MASTER

HEDL-SA-2252

CONF-801209--8

SPENT-FUEL-STABILIZER SCREENING STUDIES

N. Wynhoff
S. E. Girault
R. L. Fish

November 1980

Second Annual NWTS Information Meeting
December 9-11, 1980
Columbus, Ohio

HANFORD ENGINEERING DEVELOPMENT LABORATORY
Operated by Westinghouse Hanford Company, a subsidiary of
Westinghouse Electric Corporation, under the Department of
Energy Contract No. DE-AC14-76FF02170
P.O. Box 1970, Richland, Washington 99352

DISCLAIMER

TABLE OF CONTENTS

FIGURES

TABLES

SPENT FUEL STABILIZER SCREENING STUDIES

I. INTRODUCTION

The National Waste Terminal Storage (NWTS) program was established by the Department of Energy (DOE) to provide facilities for permanent disposal of nuclear wastes. Disposal in mined geologic repositories is presently considered the most viable method for isolating radionuclides in spent fuel assemblies from the biosphere. Currently, emphasis is being placed on developing a multibarrier waste package to serve as an isolation barrier and in part, as a handling device. The waste package consists of the spent fuel assembly, a stabilizer,* an enclosing canister and additional barriers that may include an overpack, sleeves, coatings, and an emplacement hole backfill. This multibarrier waste package concept is illustrated in Figure 1.

An understanding of the compatibility of candidate waste package materials, candidate geologies, and spent fuel under disposal conditions is important for package design as well as for evaluating radionuclide retention capabilities of the waste package system. The Spent Fuel Stabilizer program is specifically concerned with the compatibility of the stabilizer material with the waste form and candidate canister materials under expected conditions imposed by the disposal cycle. The objective of this program is to identify, test, select, and qualify stabilizer materials for use in the design of spent fuel waste packages. Throughout the program, emphasis is placed on the selection of stabilizer materials having minimal interactions with both spent fuel and canister materials.

*The stabilizer is a material used to fill the void space, wholly or partly, in a canister containing a spent fuel element.

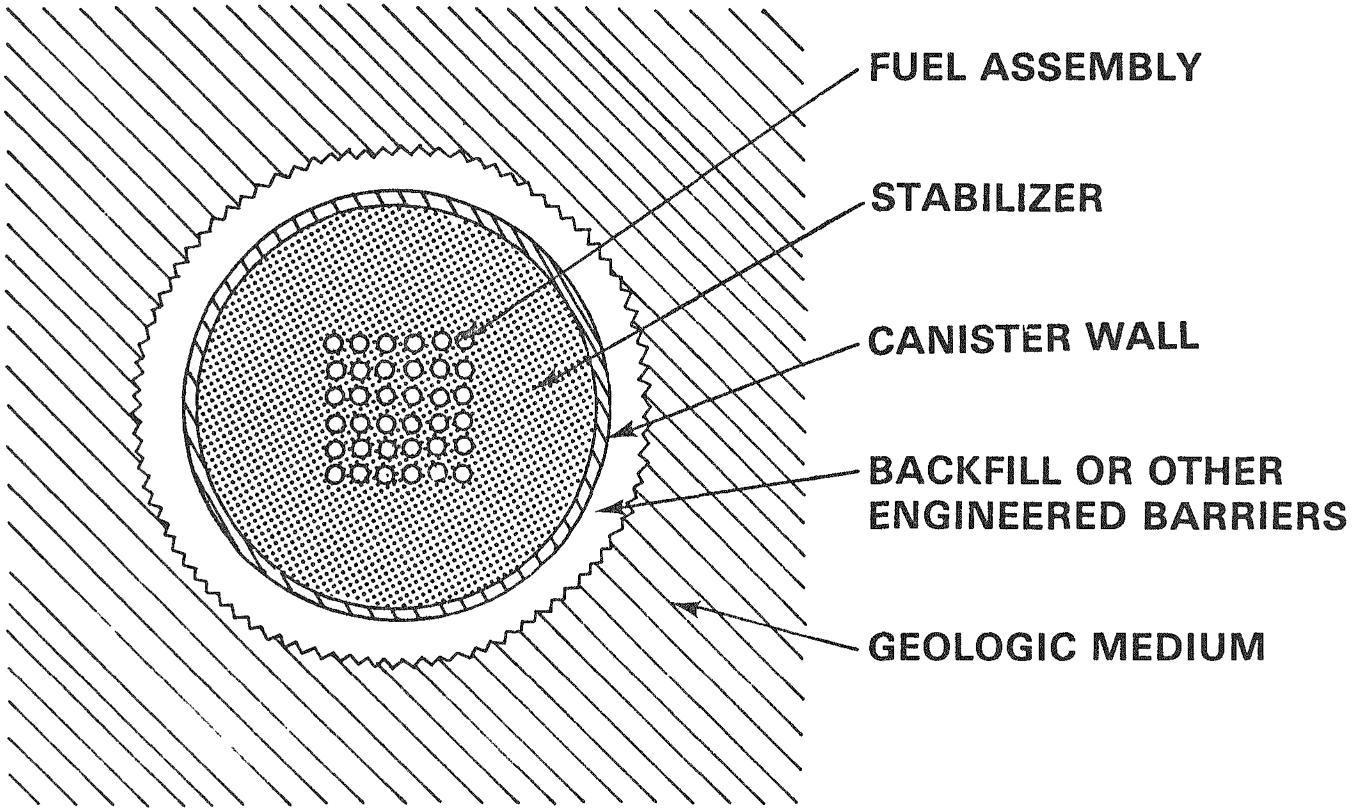

Figure 1. Schematic of Multi-Barrier Waste Package Concept.

This paper describes a study aimed at identification and screening of a broad range of potential stabilizer materials. As a result of this effort, a reduced list of recommended candidate materials was generated as well as a framework for the screening of any potential stabilizers identified at a later date. Subsequent testing will generate interaction data for further screening and qualification of stabilizer materials for use in the design of waste package systems.

II. METHODOLOGY

A. Preliminary Stabilizer Material Selection

To identify and screen potential stabilizer materials, a framework was established by first defining functions the stabilizer may be expected to perform. Potential performance functions and other considerations of interest are presented in Table 1. No attempt was made to place importance factors on these functions nor to order them relative to importance. The intent was to identify functions for the sole purpose of developing the screening framework. Material properties and attributes relevant to the performance functions were then identified. These included such properties as tensile strength, thermal conductivity, material interaction which degrades the cladding or canister, melting point, and material costs. Based on the potential performance functions and relevant material attributes, 72 stabilizer materials were identified for further evaluation and screening. The preliminary list is given in Table 2.

A primary requirement for development of spent fuel waste packages is that the stabilizer not degrade either the spent fuel cladding or the canister. For this reason, relatively inert gases were among the first materials suggested for consideration as stabilizers. As a potential barrier, functions that the stabilizer might perform include providing structural rigidity and improved package corrosion resistance. Therefore, solid and particulate stabilizers must be considered as well.

TABLE 1

STABILIZER PERFORMANCE FUNCTIONS AND OTHER CONSIDERATIONS

POTENTIAL PERFORMANCE REQUIREMENTS

*1. Help resist collapse of canister from lithostatic pressures.

2. Maintain the spent fuel assembly geometry and minimize criticality concerns.

3. Improve overall "corrosion resistance" of the waste package.

4. Impede radionuclide migration.

5. Promote heat transfer from fuel assembly and minimize fuel temperature.

6. Attenuate radiation from fuel assembly.

OTHER CONSIDERATIONS

1. Material costs and availability.
2. Loading process costs and feasibility.
3. Potential health hazards.
4. Gas generation potential.

*This function is important in salt and shale geologies. It is thought unimportant for "hard rock" geologies such as granite or basalt.

TABLE 2
POTENTIAL STABILIZER MATERIALS

Alumina
Silica (Amorphous)
Mullite
Zirconia
Rutile
Zircon
Silica (Quartz)
Graphite
Salt*
Basalt*
Granite*
Shale*
Tuff*
Sand

Bentonite[†] (in combination with any of the above particulates)
Bondate[††] (in combination with any of the above particulates)

Concrete (Cement + Aggregate)
  Portland Cement Type I
  Portland Cement Type IV
Air
Helium
Nitrogen
Aluminum Casting Alloy A413
Aluminum Casting Alloy 336
Commercial Aluminum
Copper Casting Alloy 3A
Copper Casting Alloy 8A
Copper Casting Alloy 13B
Commercial Copper
1% Antimonial Lead
Calcium Lead
Commercial Lead
Zinc Alloy AG40A
Zinc Alloy AC41A
Zinc-Copper-Titanium Alloy
Commercial Zinc
Soda Lime Glass (0079,0080,0081)
Lead Glass (0120, 1990)
Borosilicate Glass (7063)
Soldering Glass (1416, 7570, 7575, 7593, 7594)

*Source of stabilizer material would be excavated repository.

[†] Bentonite is a natural clay that swells with the adsorption of water and has good ion exchange properties.

[††] Bondate is a patented, chemical bonding agent for aggregates and fibers, marketed by Bondate Industries, Inc.

B. Evaluation of Materials

Attributes for each material under consideration were compiled on reference data sheets to the extent that data was available in the open literature and from material vendors. The format used is illustrated in Figure 2. Analytical evaluations were identified and performed to provide a basis for ranking and screening these stabilizer materials. All performance functions in Table 1 were addressed with the exception of resistance to canister collapse. In soft rock media (salt and shale) where rock creep results in external crushing forces exerted on the waste package, it has been shown that package integrity can be maintained by at least two different package designs. One design utilizes the stabilizer to prevent collapse. The second provides a borehole sleeve, and therefore eliminates the need for the stabilizer to prevent collapse. An analysis of materials cost and availability was also performed.

C. Screening Technique

After each analytical evaluation, the materials were ranked based on the results of the analysis. For those evaluations where a reasonable basis exists, specific screening criteria were established. Those materials that satisfied all screening criteria are recommended for further testing and assessment. For evaluations where no criteria were established, the materials have simply been ranked and not eliminated. Flexibilities in this screening technique allow additional materials to be considered using the same evaluation analyses and application of respective screening criteria. Alternative or additional criteria can be applied to results of any evaluation as appropriate information becomes available from waste package designers. This approach provides flexibility for further selection of materials and consideration of different screening criteria while still providing consistent results.

FIGURE 2

SPENT FUEL STABILIZER PROGRAM DATA SHEET

MATERIAL

ELEMENT							
COMPOSITION %							

NET IMPORT RELIANCE		HEALTH/ENVIRONMENT HAZARDS
MAJOR FOREIGN SOURCES		
WORLD RESERVES		NOTES
MATERIAL COST		

PROPERTY	DENSITY	TENSILE STRENGTH	COMPRESSIVE STRENGTH	TOTAL ELONGATION	THERMAL CONDUCTIVITY	THERMAL EXP. COEFFICIENT		
UNITS	g/cm^3	psi	psi	%	watt/cm °C	μ in/in/°C		

III. STABILIZER INTERACTION AND COST ANALYSES

A. Thermal Gradient Analysis

To assess the effectiveness of potential stabilizers for promoting heat transfer and minimizing fuel temperatures, a model configuration was assumed and the thermal gradient was determined for each material. For this model, a single, intact, pressurized water reactor (PWR) fuel assembly was assumed to be located in the center of a 13-inch inside diameter (ID) canister, 15 feet in length. A thermal power level of 0.964 kW, considered maximum for a PWR assembly five years after reactor discharge, was assumed in order to provide a conservative basis for the thermal gradient analysis

Figure 3 shows the temperature difference between the cladding of the hottest (central) fuel rod and the canister as a function of stabilizer thermal conductivity, with the assumption that effects of radiation and convection are negligible. This applies to solid stabilizers and, with less confidence, to particulate stabilizers with helium fill. Effective thermal conductivities for the particulate stabilizers were calculated assuming that particles are spherical and that equal contributions are made by series and parallel heat transfer. The maximum cladding temperature as a function of canister temperature is given in Figure 4 for helium and nitrogen stabilizers. Effective thermal conductivities of these gas stabilizers were estimated from this relationship in conjunction with information from Figure 3.

B. Stress Analysis

A solid stabilizer can provide isolation enhancement for spent fuel during disposal by preventing intrusion of water and containing gaseous and volatile fission products in the event of localized canister breach. A fractured solid stabilizer could not perform these functions. Tensile stresses induced by expected thermal conditions

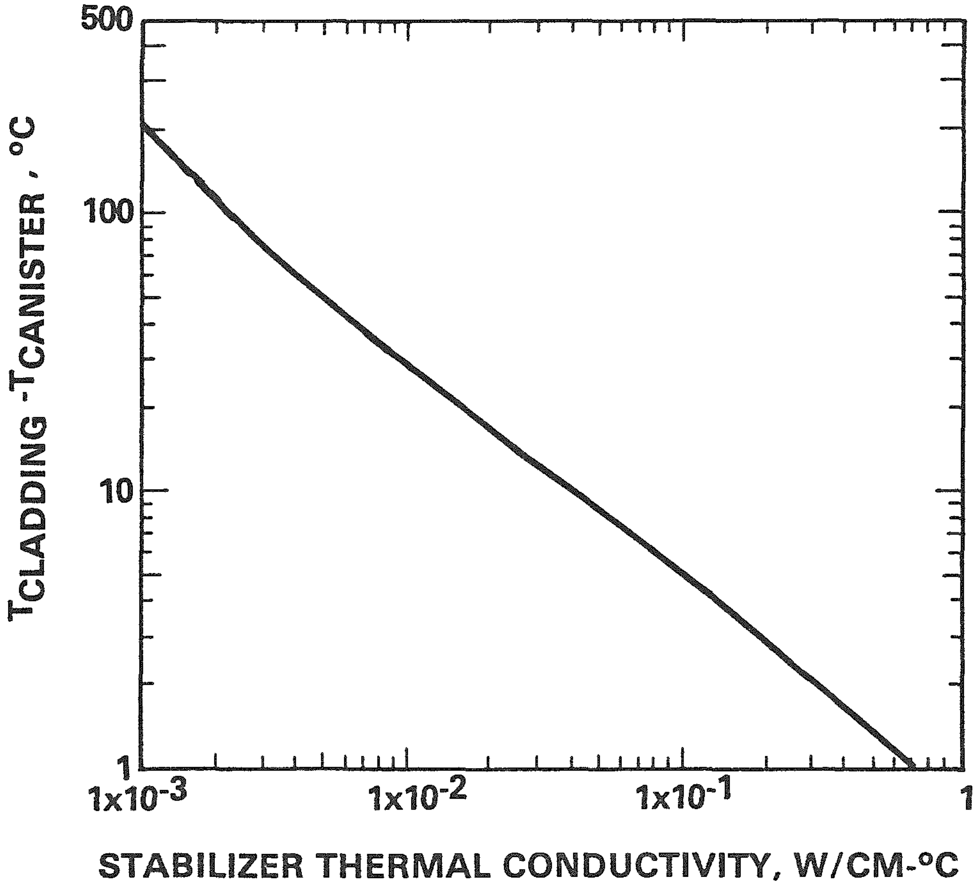

Figure 3. Temperature Difference Between Cladding of the Hottest Rod and Canister vs. Thermal Conductivity of Stabilizer (5 year old spent PWR fuel).

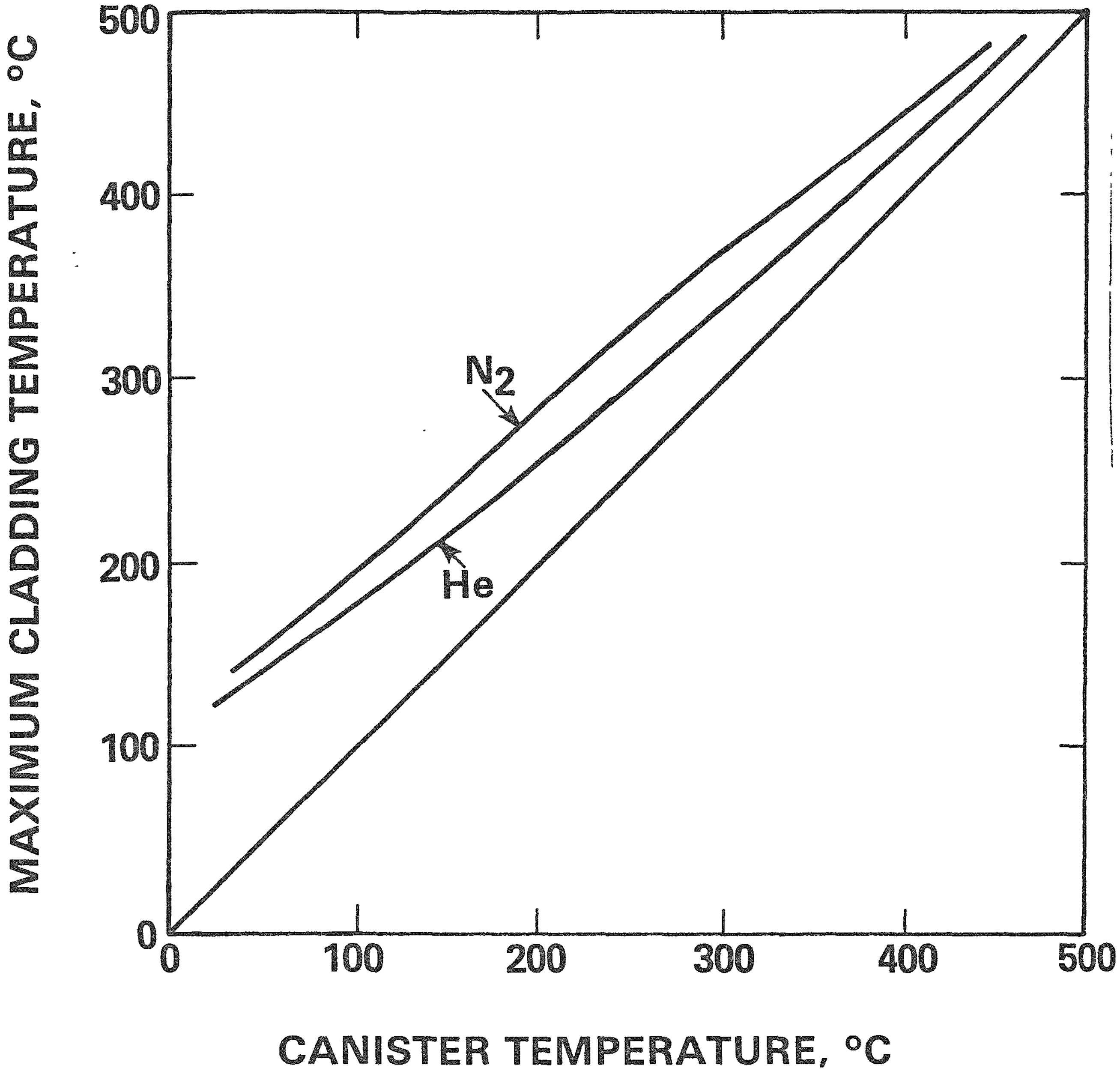

Figure 4. Maximum Fuel Cladding Temperature vs. Canister Temperature for He and N_2 Stabilizers (5 year old spent PWR fuel)

were calculated to permit screening of solid materials which may fracture during waste package preparation or disposal. Two analyses were performed: (1) A calculation of tensile stresses induced by the thermal gradient was made for all potential solid stabilizer materials; (2) An analysis of tensile stresses induced during cooldown by differential thermal expansion between the cladding and stabilizer was made for stabilizer materials which require melting during the fill process.

For brittle materials, such as glass and cement, a direct comparison of induced stress with material strength provides a basis for screening. Since stresses in metals can be relieved or reduced by plastic deformation, credit can be taken for their inherent ductility. Therefore, if the calculated stress (assuming the strains to be elastic for simplicity) induced in a metallic stabilizer exceeded the ultimate tensile strength of the metal, the elastic strain required to induce that stress was calculated and compared to the actual strain capacity of the metal. The strain capacity was taken as tensile elongation of the stabilizer material.

1. Thermal Gradient Stress Analysis

For screening purposes, tensile stresses induced by thermal gradients were calculated by considering the stabilizer to be a long, hollow cylinder. Thermal stress in a thick-walled cylinder was calculated assuming a cylindrical spent fuel assembly, a uniform radial temperature distribution with respect to the axis of the cylinder, a constant temperature along the length of the axis, and free expansion of the stabilizer cylinder. Since the canister may restrain expansion, this assumption will provide conservative results. Dimensions used were those for a PWR fuel assembly and a canister with a 13-inch ID.

2. Differential Thermal Expansion Stress Analysis

Tensile stresses may be developed in a cast, solid stabilizer by differential thermal expansion between the cladding and stabilizer during cooling from the high temperature fill process. These stresses were evaluated by determining stresses developed around a single fuel rod in a cast, solid material. This model assumes isolation of each fuel rod and its uniform stabilizer layer from all other fuel rods. The following basic assumptions were also made:

- Above the strain temperature for a glass or half the solidus temperature for an alloy, all stresses are relieved through strain accommodation. Below these temperatures, there is no stress relief.

C. Nuclear Criticality Analysis

In response to the stabilizer performance function of minimizing criticality concerns, the effect potential stabilizers would have on nuclear criticality was assessed. KENO IV, a Monte Carlo criticality code along with a 16 group Hansen Roach cross section library, was used to calculate K_{eff}, the multiplication factor. It was assumed that the stabilizer completely fills all void space in a thirteen inch canister with an intact PWR fuel assembly in place with no neutronic interaction between canisters. Calculations were based on the probable case of a spent fuel assembly with a burnup of 28,500 MWd/MTU at 5 years after reactor discharge.

D. Radiation Attenuation Analysis

To address radiation attenuation, the shielding effectiveness of potential stabilizers was addressed. The gamma dose rate at 12 detector locations around a spent fuel canister surface was calculated by the point kernal code, QAD-P5A. The stabilizer material was assumed to fill all void space in a thirteen inch diameter canister with an intact PWR fuel assembly in place. The fuel assembly was assumed to have a burnup of 28,500 MWd/MTU and a time since discharge of 5 years. The gamma source spectrum was computed by the ORIGEN isotope generation and depletion code.

E. Cost and Availability Analysis

Due to the concern for maintaining reasonable material cost and availability, the cost per waste package was calculated and an analysis of material availability was made for each potential stabilizer.

To calculate the cost per canister, a right circular cylinder canister with a 15-foot length and one-foot interior diameter was assumed (empty canister volume 10.4 $ft.^3$). A Westinghouse 17 x 17 array PWR assembly was taken as the typical spent fuel assembly with a calculated volume of approximately 2.97 $ft.^3$ This volume includes fuel rods, control rod guide tubes, and the instrument tube, but omits items such as spacer grids and orifice plates. A conservative canister void volume of 7.43 $ft.^3$ was thus obtained for the amount of stabilizer material per canister.

The material cost per canister, based on this volume, material densities, and on industrial average prices was calculated. For particulate materials, densities were adjusted to 70% of the bulk density to reflect the estimated void volume that will exist between particles even when tightly packed. For gas stabilizers, it is expected that the pressure in the canisters will be low (100 psi maximum). This maximum pressure requires a volume of 50.5 $ft.^3$ of gas per canister at 21°C. Costs for gas stabilizers were then calculated based on industrial average cost per cubic foot.

For this preliminary screening effort, the determining factor used in assessing material availability was import reliance. Dependence of domestic industry upon foreign suppliers is undesireable. The U.S. Bureau of Mines defines and computes a net import reliance value which is based on quantities imported and exported. This information was used to evaluate materials on a comparative scale. When stabilizer materials are selected and qualified for spent fuel waste packages, it will be important that this complex consideration be more completely evaluated.

IV. INTERPRETATION OF STABILIZER INTERACTION AND COST ANALYSES

A. Thermal Gradient Evaluation

Since no spent fuel canister temperature is currently available and the maximum allowable cladding temperature of 380°C is currently recommended on an interim basis only, no specific thermal gradient criterion has been established. The thermal gradient analysis provided relative information on the effectiveness of potential stabilizer materials in minimizing fuel temperature by heat transfer. The results also provided input to the evaluation of tensile stresses induced by the thermal gradient.

B. Stress Evaluation

Based on the general criterion of precluding fracture of the stabilizer, the following stress criterion was established: Calculated tensile stress induced in a potential stabilizer shall not exceed the material strength. If this condition is violated for metallic stabilizer materials, the calculated strain shall not exceed the tensile elongation. The material strength was taken as the design strength for glass stabilizers, as ultimate tensile strength for metals and as tensile strength for cement and Bondate.

Applying this stress critertion to results from analysis of tensile stresses induced by thermal gradient, it is recommended that cement and Corning glasses 7570, 1416, 0120, 0081, 7575, and 1990 be rejected from further consideration.

For stresses induced by differential thermal expansion of the stabilizer and cladding, the stress criterion was applied to Zircaloy-4 cladding with a check of Type 304 stainless steel to determine if additional materials should be rejected. Examination of the results with Zircaloy-4 indicated the following materials do not meet the stress criterion: Corning glasses 0081, 0120, 1990, 1416, 7570, and 7575, commercial zinc, commercial copper, 1% antimonial lead, calcium lead, commercial lead, and commercial aluminum.

An inspection of Type 304 stainless steel results indicated no additional materials fail this stress criterion. Tensile elongations for the metals are significantly greater than the calculated elastic strains. Therefore, it is recommended that only the six glasses listed above be rejected, using this criterion.

C. Nuclear Criticality Evaluation

To preclude nuclear criticality, the following criterion was established: Calculated K_{eff} shall not exceed 0.95. This is considered adequately subcritical. Application of the criterion to the results indicated that none of the potential stabilizer materials interact with the spent fuel assembly in a manner to create a criticality concern.

D. Radiation Attenuation Evaluation

Currently no gamma dose rate limit has been established for the spent fuel canister. Therefore, no criterion for gamma dose rate was established. The results of this evaluation provide only relative shielding effectiveness information for potential stabilizer materials.

E. Cost and Availability Evaluation

Since the stabilizer is only a portion of the multibarrier waste package, it is logical that its cost should be relatively low. It has been estimated that it will cost the utilities $200,000 to dispose of each fuel assembly.(18) From this figure, an arbitrary maximum stabilizer cost of $5000 per canister (2.5% of total cost) was established. Applying the cost criterion results in a recommendation to reject all glass stabilizers currently under consideration.

To screen materials on the basis of material availability, potential stabilizer materials were recommended to be rejected from further testing if a known net import reliance of 80% or greater exists. When this criterion is applied, it is recommended that alumina, aluminum, aluminum alloys, and rutile be rejected from further consideration.

F. Other Considerations

In considering corrosion resistance, it is recommended that stabilizers with water soluble components having known serious corrosion effects on cladding be rejected from consideration. For example, a crushed salt stabilizer will form a chloride solution with any residual moisture and both zirconium alloys and stainless steels exhibit serious pitting corrosion or stress corrosion cracking in chloride solutions. It is, therefore, recommended that salt be removed from further consideration as a stabilizer material.

V. RECOMMENDATIONS

Potential stabilizer materials initially identified are summarized in Figure 5(a). Seventeen materials failed to satisfy all screening criteria and are not recommended for subsequent testing. Two of these materials are particulates; therefore, bentonite and Bondate combinations with these particulates were also rejected. This reduces the number of materials under consideration to fifty-one.

Since initial experimental work will focus on further screening of materials, it was deemed adequate to test bentonite and Bondate with representative particulates rather than with all particulates. The following particulates are considered as representative: (1) silica-quartz, mullite and zirconia; (2) basalt, because it is the only candidate geology with a presently known likely site; and (3) graphite. Graphite will be tested with Bondate only. The combination of bentonite with graphite seriously reduces the heat transfer effectiveness of graphite without providing any significant advantage. This reduces the number of stabilizer materials to thirty-four, as summarized in Figure 5(b). The complete list of candidate stabilizers recommended for subsequent testing and screening is given in Table 3.

FIGURE 5

SUMMARY OF POTENTIAL AND RECOMMENDED CANDIDATE STABILIZER MATERIALS

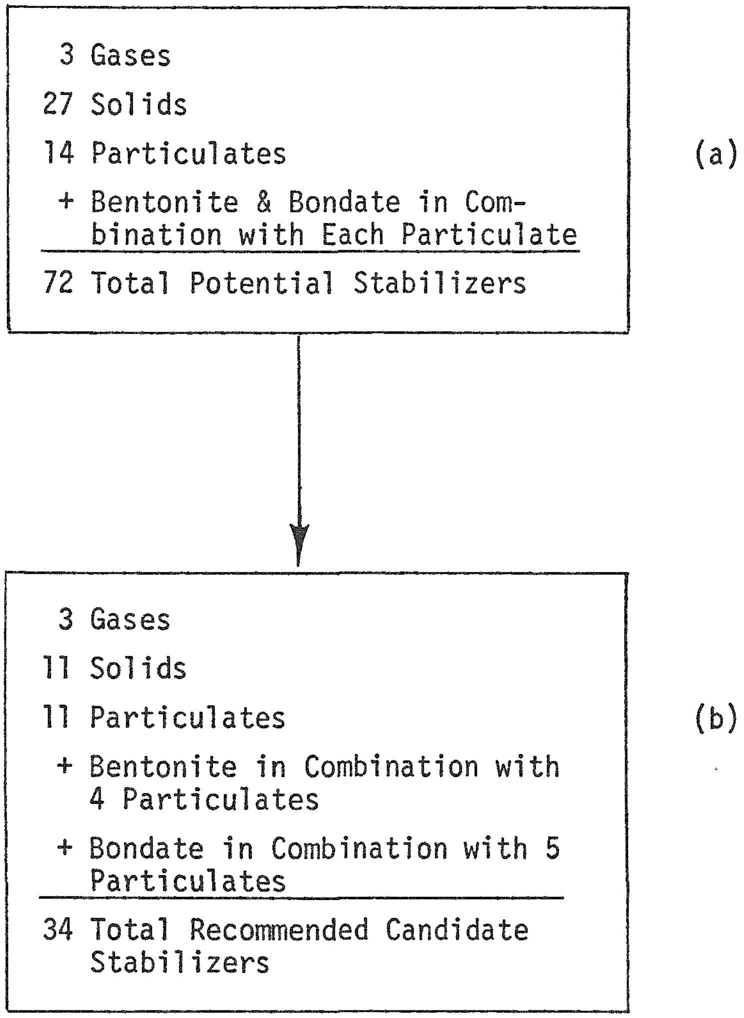

TABLE 3
RECOMMENDED CANDIDATE STABILIZERS

silica - amorphous
silica - quartz
silica - quartz/bondate
85% silica - quartz/15% bentonite
mullite
mullite/bondate
85% mullite/15% bentonite
zircon
zirconia
zirconia/bondate
85% zirconia/15% bentonite
*basalt
basalt/bondate
85% basalt/15% bentonite
*granite
*shale
*tuff
†sand
graphite
graphite/bondate
air
He
N_2
1% antimonial lead
calcium lead
comm. lead
Zn alloy AG40A
Zn alloy AC41A
zinc-copper-titanium alloy
comm. zinc
copper casting alloy 3A
copper casting alloy 8A
copper casting alloy 13B
comm. copper

*Geologies will be tested when a candidate repository site is identified by ONWI.

†Sand will be tested when a large, stable, industrial scale supply is identified.