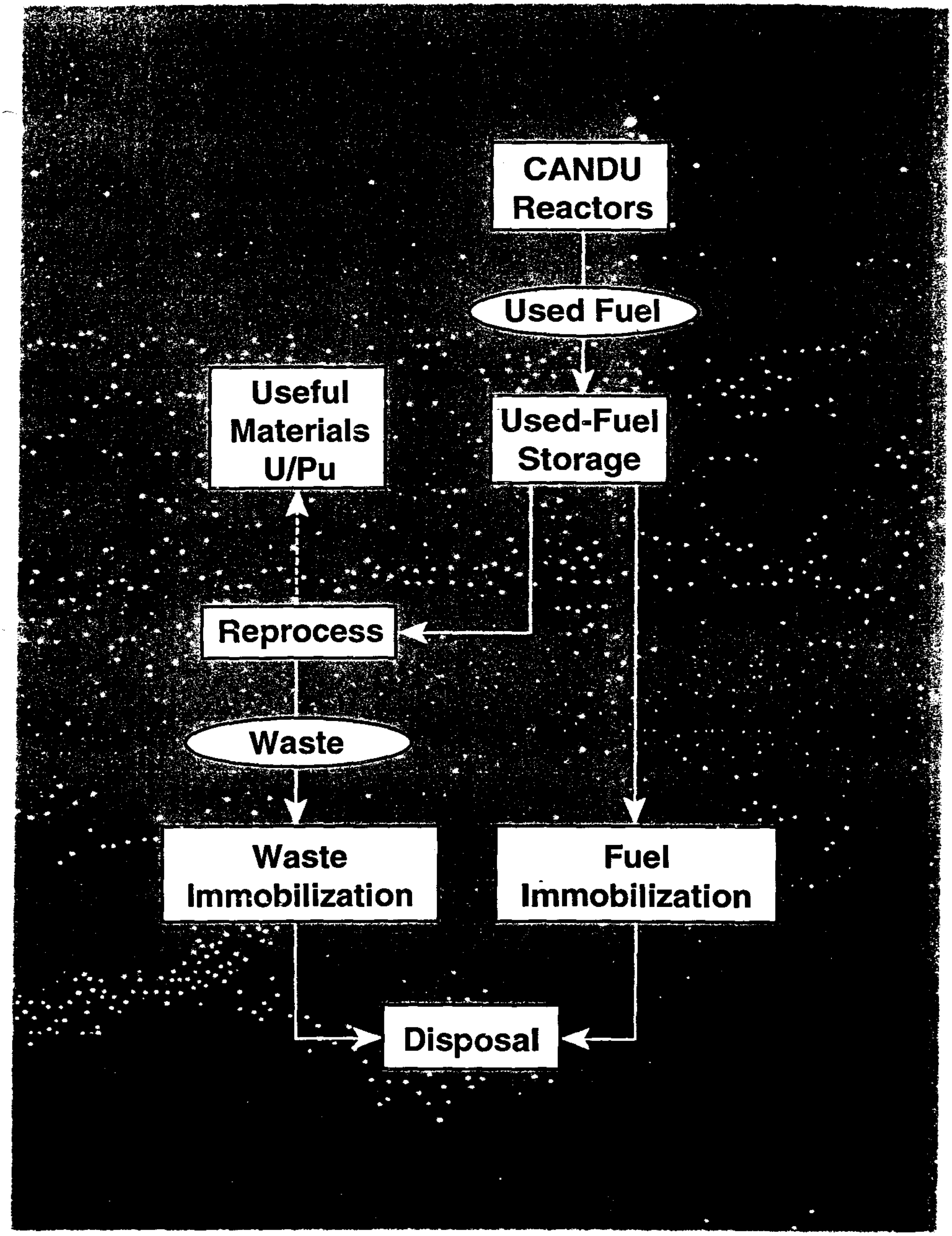
- OPTIONS
- CHARACTERIZATION
- RADIONUCLIDE INVENTORIES
- RELEASE CHARACTERISTICS
- CONCEPTUAL MODELS FOR RELEASE
- MATHEMATICAL MODELS FOR RELEASE

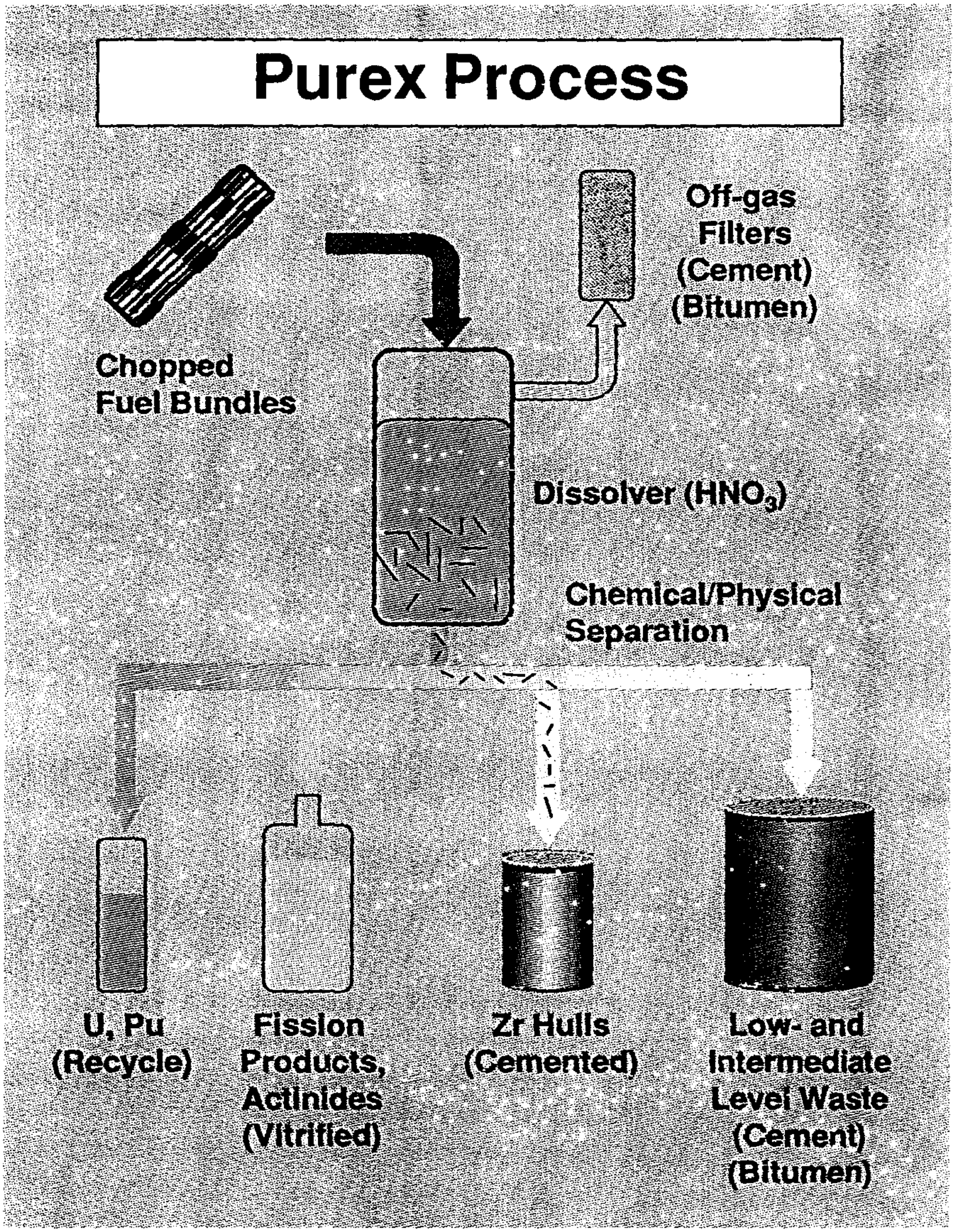


102 mm
7
End View
2
3
5
6
1
4
495.3 mm
1 Zircaloy Bearing Pad
2 Zircaloy Fuel Sheath
3 Zircaloy End Cap
4 Zircaloy End Support Plate
5 Uranium Dioxide Pellets
6 CANLUB Graphite Interlayer
7 Interelement Spacers



LWR FUEL ASSEMBLY


CANDU Reactors
Used Fuel
Used-Fuel Storage
Useful Materials U/Pu
Reprocess
Waste
Waste Immobilization
Fuel Immobilization
Disposal


Purex Process
Chopped
Fuel Bundles
Off-gas
Filters
(Cement)
(Bitumen)
Dissolver (HNO_3)
Chemical/Physical
Separation
U, Pu
(Recycle)
Fission
Products,
Actinides
(Vitrified)
Zr Hulls
(Cemented)
Low- and
Intermediate
Level Waste
(Cement)
(Bitumen)

Products for Immobilization of High-level Liquid Fission Product and Actinide Reprocessing Wastes

Vitrification (Immobilization in glass or glass-ceramic)

Borosilicate Glass - International product choice a reprocessing facilities.

AECL-developed products

Borosilicate Glasses

Aluminosilicate Glasses

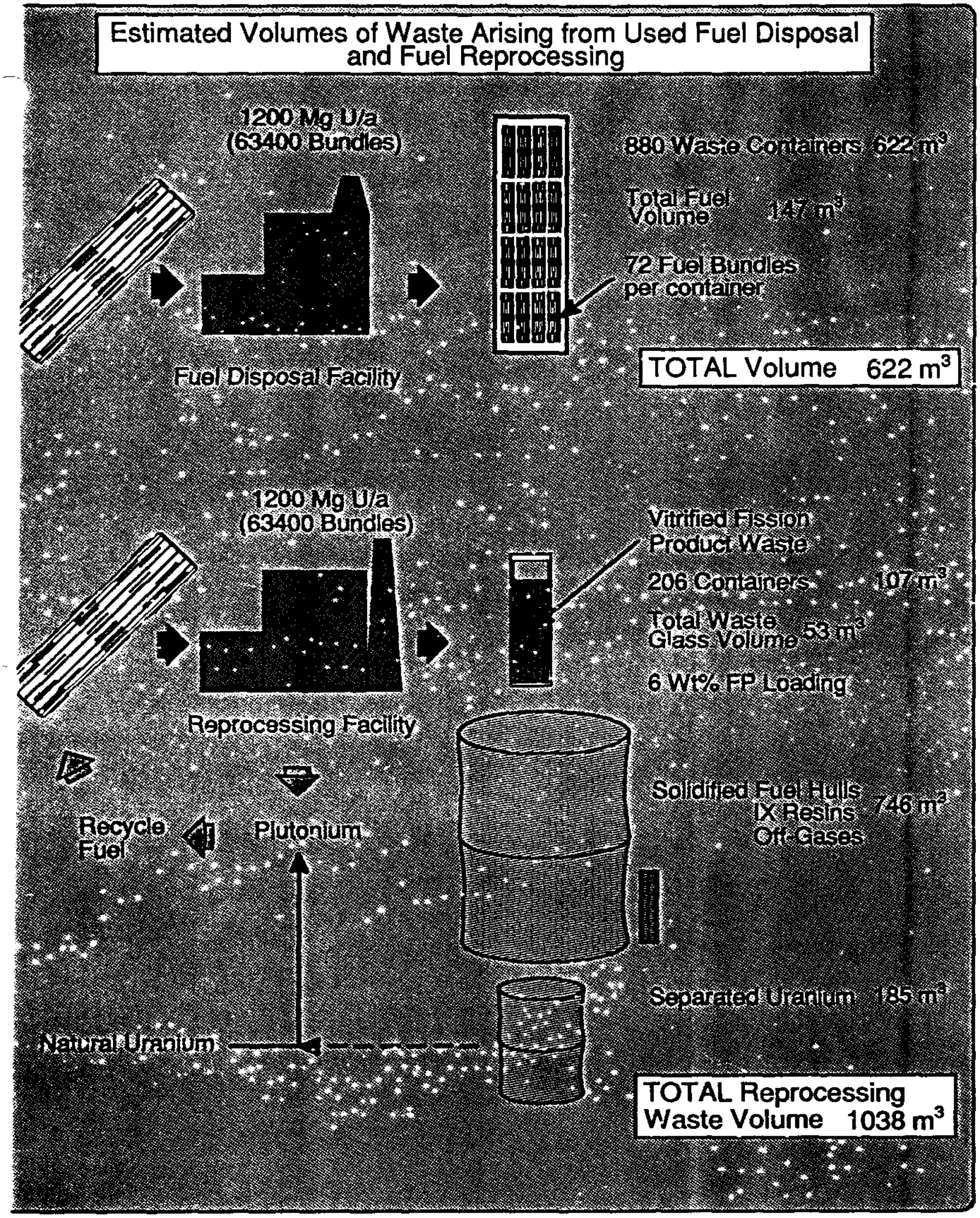
Sphene-based Glass-ceramics

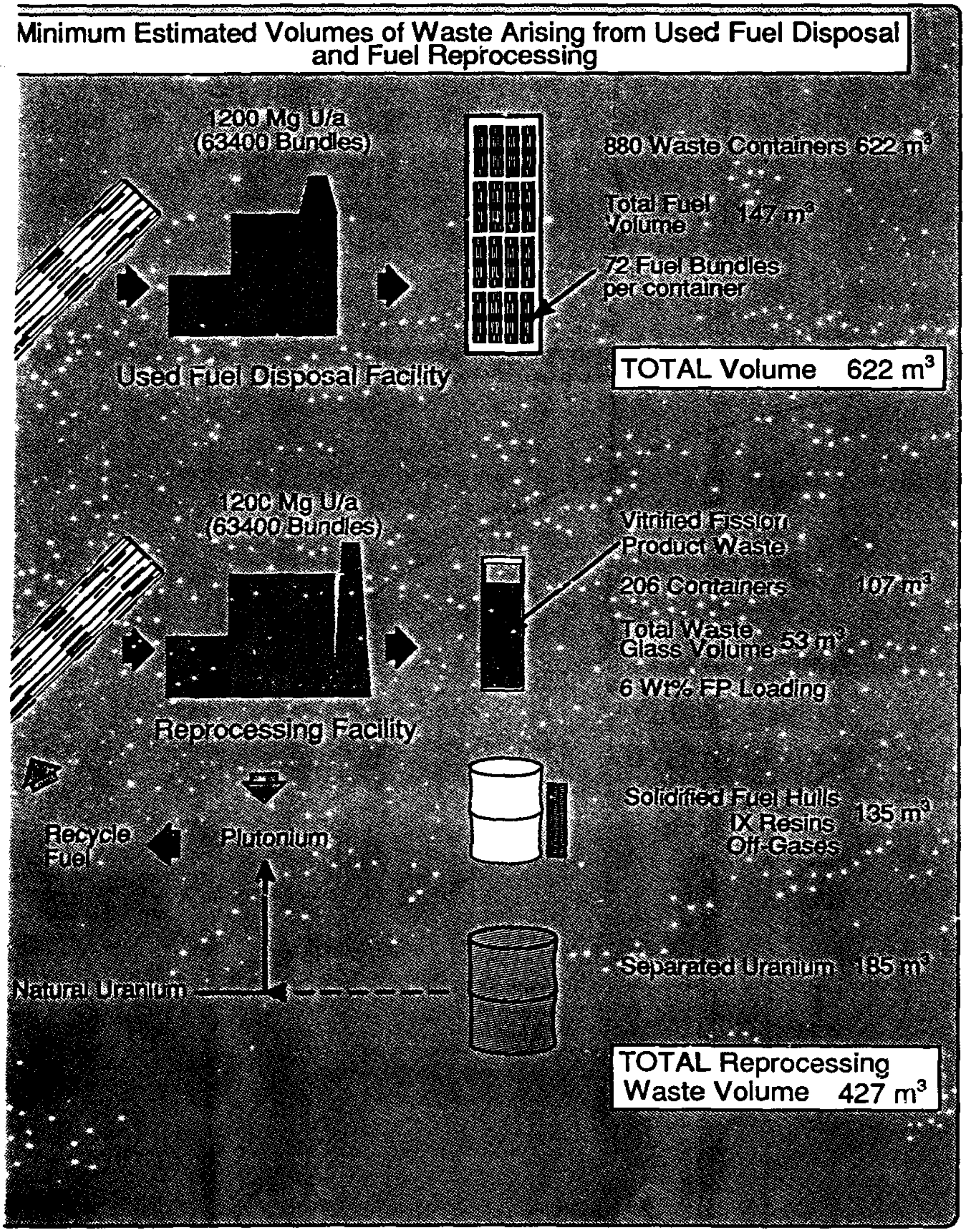
Products were developed to provide enhanced durability ove conventional borosilicate glasses and to be compatible with disposal ii a granitic rock repository.

Models were developed to describe the dissolution behaviour of thes products.

Glass-melter technology was developed for the fabrication of thes waste-forms.

Any future work would involve product optimization and scaling-up o production technologies.


Estimated Volumes of Waste Arising from Used Fuel Disposal and Fuel Reprocessing
1200 Mg U/a
(63400 Bundles)
880 Waste Containers 622 m³
Total Fuel Volume 147 m³
72 Fuel Bundles per container
Fuel Disposal Facility
TOTAL Volume 622 m³
1200 Mg U/a
(63400 Bundles)
Vitrified Fission Product Waste
206 Containers 107 m³
Total Waste Glass Volume 53 m³
6 Wt% FP Loading
Reprocessing Facility
Recycle Fuel
Plutonium
Solidified Fuel Hulls
IX Resins
Off-Gases
746 m³
Separated Uranium 185 m³
Natural Uranium
TOTAL Reprocessing Waste Volume 1038 m³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Used Fuel Disposal Facility
TOTAL Volume 622 m³
1200 Mg U/a
(63400 Bundles)
Vitrified Fission Product Waste
206 Containers 107 m³
Total Waste Glass Volume 53 m³
6 Wt% FP Loading
Reprocessing Facility
Solidified Fuel Hulls
IX Resins
Off-Gases
135 m³
Recycle Fuel
Plutonium
Separated Uranium 185 m³
Natural Uranium
TOTAL Reprocessing Waste Volume 427 m³

Impact of Reprocessing on Waste Disposal

Disposal of 63,400 fuel bundles (1200 Mg U) per year
10 years cooled

	Used Fuel	High-Level Reprocessing Waste
# Containers	880	206
Container Volume	622 m^3	107 m^3
Fission Product Waste per Container	11 kg	47 kg
Total Fission Product Waste	9700 kg	9700 kg
Heat per Container	286 W	1223 W
Total Heat	252 kW	252 kW

To maintain a maximum temperature of 90°C at the container surfac reprocessing waste containers would require wider spacing.

Volume of vault required for disposal of waste cooled for equivalent tim period would be identical.

ADVANCED FUEL CYCLES

- Slightly Enriched Uranium Oxide (0.9 - 1.5% U-235)
- Mixed-Oxide - $(U, Pu)O_2$
- Tandem - LWR → CANDU
- Thorium Fuel

Detailed Analysis of Environmental Impacts and Economic Aspects Not Yet Performed

UO_2 FUELS

	CANDU	PWR
Fuel	Nat. UO_2 0.7% ^{235}U	Enriched UO_2 ~3-4% ^{235}U
Cladding	Zircaloy	Zircaloy
Assembly length (m)	0.5 m	~4 m
Rods/assembly	28 or 37	17 x 17
UO_2 wt. (kg)	21 kg	523 kg
Burnup (MWd/MTHM)	6000 - 12000	8000 - 40000
Linear Power (kW/m)	20 - 55	15 - 25
T centerline (°C)	800 - 1700	800 - 1200

FACTORS AFFECTING CHARACTERISTICS OF USED FUEL FOR DISPOSAL

- Fuel history - burn up, linear power
- Fuel defects
- External contamination, crud
- Storage time, changes during storage
- Changes during transportation

EFFECTS OF IRRADIATION ON UO_2 FUEL

- o During irradiation of fuel in reactor, Actinides and many Fission Products remain homogeneously distributed in UO_2 crystalline lattice.

- o Some Fission Products may segregate to specific locations in the UO_2 fuel:

  - Tc, Rh, Pd, Ru and Mo are insoluble in lattice and are submicroscopically dispersed in UO_2 lattice and migrate to UO_2 grain boundaries.

  - Volatile Fission Products (e.g. Cs, I) and fission gases (Xe, Kr) migrate to UO_2 grain boundaries and also along grain boundaries to the fuel/Zircaloy sheath gap region.

- o High power/high temperature fuels will show the greatest segregation and the highest concentrations of Cs, Xe and I in the gap region.

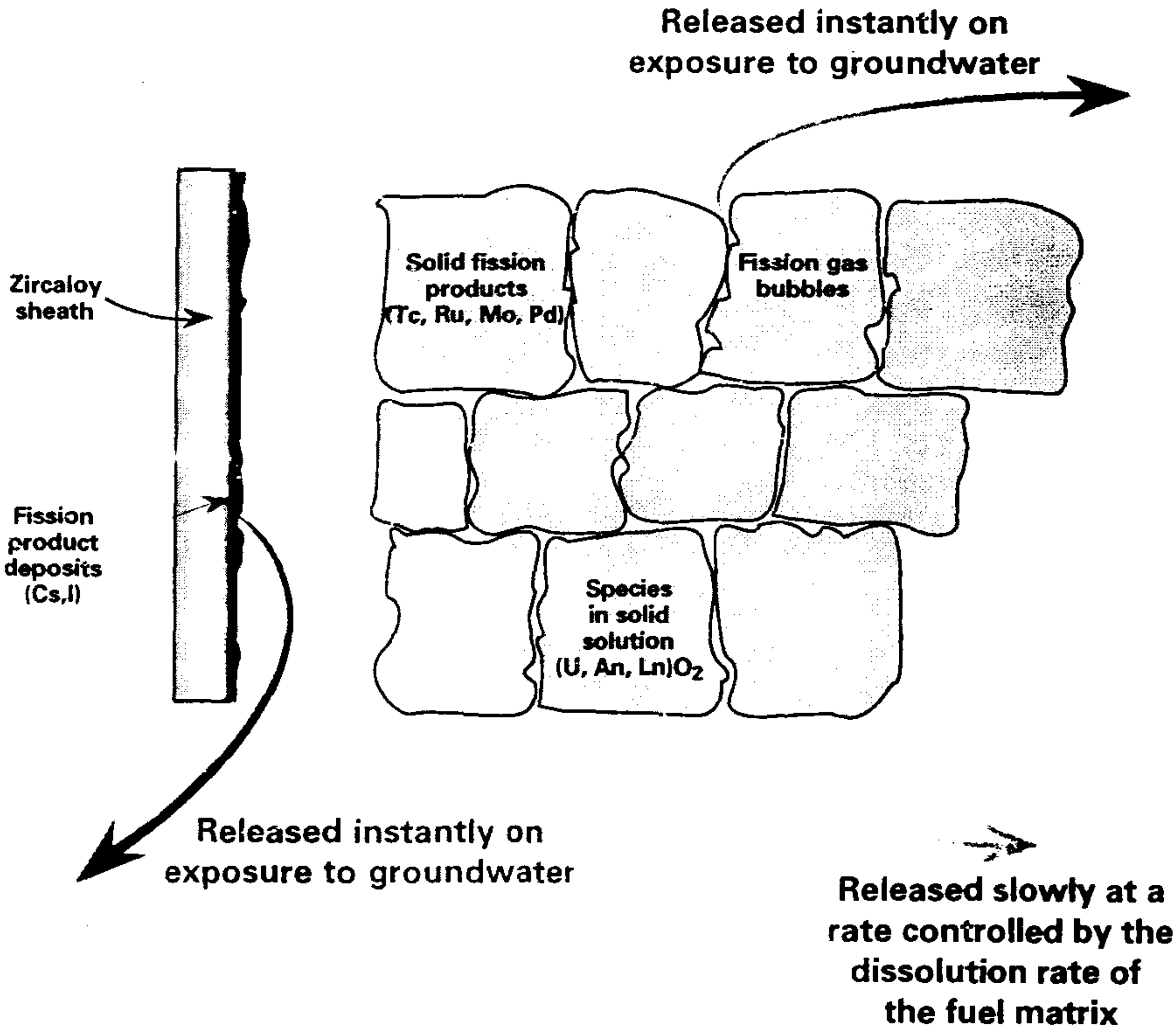
For typical CANDU fuels, total fission product inventory may be:

$\sim$0.05 to 15% (average $\sim$2%) in the gap region,

$\sim$5% in grain boundaries,

remainder ($\sim$80-95%) in UO_2 matrix.

Release of Radionuclides from used Fuel



MP96-028.32

RESEARCH TO EVALUATE DURABILITY OF USED FUEL DURING DISPOSAL

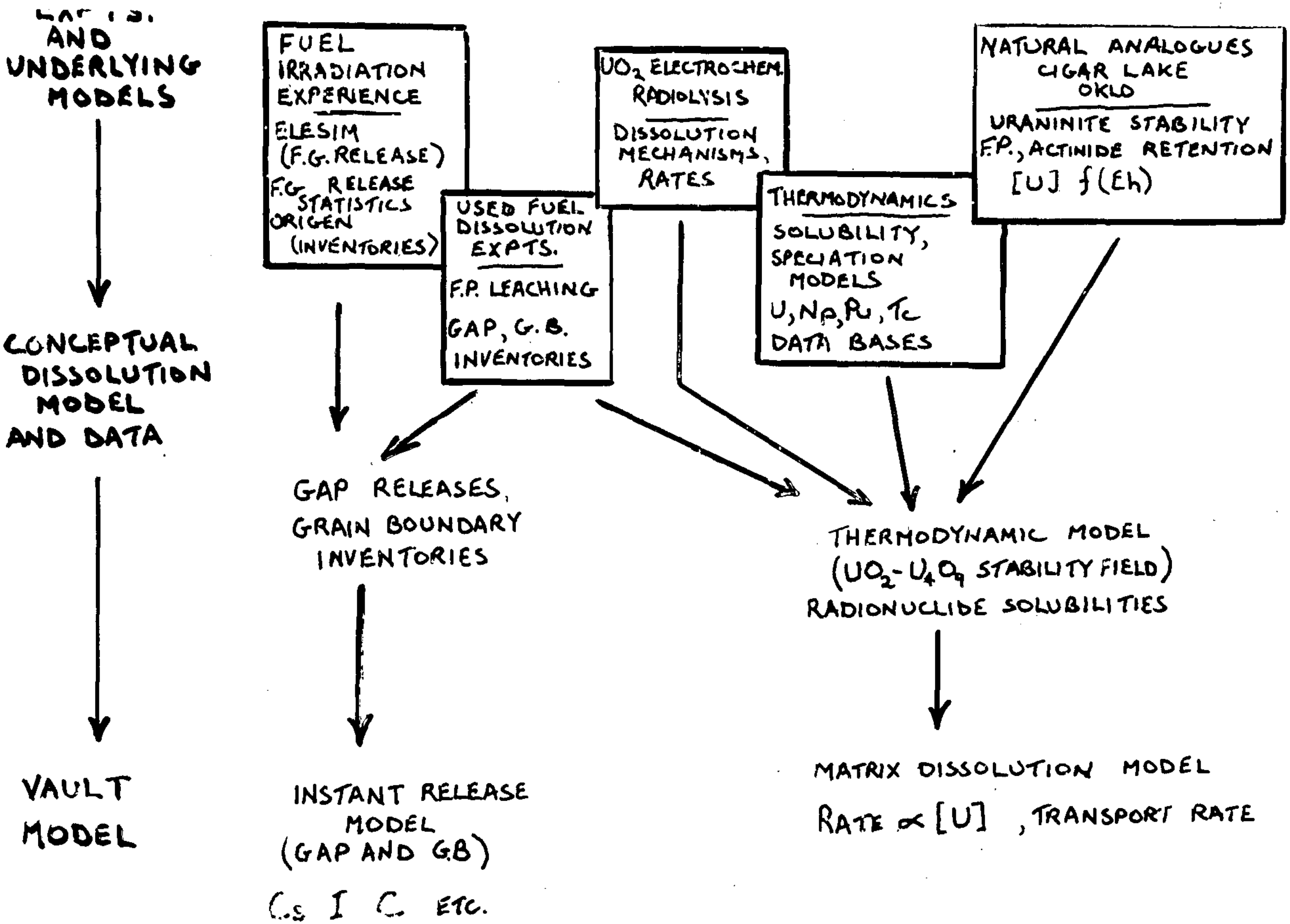
- **INVENTORIES AND DISTRIBUTION OF RADIONUCLIDES IN USED FUEL**
  - predictive codes, e.g., ORIGEN-S
  - measurements to validate predictions

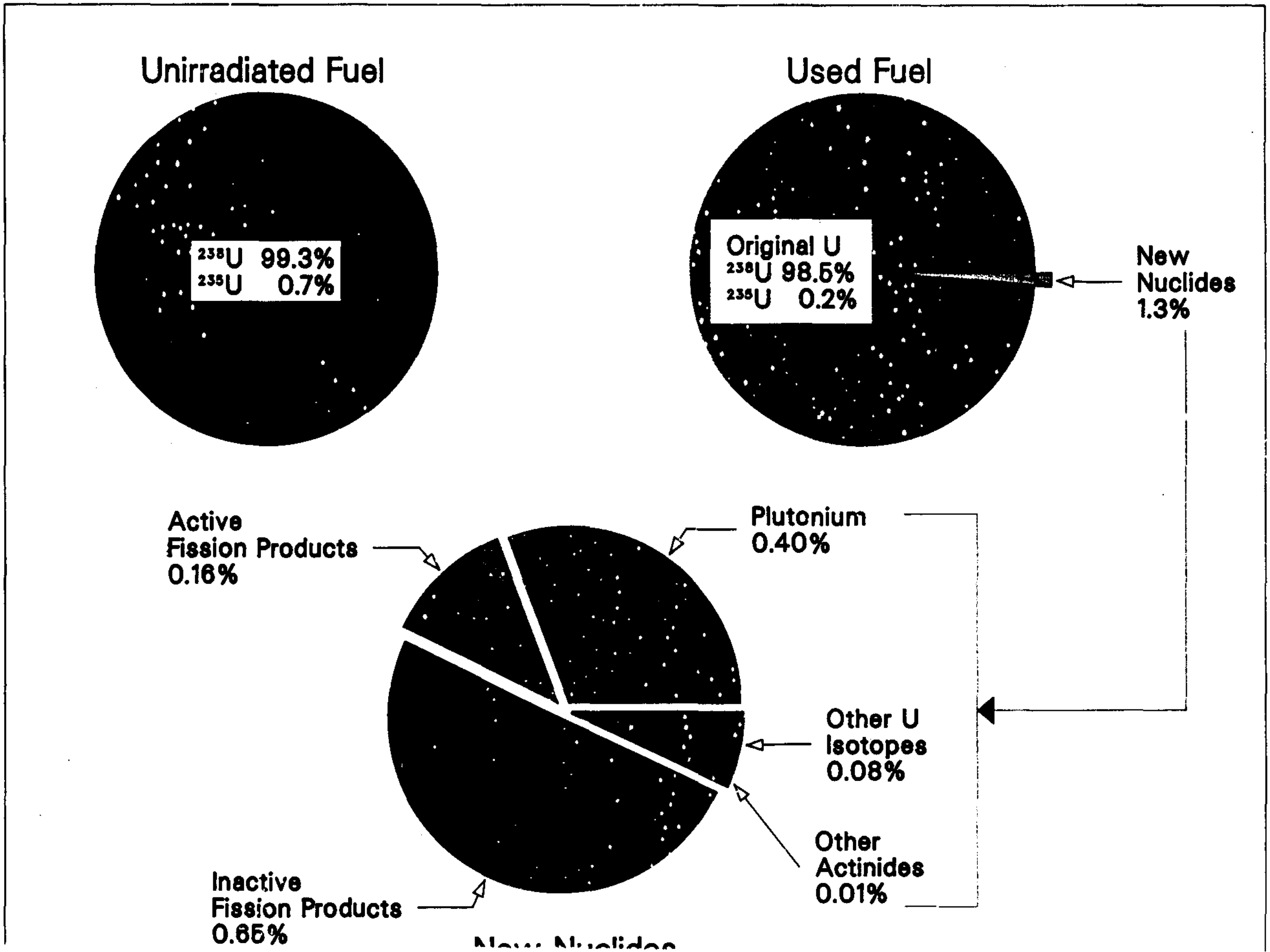
- **STUDIES OF DISSOLUTION OF USED FUEL**
  - radionuclide release from fuel/cladding gap
  - radionuclide release from grain boundaries
  - radionuclide release from fuel matrix
  - fuel/groundwater, multicomponent tests

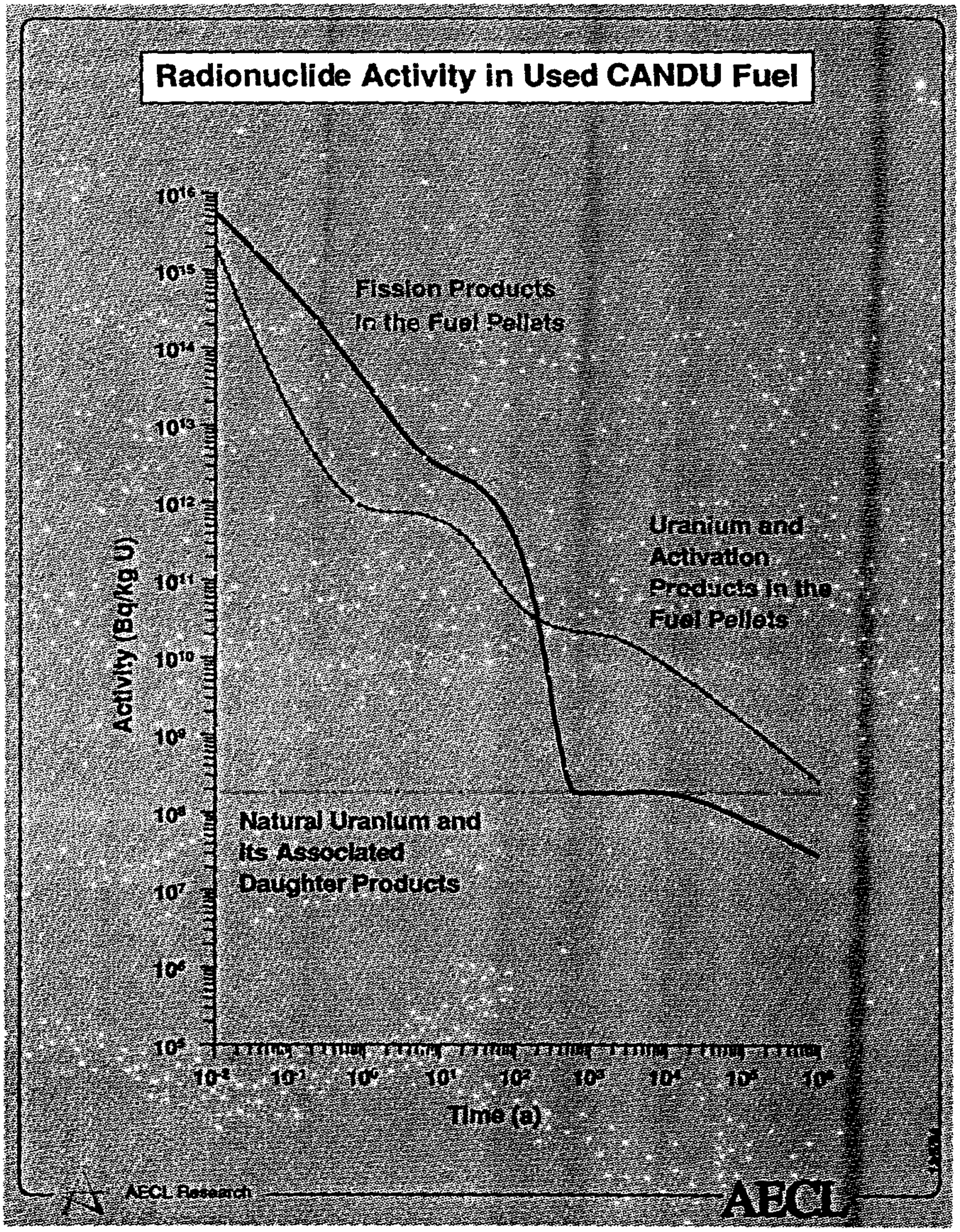
- **STUDIES OF DISSOLUTION OF UO_2**
  - in solutions containing O_2 and H_2O_2
  - effects of α and γ radiolysis
  - oxidative dissolution model development

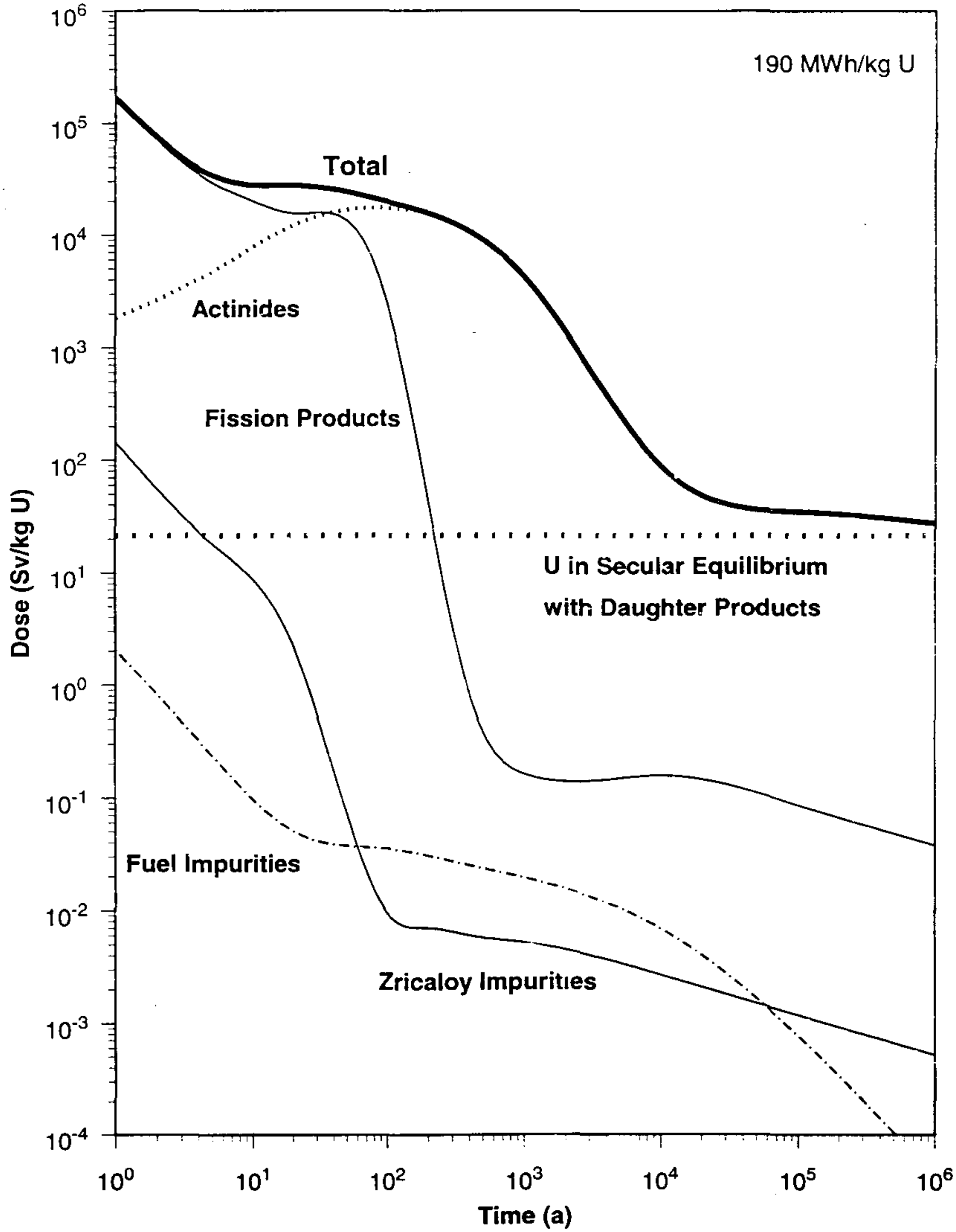
- **USED FUEL DISSOLUTION MODEL**
  - evaluate instant release
  - uranium solubility function
  - radionuclide solubilities

- **STUDIES OF NATURAL ANALOGUES**
  - U-ore deposits


AND UNDERLYING MODELS
CONCEPTUAL DISSOLUTION MODEL AND DATA
VAULT MODEL
FUEL IRRADIATION EXPERIENCE
ELESIM (F.G. RELEASE)
F.G. RELEASE STATISTICS
ORIGEN (INVENTORIES)
USED FUEL DISSOLUTION EXPTS.
F.P. LEACHING
GAP, G.B. INVENTORIES
UO2 ELECTROCHEM. RADIOLYSIS
DISSOLUTION MECHANISMS, RATES
THERMODYNAMICS
SOLUBILITY, SPECIATION MODELS
U, Np, Pu, Tc DATA BASES
NATURAL ANALOGUES CIGAR LAKE OKLO
URANINITE STABILITY
F.P., ACTINIDE RETENTION
[U] f(Eh)
GAP RELEASES, GRAIN BOUNDARY INVENTORIES
THERMODYNAMIC MODEL (UO2-U4O9 STABILITY FIELD)
RADIONUCLIDE SOLUBILITIES
INSTANT RELEASE MODEL (GAP AND G.B)
Cs I C ETC.
MATRIX DISSOLUTION MODEL
RATE ∝ [U], TRANSPORT RATE

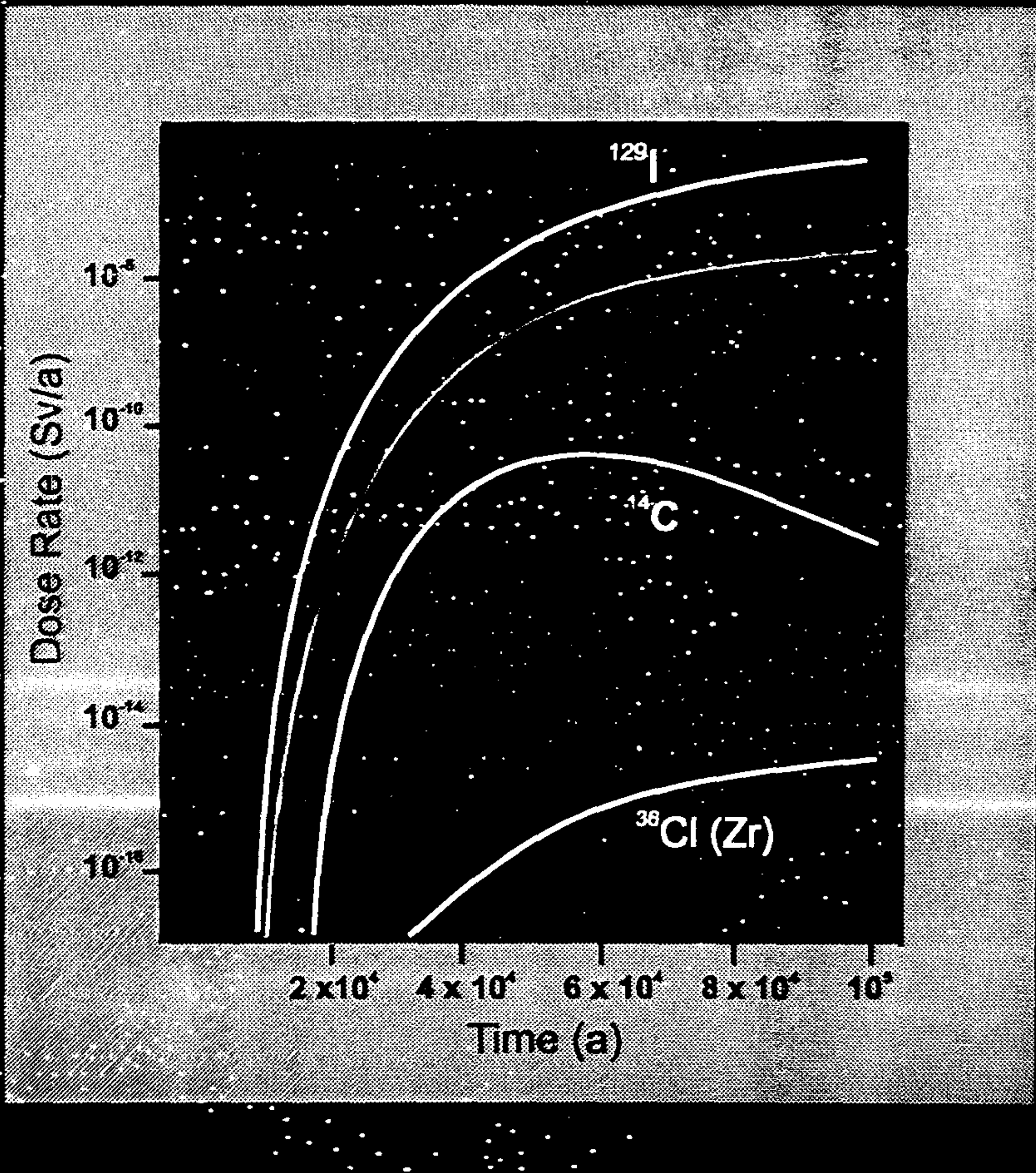

Unirradiated Fuel
^{238}U 99.3%
^{235}U 0.7%
Used Fuel
Original U
^{238}U 98.5%
^{235}U 0.2%
New Nuclides 1.3%
Active Fission Products 0.16%
Plutonium 0.40%
Other U Isotopes 0.08%
Other Actinides 0.01%
Inactive Fission Products 0.65%

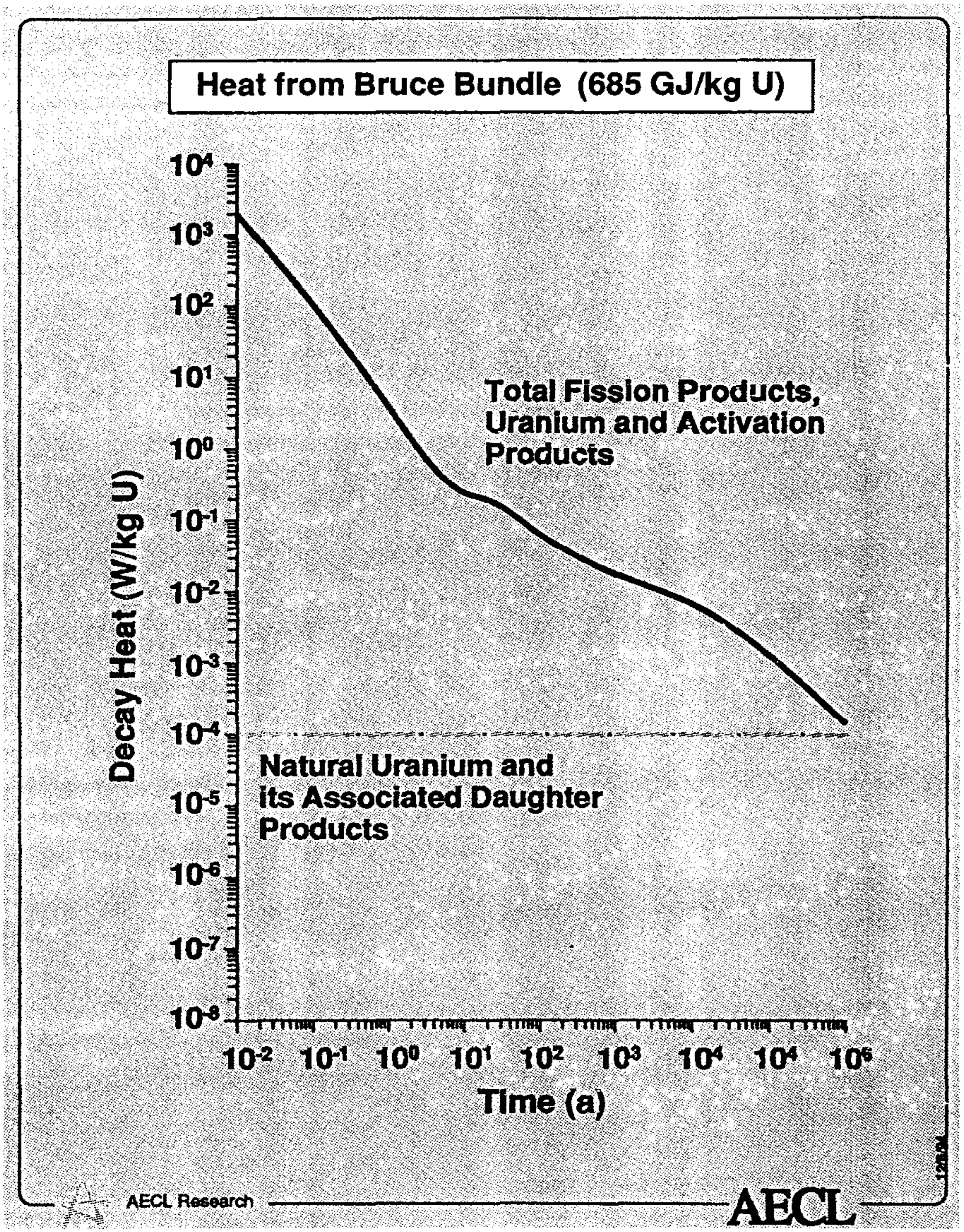

Radionuclide Activity in Used CANDU Fuel
Fission Products
in the Fuel Pellets
Uranium and
Activation
Products in the
Fuel Pellets
Natural Uranium and
its Associated
Daughter Products
Activity (Bq/kg U)
Time (a)
AECL Research
AECL



190 MWh/kg U
Total
Actinides
Fission Products
U in Secular Equilibrium
with Daughter Products
Fuel Impurities
Zricaloy Impurities
Dose (Sv/kg U)
Time (a)
10^{6}
10^{5}
10^{4}
10^{3}
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{0}
10^{1}
10^{2}
10^{3}
10^{4}
10^{5}
10^{6}

Median-Dose for ^{129}I, ^{36}Cl and ^{14}C



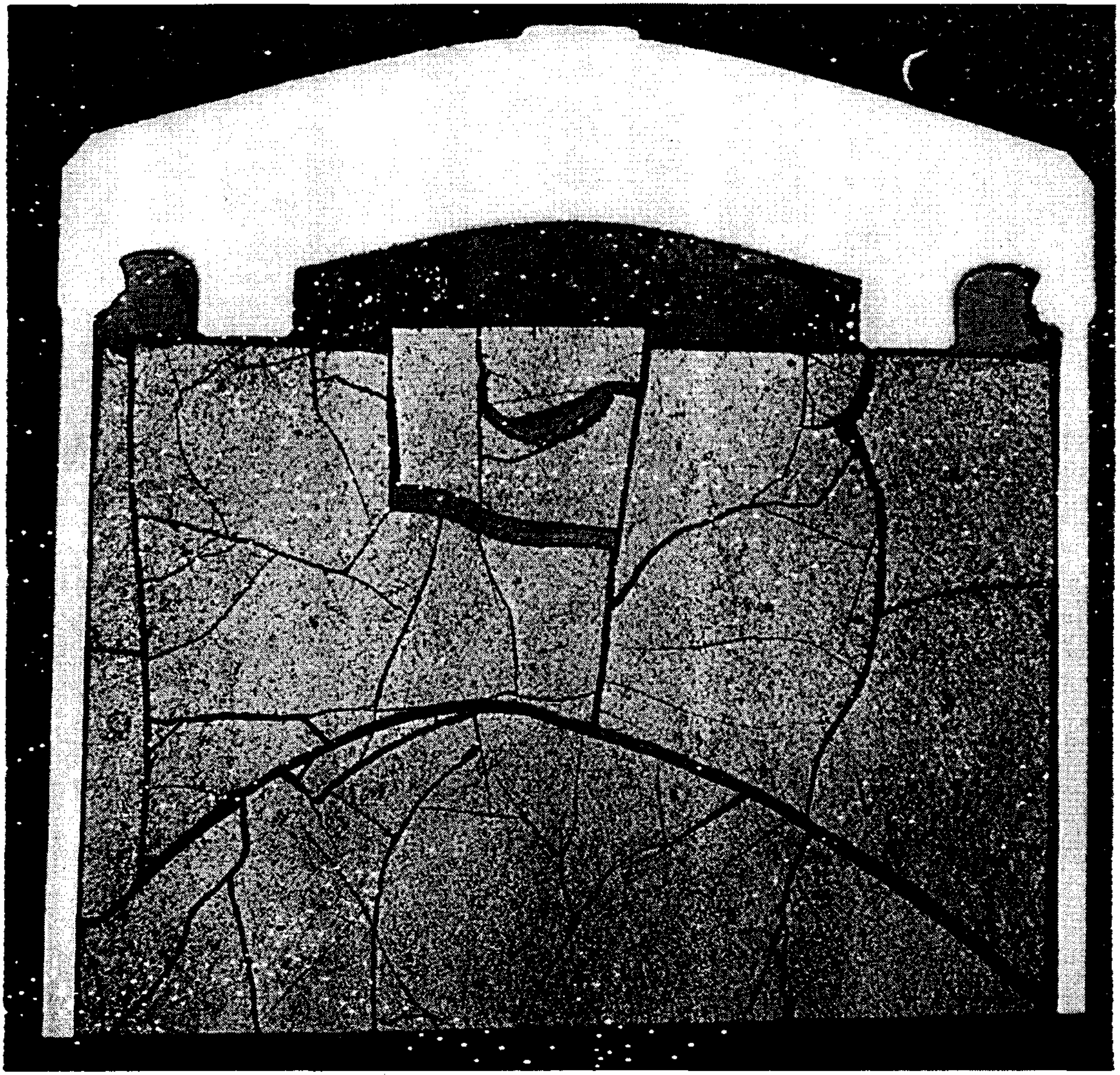


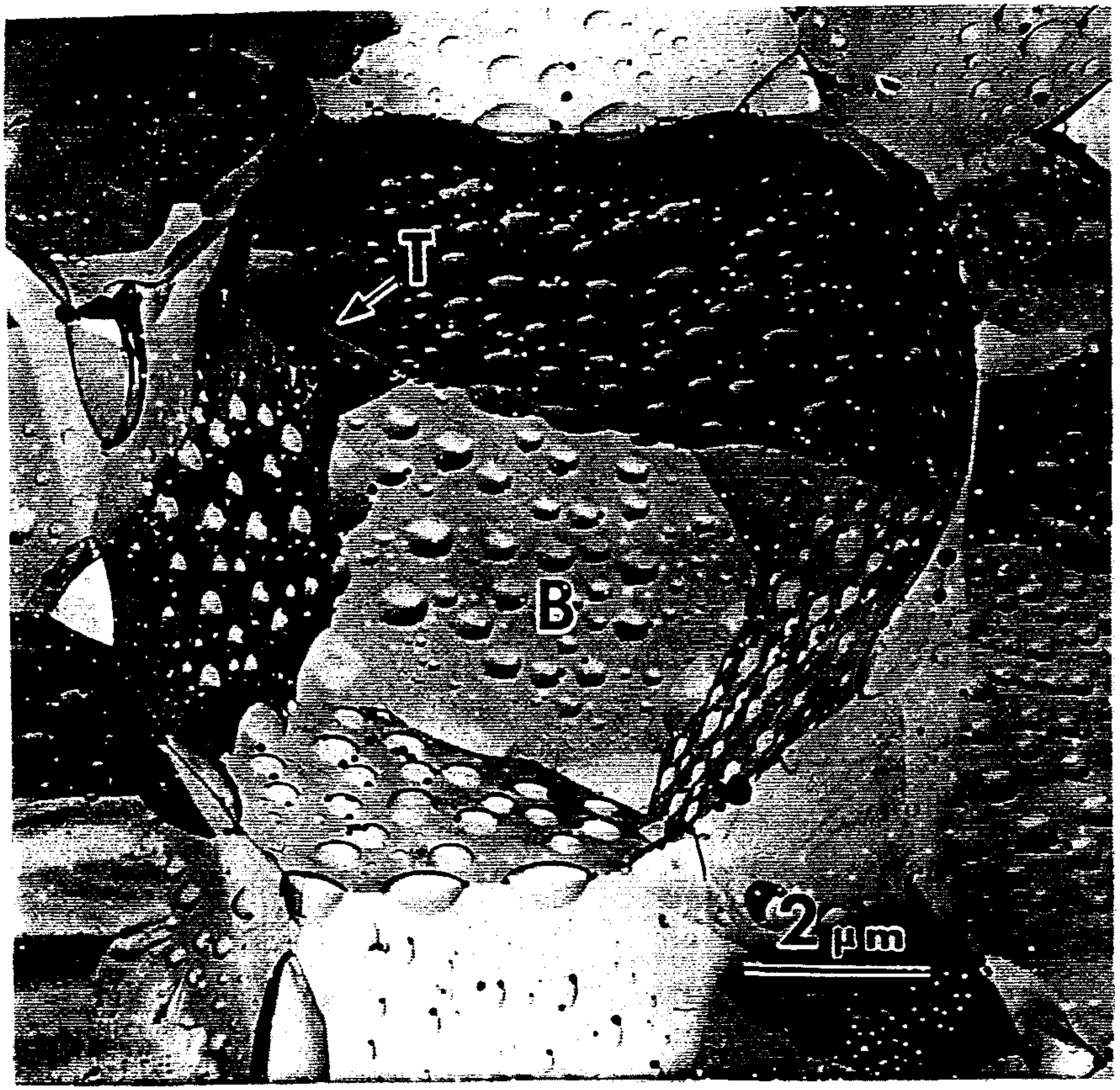
Heat from Bruce Bundle (685 GJ/kg U)
Decay Heat (W/kg U)
10⁴
10³
10²
10¹
10⁰
10⁻¹
10⁻²
10⁻³
10⁻⁴
10⁻⁵
10⁻⁶
10⁻⁷
10⁻⁸
Total Fission Products, Uranium and Activation Products
Natural Uranium and its Associated Daughter Products
10⁻² 10⁻¹ 10⁰ 10¹ 10² 10³ 10⁴ 10⁴ 10⁶
Time (a)
AECL Research
AECL

Origen-S Pickering Fuel Comparison

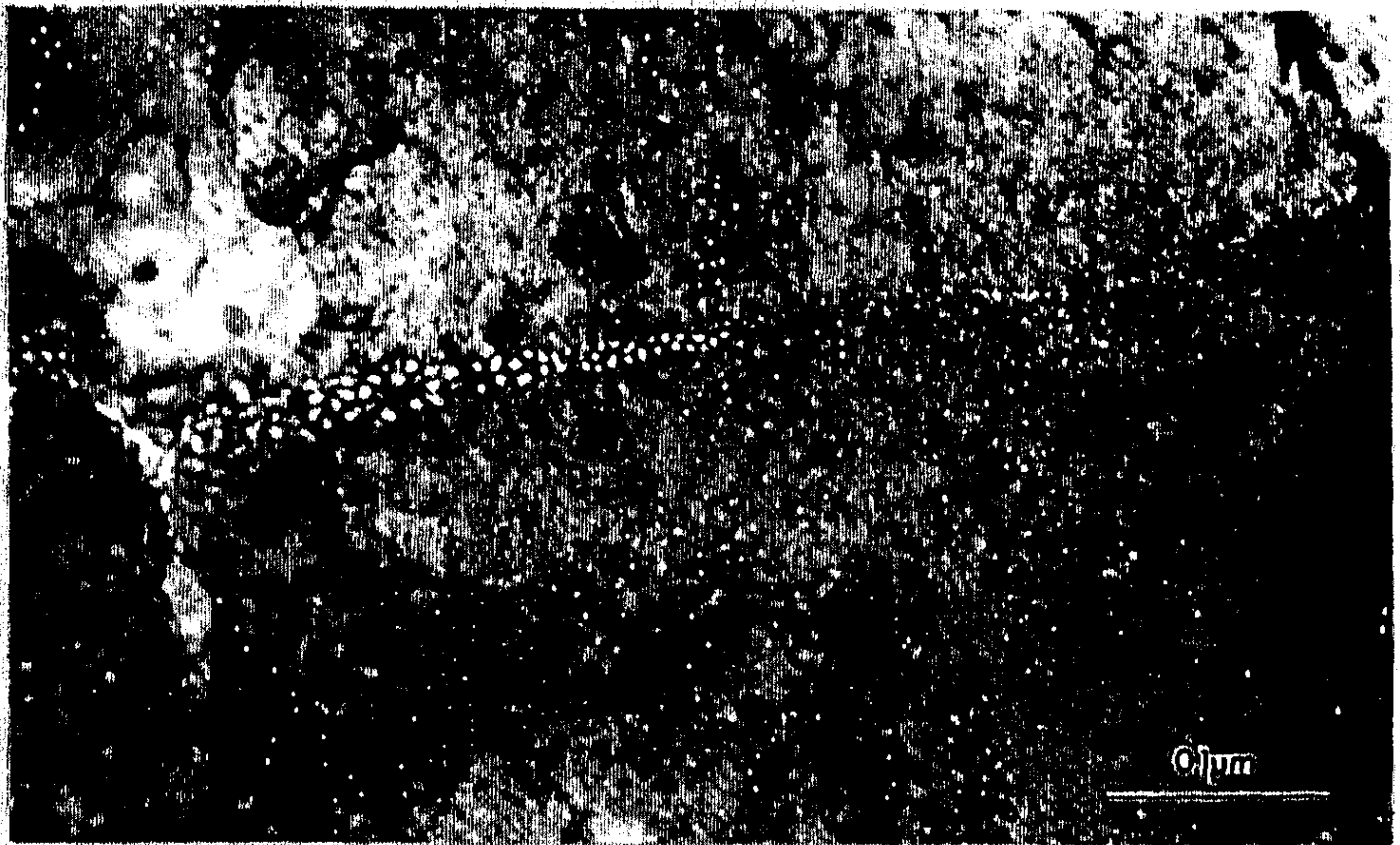
	Measured Inventory Bq/kg U	Origen-S Bq/kg U	Ratio Meas/Calc	Error +/-
Cm-244	7.12e+08	7.92e+08	0.90	0.13
Am-241	1.86e+10	1.87e+10	0.99	0.20
Np-237	9.99e+05	8.99e+05	1.11	0.22
Eu-155	3.35e+09	4.29e+09	0.78	0.06
Eu-154	8.14e+09	1.55e+10	0.52	0.03
Cs-137	8.05e+11	7.84e+11	1.03	0.05
Cs-134	4.16e+09	4.07e+09	1.02	0.07
I-129	2.44e+05	3.61e+05	0.68	
Sb-125	2.20e+09	2.56e+09	0.86	0.16
Ru-106	8.72e+07	2.52e+08	0.35	0.02
Tc-99	1.08e+08	1.50e+08	0.72	0.07
Sr-90	4.86e+11	5.03e+11	0.97	0.04
Co-60	7.44e+07			
H-3	2.07e+09	2.23e+09	0.93	0.06

	Measured g/kg U	Origen-S g/kg U	Ratio Meas/Calc	Error +/-
U-233	<0.01	2.32e-07		
U-234	3.39e-02	4.22e-02	0.80	0.44
U-235	1.63e+00	1.63e+00	1.00	0.02
U-236	8.01e-01	8.28e-01	0.97	0.04
U-238	9.83e+02	9.83e+02	1.00	0.00
Pu-238	5.76e-03	5.53e-03	1.04	0.06
Pu-239	2.69e+00	2.73e+00	0.99	0.03
Pu-240	1.22e+00	1.25e+00	0.98	0.04
Pu-241	1.34e-01	1.38e-01	0.97	0.09
Pu-242	9.40e-02	1.01e-01	0.93	0.06

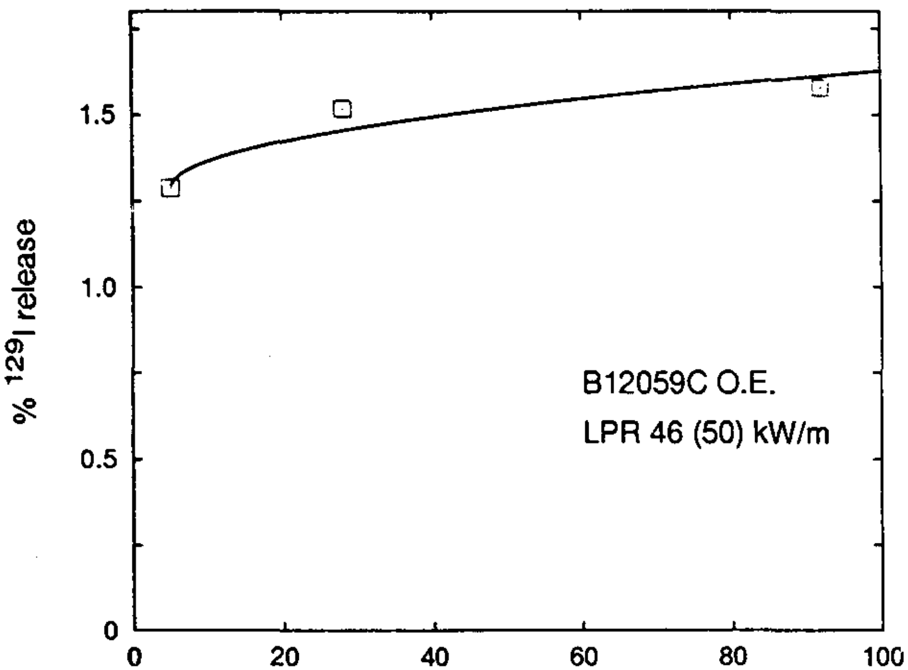


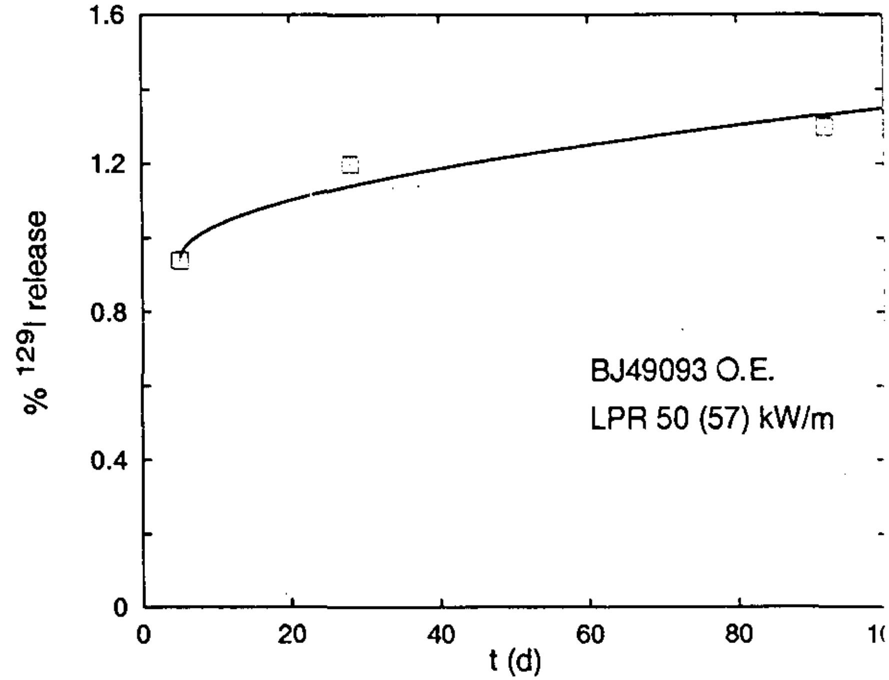


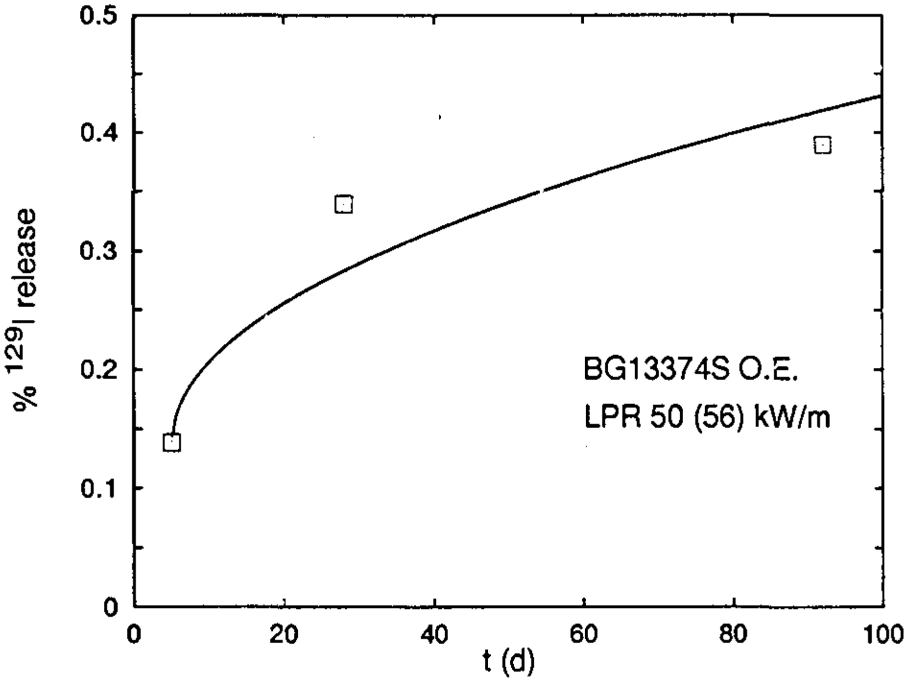
FRACTURE SURFACE OF HIGH LINEAR POWER CANDU FUEL



H. B. Robinson spent fuel. 30000 MWd/MTU
Transmission electron micrograph showing fission gas bubbles at grain boundary
250000X
L. E. Thomas, Westinghouse Hanford Co.



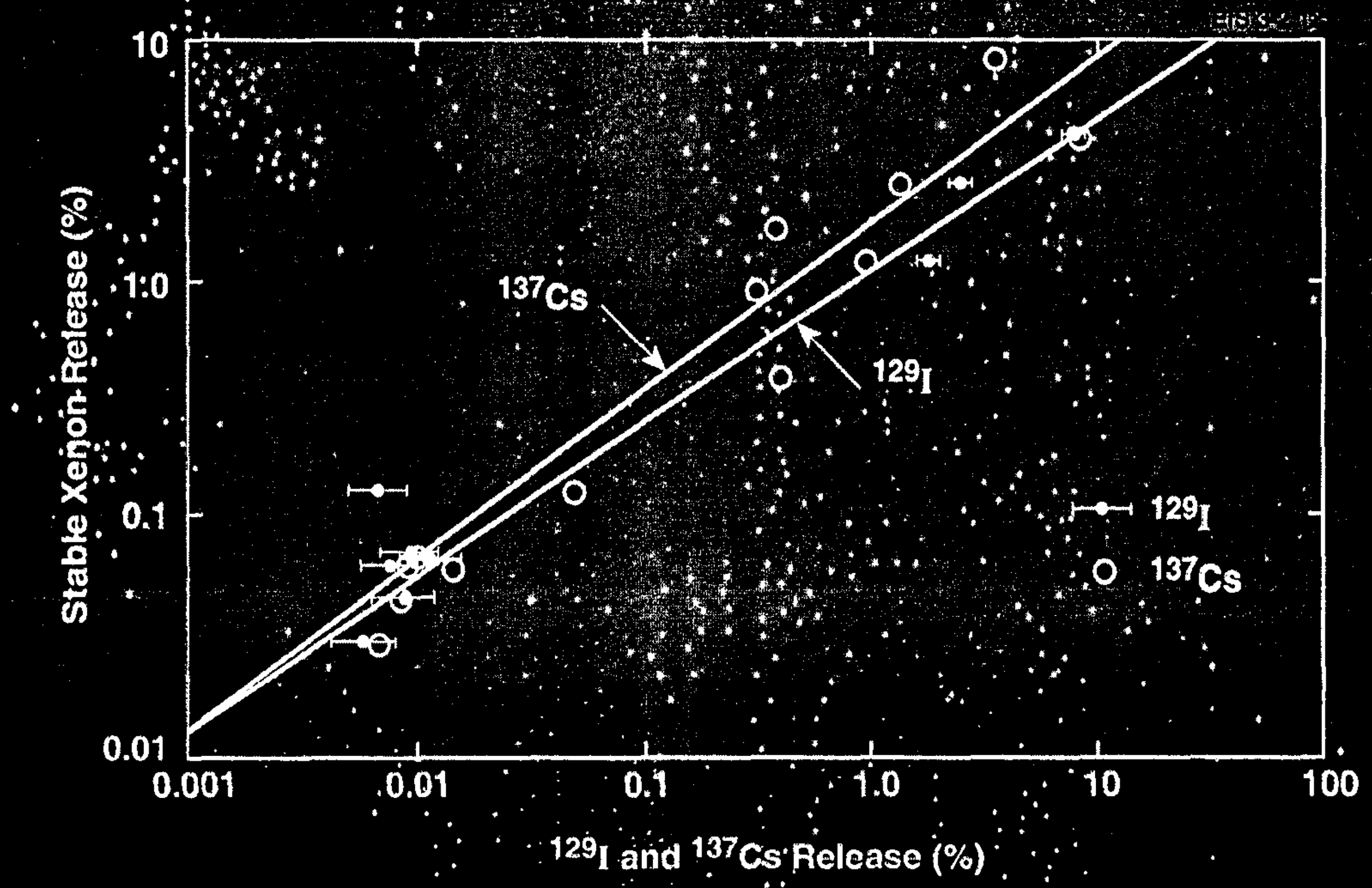




Kinetics of ^{129}I release

from used CANDU fuel segments for fuel with average LPR ≥46 kW/m and peak LPR ≥50 kW/m

Fitted to $I_t = I_5 + a\,(t\text{-}5)^{1/2}$



Stable Xenon Release (%)
10
1.0
0.1
0.01
0.001
0.01
0.1
1.0
10
100
137Cs
129I
129I
137Cs
129I and 137Cs Release (%)