UT-BATTELLE

Processing of Spent TRISO-Coated GEN IV Reactor Fuels

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Session: Knowing and Improving Resource Quality — Spent-Fuel Processing, Partitioning and Transmutation

2004 ANS Annual Meeting: A Golden Anniversary — A Golden Opportunity

Pittsburg, Pennsylvania

Tuesday June 15, 2004

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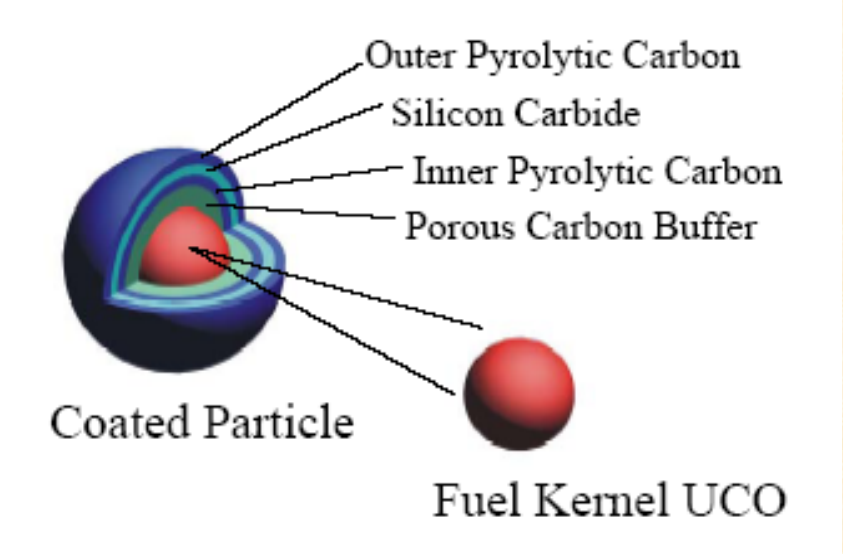
Coated-Particle Fuels are Under Development for the NGNP (GEN IV)

- Durable fuel form for HTGRs/AHTRs that support
  - High-temperature operations with high efficiency
  - HT processes (e.g. H_2 production)
  - Much higher burn-up than LWRs
    - Initial higher enrichment
    - Outperforms metal-clad fuels
- Excellent characteristics for transmutation and Pu-burning reactors
  - Supports deep-burn strategy
  - Neutron transparent
  - Supports full Pu or MOX core

Configuration of HTGR/AHTR Fuels
—General Description—

- Fuel construction begins with small kernels
- Coatings are applied for fission product containment
- Coated particles are mixed with a carbon-based mastic to form fuel compacts ("rods")
- The compacts are loaded into machined graphite, prismatic fuel elements ("blocks")
- A large amount of carbon is associated with a small amount of fuel
  - Unique challenges to processing



Particles

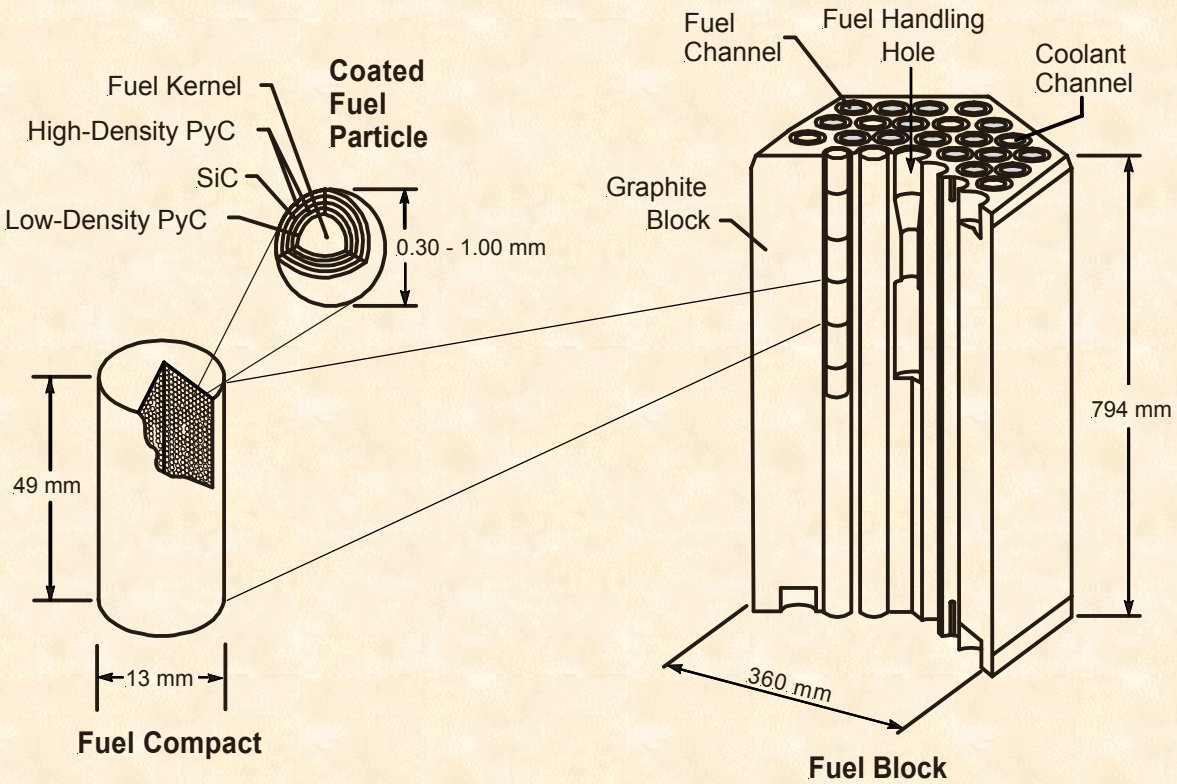
Compacts

Fuel Elements

Processing of Coated Particle Fuels —e.g. TRISO-Coated Fuels—

- Purpose is to recover
  - Actinides for recycle
  - Off-spec or defective fuel for recycle
  - TRU actinides and long-lived FPs for transmutation
- Challenges
  - Fuel particles encapsulated in inert coatings
  - Little fuel material in a sea of carbon
  - Alternative head-end required to separate fuel material from graphite matrix
  - Interfacing to spent LWR fuel separations, or equivalent technology, required

HTGR/AHTR Fuels
—Dimensions and Physical Properties—

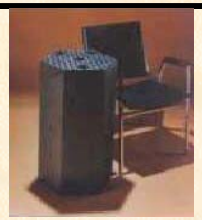


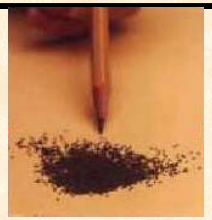
Leads to……

HTGR/AHTR Fuels
—Composition and Material Balances—

	Mass in one fuel element (kg)*		
Component	Complete Element	Compacts†	Particles
Graphite	90.00	0	0
Filler C	12.14	12.14	0
Pyro C	6.89	6.89	6.89
Porous C	1.72	1.72	1.72
SiC	4.63	4.63	4.63
Fuel + FPs	5.68+1.25	5.68+1.25	5.68+1.25
TOTAL	122.31†	32.31	20.17

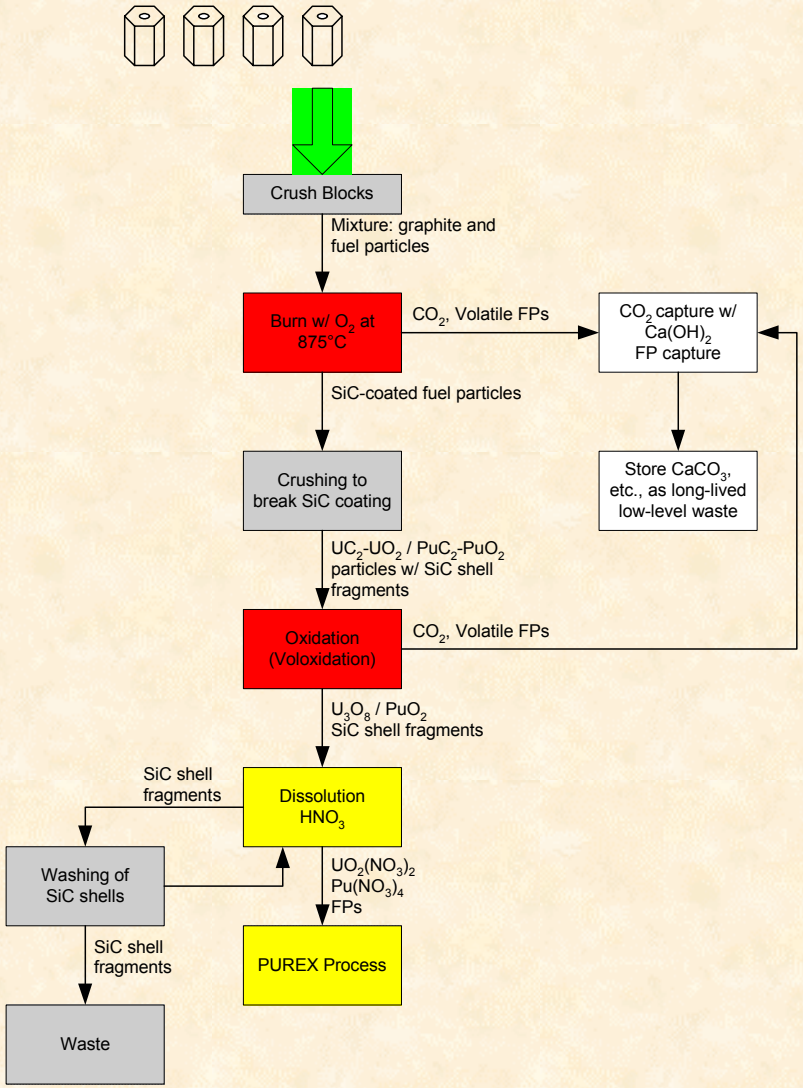
*Assume: 20% initial enrichment; after burn ~18% fission product by weight and 2.5% enriched in ^{235}U.





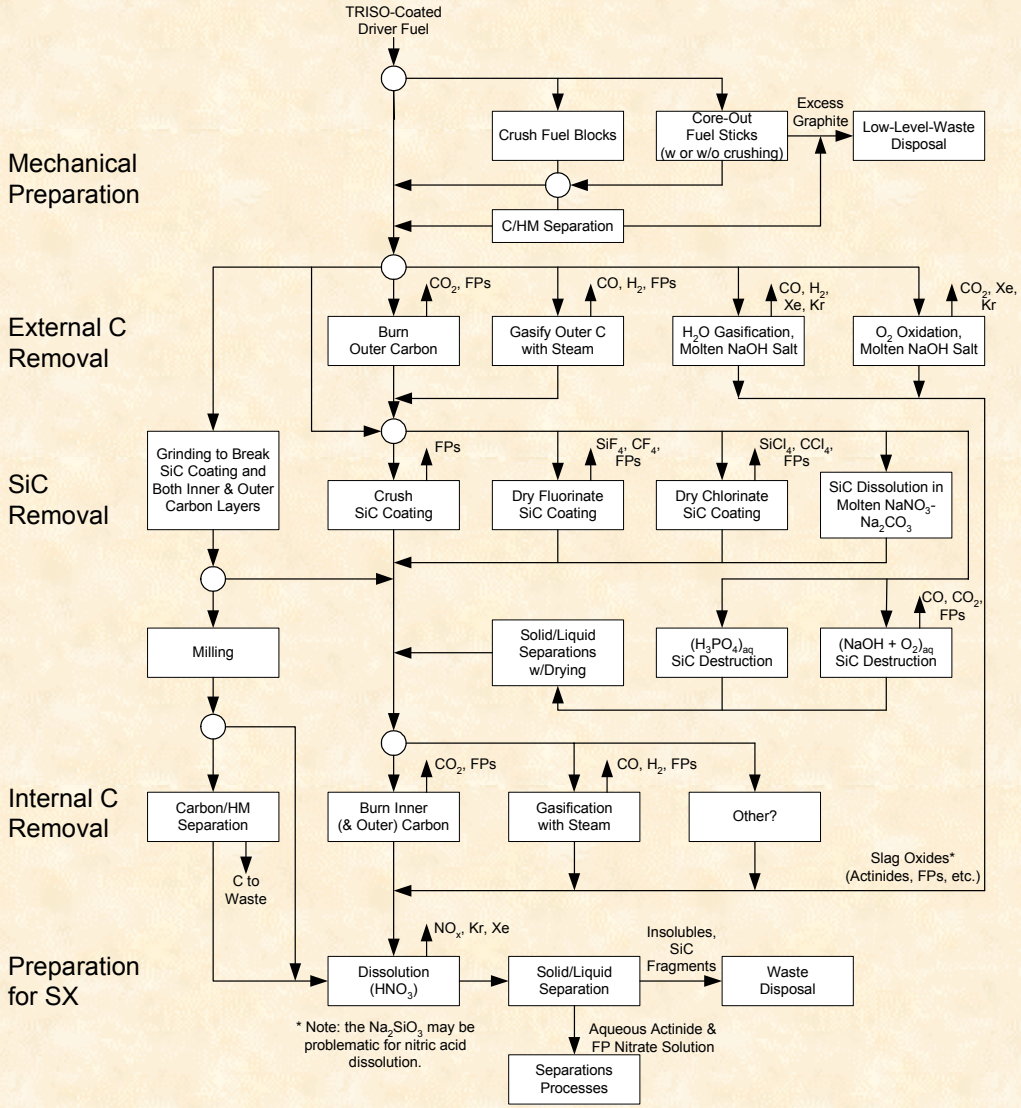
†Mass of C in the compacts alone is 20.75 kg.

Traditional Methods
—Crush-Burn-Leach—



- **HTGR fuel processing ca. 1970**
  - **Crush**
  - **Burn (filler/outer carbon)**
  - **Crush SiC coatings**
  - **Burn (inner carbon)**
  - **Dissolve oxides**
- **Thought to have few problems**
  - **CO_2 disposal to atmosphere thought acceptable pre-70s**
  - **Readily dissolved oxide product**
- **Problems**
  - **Large quantities of CO_2**
  - **^{14}C disposal to atmosphere**
  - **Large off-gas processing equipment**
  - **Capture greatly increases waste mass and volume (e.g. $CaCO_3$)**

Many Processing Options Identified
—From Studies on Processing Deep-Burn Fuel—

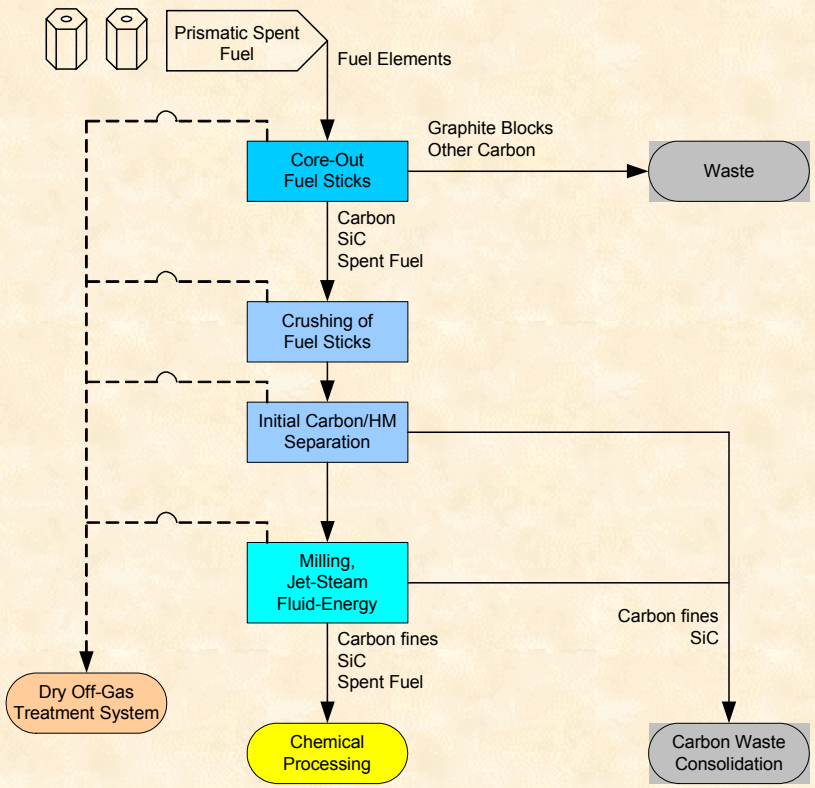


Ref. ORNL/TM-2002/156

Dealing with the Carbon in the Most Efficient Way Is Key

- Need to process about 120 kg of C to recover the fuel (<6 kg) in each fuel element
- A mechanical head-end is needed to separate the bulk of the C from the spent fuel
  - Remove compacts from prismatic block
- Combining the C component with other elements should be minimized
  - Much larger amount of waste for disposal (CO_2, CF_4, CCl_4, etc.)
  - Gaseous forms of C are very expensive to sequester
    - Geological co-sequestration of CO_2 and Kr by deep-well injection may be an economical alternative
- Two promising alternatives to crush-burn-leach identified
  - Grind-leach
  - Carbochlorination

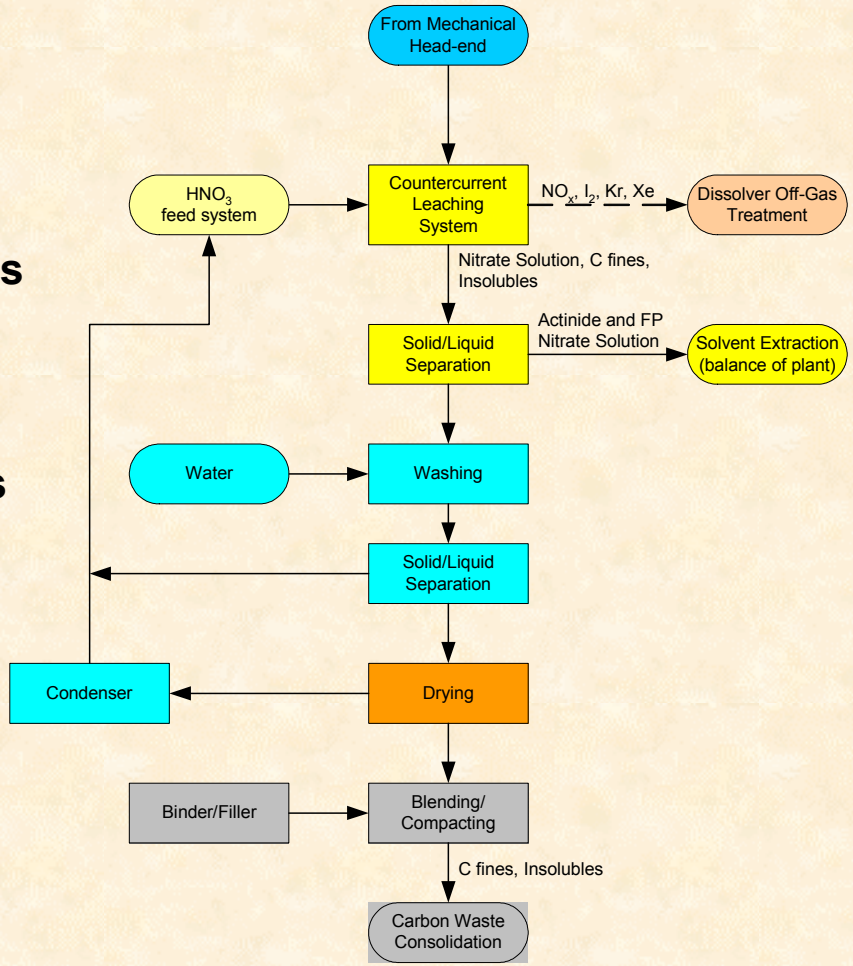
Mechanical Head-End
—Common to Best Approaches—



- Coring to remove fuel sticks
  - Eliminates bulk of carbon (graphite block)
  - Is a demonstrated technology
    - Limited data from a GA test
- Crushing fuel sticks to remove coated fuel particles
  - May permit more separation of C
  - Jaw crusher and roller mills have been successful
  - Further development recommended
- Fluid energy mill
  - Existing technology
  - Ensures breaking SiC shells
  - Has some separation capability, to be demonstrated

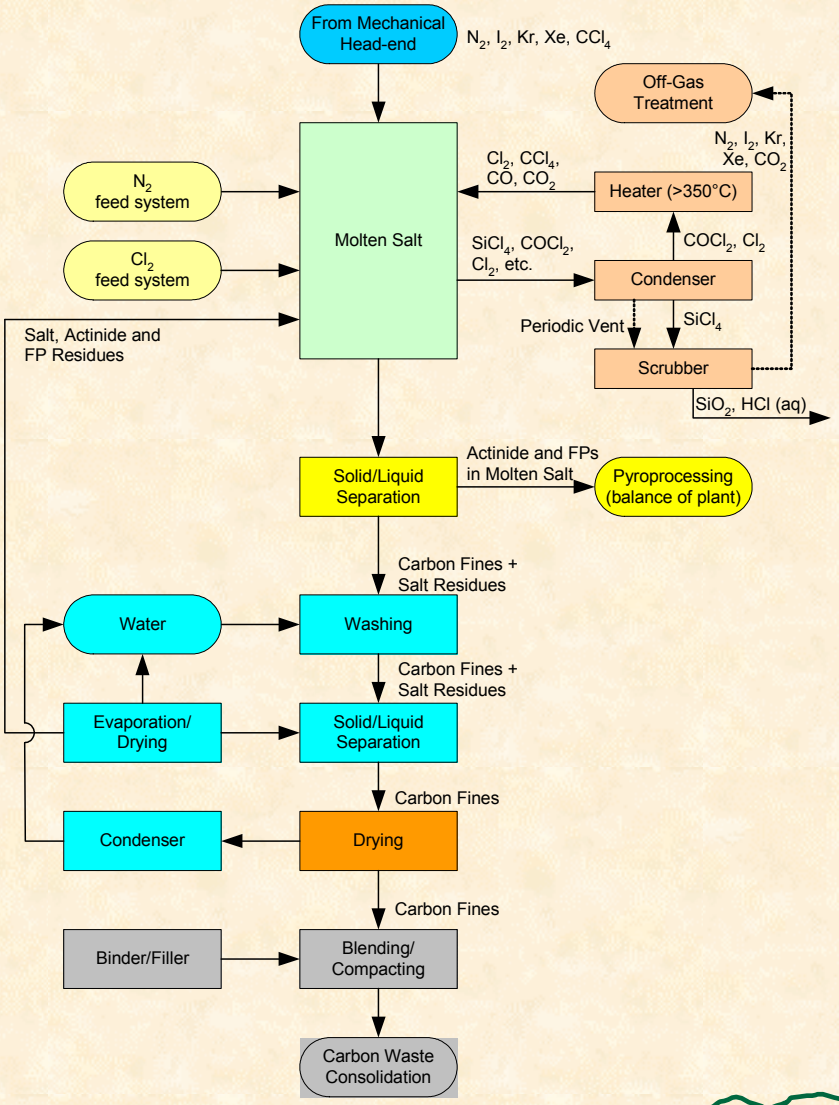
Two Promising Options Identified
—1. Aqueous Processing Option—

- Many process steps are industrial operations
- Acid leaching of fuel and FP carbides/oxides
  - Carbon is effectively inert
    - Multifunctional carboxylic acids problematic only with metal carbides (e.g. Mellitic acid)
- Carbon remains as a waste form
  - Will contain insoluble noble metals
  - Binder used to make monolithic waste form
  - Other wastes may be added
    - further consolidation
  - Waste form very resistant to leaching
- Aqueous nitrate solution to standard aqueous processing



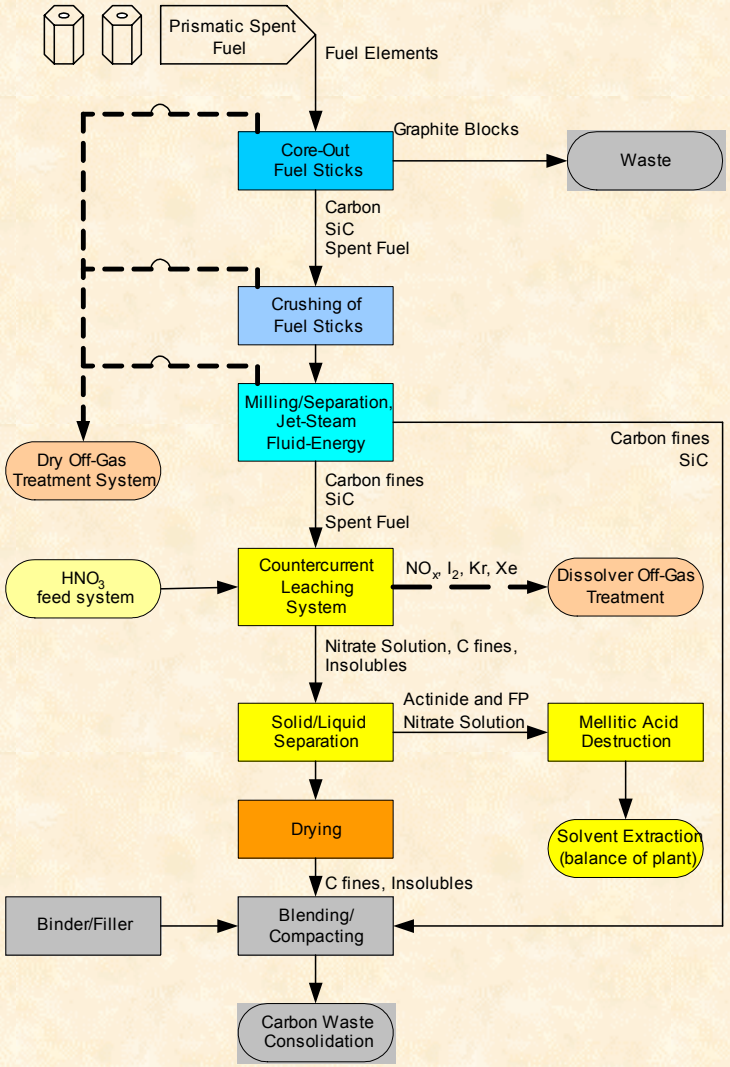
Two Promising Options Identified
—2. Pyroprocessing Option—

- Carbochlorination separates the carbon and fuel components
  - Small fraction oxidized to CO_2
  - Efficient mechanical head-end reduces CO_2 production
  - SiC is destroyed
  - Molten salt provides fast kinetics
  - Water washing removes residual salts
  - Process development and demonstration are required
- Carbon waste form as before
- Actinide- and FP-bearing salt to a pyro-chemical process
  - e.g. spent LWR fuel pyro-process

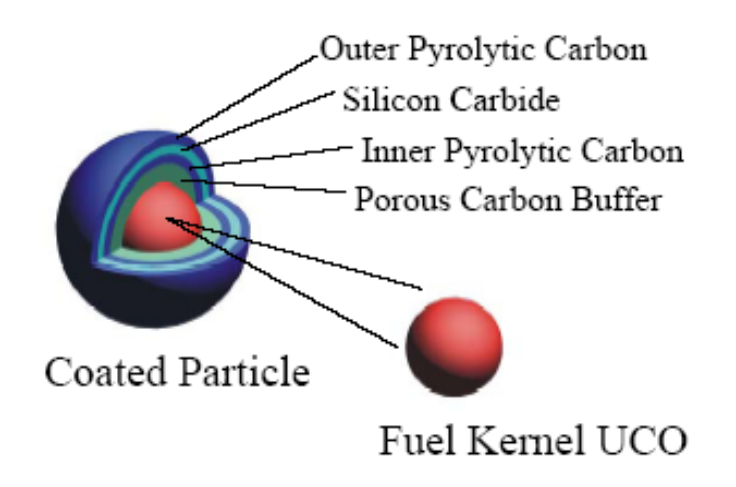


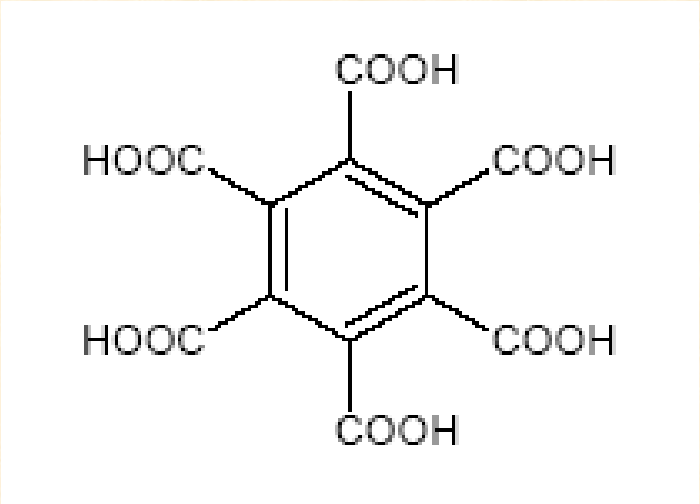
Aqueous Treatment Process
—Concept Selected for Further Study—

- Harvest fuel compacts
- Process to recover fuel
  - Crushing and grinding
  - Optional solid-solid separation
  - Leaching with nitric acid
    - Adaptation of commercial graphite purification process
  - May require steps to destroy carboxylic acids
  - Fuel laden acid solution routed to standard aqueous process
  - Residual carbon processed into waste form
- Methods result in smallest volume of carbon waste



Focus of Initial Research
—UCO Fuel Processing—





- **Dissolution of UCO fuel**
  - Some organic acids may form
    - Known to occur with mono- or di-carbide fuels
    - Buffer carbon may contribute in a lesser manner
  - Evaluate extent of problem
- **Evaluate separation of carbon**
  - Solution recovery
  - Carry-over
  - Stage efficiency
- **Address organic acid problems**
  - Acts as a foaming agent
  - May interfere with solvent extraction
  - An ozonation, or similar, step could destroy the mellitic acid

Summary

- Traditional processing of HTGR fuel reviewed
- New and promising alternative methods identified
- Waste reduction goals favor retaining elemental carbon
- Two flowsheets were identified for further development
  - Aqueous based: relies on industrial carbon-processing technology
  - Pyrochemical based: relies on well-developed thermo-chemical processes
- Selected options produce a highly inert carbon/graphite waste form
- Potential problematic areas identified for resolution