APPLICATION OF HIGH LEVEL WASTE-GLASS TECHNOLOGY TO THE VOLUME REDUCTION AND IMMOBILIZATION OF TRU, LOW LEVEL, AND MIXED WASTES (U)

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ABSTRACT

The EPA has designated vitrification as the "Best Developed Available Technology" for immobilization of high-level nuclear waste. Recent public pronouncements and private conversations with the EPA indicate that the agency is considering long-term waste immobilization as a preferred mode of treatment for many types of hazardous waste. It is concluded that a program for adaptation of HLW vitrification technology to other wastes is clearly needed, and would have a high confidence of making major beneficial impact on the safe and efficient disposal of mixed and transuranic wastes.

INTRODUCTION AND SUMMARY

The EPA has designated vitrification as the "Best Developed Available Technology" for immobilization of high-level nuclear waste.(1) Recent public pronouncements and private conversations with the EPA indicate that the agency is considering long-term waste immobilization as a preferred mode of treatment for many types of hazardous waste.

Slurry Fed Melters (SFM) have been developed in the U.S., Europe, and Japan for the conversion of high level radioactive waste (HLW) to borosilicate glass for disposal.(2-7) Laboratory and pilot scale operations have been conducted to develop equipment, glass compositions, and control methods. The melters developed fall into four categories: batch melters, continuous pot melters, Joule-heated ceramic-lined melters, and stirred melters. The newest design, the stirred melter,(7) combines the high production rates and high glass quality features of the Joule-heated melters with the low-cost, compact, simple maintenance of the pot melters.

The Savannah River Laboratory has been active for many years in developing glass compositions and melter technology for high-level wastes at the Savannah River Site (SRS). This same expertise could be applied to other waste forms presently being stored on the SRS and other nuclear sites.

The goals of this paper are as follows:

- determine whether and how DOE's vitrification technology can be brought to bear on other types of waste
- identify promising candidate waste types for vitrification
- identify research and development programs needed

Consequently, this paper reviews suitable waste types produced at SRS, evaluates whether the available HLW technology is appropriate to the immobilization of these wastes, and outlines development programs necessary to demonstrate the immobilization.

It is concluded that a program for adaptation of HLW vitrification technology to other wastes is clearly needed, and would have a high confidence of making a major beneficial impact on the safe and efficient disposal of Mixed and Transuranic wastes. The high probability of success is based on the conclusion that, for the most part, wasteforms and equipment of the type necessary have already been developed as part of the pilot scale development and testing for HLW immobilization. Non-HLW wastes generally have no or relatively low penetrating radiation levels. Thus, treatment of these wastes is possible by adapting equipment and glass compositions developed for the treatment of High Level Waste to use in facilities with contact maintenance. This would produce a stable, transportable and low-release-rate waste form, thereby increasing the safety of shallow burial, engineered, or geological disposal. Many types of mixed Resources Conservation and Recovery Act (RCRA) wastes can be delisted, permitting environmental compliance and risk management to focus on the radioactive hazards.

Similarly, the program to develop borosilicate glass waste forms for HLW has investigated a broad range of silicate glass compositions which are adaptable to other waste types.

DISCUSSION

Available Melter and Glass Technology

The Defense Waste Processing Facility (DWPF) at the U.S. Department of Energy's Savannah River Site has an anticipated operating life of over 16 years, using a remotely replaceable melter with a minimum design life of 2 years. Similar equipment and facility development efforts are underway for HLW disposal at the West Valley Demonstration Project (WVDP), Hanford Waste Vitrification Project (HWVP), and in France, Belgium, the Federal Republic of Germany, and Japan. As part of this process, a number of melter systems of various capacities have been designed, constructed, and operated. Most of these systems have been operated and maintained in non-radioactive and non-hazardous environments as part of equipment development

and demonstrations. However, several large scale pilot and production units have been operated with hazardous melter feeds, radioactive spikes, or HLW. Design, construction, and operation of these facilities provides a basis for the evaluation of the technology of volume reduction and immobilization of other wastes in similar facilities.

The scope of the melter development at Savannah River has been restricted to the immobilization of SRS high-level nuclear waste. However, SRS, as is true of other U.S. Department of Energy facilities, also produces large amounts of non-high-level radioactive wastes which have relatively low penetrating radiation levels. It is important to make the best use of the available technology developed for high level radioactive waste to aid in the safe and efficient disposal of these wastes. Therefore, this study considers how the glass technology developed for HLW might be applied to the immobilization, volume reduction and stabilization of Transuranic (TRU), Pu^{238} (alpha), low-level, and chemical (RCRA) wastes, as well as mixed radioactive/chemical wastes (mixed wastes). Wastes of these types generally do not require heavy shielding to control radiation doses to operating and maintenance personnel, permitting direct (hands on) maintenance. However, these wastes share many contamination and environmental release concerns with high level waste. Thus, it can be expected that they would require similar wasteform quality and production control practices as the high-level wastes, but the scale of operations and equipment design would differ.

Potential benefits of the application of the existing waste glass technology to the disposal of other waste types include:

- well characterized waste form stability and extremely low release rates in a variety of environmental conditions
- excellent mechanical and thermal stability
- no combustible or pyrophoric properties
- retention of essential release properties even if disturbed or mechanically damaged during storage or disposal
- low generation of potentially respirable particles
- the ability to accept high loadings of heavy metals
- ability to accommodate fluctuating waste types with negligible effect on release properties
- ability to combust limited amounts of organics (including carcinogens)
- total destruction of asbestos
- scale of facility adaptable to accommodate a wide range of disposal needs.

An important indication of the waste retention capabilities of waste glass is a dimensional analysis of the respective waste leach tests used for radioactive waste glass, commercial glass, and hazardous wastes.(8) It can be inferred from this analysis that typical waste glass is 1,000 to 100,000 times more durable than is necessary for the delisting of hazardous chemical wastes. Typical waste glass durabilities are comparable to granite, basalt and other durable rocks.(9) In similar studies, soda-lime-silicate glass melters have been demonstrated for the immobilization of glove box wastes.(10-12) These glasses are expected to have durabilities within a factor of four of those developed for HLW. Thus, it is practical to consider the construction of a facility for converting certain mixed chemical/radioactive wastes into stable radioactive wasteforms that do not exceed RCRA criteria. This could have significant impact on the cost of disposing of such wastes, and could also minimize radiation doses that are expected because of certain surveillance procedures (such as sampling of sumps) necessary for the storage of mixed wastes that would not be needed for wastes delisted to low-level radioactive waste.

The HLW disposal programs have gone a long way in addressing the concerns for the other waste types. In many ways HLW disposal is the most difficult, since it combines the requirement of very durable and stable waste forms with remotely maintained equipment. The other waste types require various levels of contamination control, but none would require large remotely maintained facilities. Thus, the waste glass formulations developed for HLW should be applicable, as well as the materials of construction, but the lack of need for remote maintenance should permit more flexibility in the design and construction of the various melters and related equipment.

Melter Designs Applicable to the Vitrification of Non-High Level Wastes

The majority of the current melter programs at SRS are in support of the operation of the DWPF with the existing melter design, with possible evolutionary design changes. However, these and similar programs at other sites provide much of the necessary basis for the development of Non-HLW Glass Melters.

The first waste glass melters were designed for batch operations, and were a direct increase in scale from crucible tests. This approach was found unsuitable for HLW production facilities because of slow melt rates caused by slow heat transfer from the external heaters through the canister into the reacting batch. Lack of agitation and lack of temperature uniformity made it difficult to homogenize the glass. Calcination of feed before introduction into the canister increased the melt rate, but also increased the tendency for crystal formation in the glass and entrained waste in the calciner off-gas system. This method was finally eliminated for HLW based on the large number of melters, operating in parallel, required to meet the production rates necessary

to dispose of HLW inventories. The method remains attractive for small facilities where melt rates are not restrictive, such as the at-line vitrification of Pu wastes. Subsequent improvements in melter materials and glass compositions(13-15) permit the slow production of small amounts of waste glass with satisfactory durabilities.

The second class of melters developed were continuous pot melters. In this type the melt rate was increased by increasing the diameter of the pot, by direct heating of the pot by radio frequency induction heating, and by continuous feeding of raw materials. Glass homogeneity was improved by using gas bubblers to agitate the melt. The largest of this type of melter is the French AVH system which melts 25 kilograms per hour. This is the processing rate limit per pot melter using dried feed. With slurry feeding the melter capacity limit would be about one half of this, or 12 kilograms per hour per melter. The melter design temperature is limited to about 1150°C by creep resistance of the Inconel™ alloy used for the pot. The use of this system was not practical in the DWPF because of the large number of parallel melters, calciners, and off-gas scrubbing systems required. However, this class of melter is modularized, with parts that are relatively easy to replace. An additional benefit of this approach is that only the failed components need be replaced, minimizing the amount of waste generated with melter changeout, and maximizing the useful life of each component. This approach therefore has merit when dealing with homogeneous wastes, and where melt rate restrictions are not limiting. For non-homogeneous wastes the close coupling of the calciner/incinerator functions with the melter is not desirable, since it is necessary to classify the calcined waste to remove large metallic pieces.

The third category of melters are the joule-heated, ceramic-lined melters. This is the result of several generations of melter development based originally on commercial, electrically heated melters. Major differences between HLW Melters and Commercial Melters have been:

- the use of metal shells to contain glass contact refractories and thermal insulation
- development of specialized slurry feeding and glass pouring systems
- the use of nickel based alloys for electrodes carefully matched with glass composition control.

In this type the melter is lined with refractory, and the glass is directly heated by conducting electricity through the melt. This system with slurry feeding has been selected for all the production melter systems in the U.S., W. Germany, and Japan because of high production rate and high glass quality. The size of these systems is effectively limited only by operating facility constraints (e.g. cell space, crane capacity), since all the structural support is provided by a room temperature metal box which contains the refractory. The Inconel 690™ alloy electrodes only need to be self-supporting, and high current densities are possible on the faces of the electrodes.(16) Therefore, nominal melt temperatures can be as high as 1150°C, which is only 200°C lower than the alloy melting point.(13) Glass production rates are proportional to the surface area of the melt, but convection caused by the joule heating is enhanced as the size of the melter is increased, so larger melters have proportionately higher melt rates. Small laboratory melters operate below 22 kilograms per hour per square meter, production melters operate at about 39.1 kilograms per hour per square meter. Melt rates can be doubled by dry feeding. The combination of higher temperature and convective mixing makes the glass very homogeneous. The major difficulty in slurry-fed ceramic-lined designs is the large number of individual refractory bricks, supporting shell and other components that must be assembled to make this type of melter. This complexity increases the melter construction, installation, and disposal costs. In radioactive service only limited repair is possible, so failure of individual components can require removal and disposal of the entire assembly.

Melt reaction studies, computerized staged reaction models, organic combustion models and melter redox models are being developed to support the DWPF melter, and have led to the development of a fourth class of melters, known as Advanced or Stirred Melters. Evaluation of the melt reaction sequence indicated that the melter operating temperature need not be as high as the nominal 1150°C to assure adequate glass durability.(17) This made possible the consideration of melter designs where Inconel 690™ components carry dynamic loads, which is not possible with the present nominal operating temperature of 1150°C.

Several melter design companies were consulted to determine what commercial technology might be applicable. The most promising of these was a proposal by Associated Technical Consultants to develop mechanically stirred melters comparable to those originally investigated by Owens-Illinois Co..(6) This approach offered the possibility of combining the size of the continuous pot melters with the high production rates of Slurry Fed Melters: Owens-Illinois demonstrated a compact 0.57 cubic meter melt chamber that produced 10,900 kilograms per day of partially melted commercial glass from raw materials. It features a simple geometry, with a simple mechanical drive system, plus rapid start, drain and restart capabilities, all of which are desirable properties for radioactive service.

The existing Owens-Illinois design did not meet the requirements of radioactive waste disposal because it:

1. required natural gas combustion to start,
2. was constructed with electrode and agitator materials chemically incompatible with waste glass feeds,

3. produced partially-reacted foam rather than fully-reacted, dense glass,
4. could not guarantee that unreacted batch would not be delivered with the final product,
5. had restricted electrode surface area, and
6. used refractory lining, sharing scale up, glass sealing and disposal concerns with the existing ceramic-lined melters.

A major uncertainty in attempting to apply stirring technology to waste vitrification was the effect of stability of slurry feeding on the melting process. Instabilities might cause an uncontrolled amount of entrainment in the off-gas system, result in glass freezing on the agitator, or have uncontrolled flow from the output spout.

It was determined that necessary characteristics of a practicable agitated waste-glass melter are:

1. adequate durability of glass product,
2. all electrical heating,
3. use of electrode and agitator materials compatible with oxidizing melts,
4. use of alloys with known creep characteristics for predictable melter life,
5. maintenance of temperatures and stresses below creep rupture conditions,
6. elimination of porosity in product glass,
7. stable glass flow with slurry feeding of raw materials,
8. self-sufficient startup power from resistance heaters,
9. vapor space resistance heaters available for melt rate stabilization,
10. vapor space temperature above 600°C to combust organics,
11. ability to drain the tank, and
12. predictable melter life greater than 6 months.

To minimize glass sealing concerns, as well as the scale up, disposal, and repair costs, it is desirable to have all glass contact materials out of one metal alloy rather than a mixture of alloys and refractories. Based on high chromium alloys corrosion studies,(15) waste melter operating experience, and limited creep rupture and creep strength data,(13) it was specified that all glass contact materials be constructed of Inconel 690™ with a maximum operating temperature of 1075°C.

It was determined that for a first attempt at slurry feeding a stirred melter, the melter should have about 0.093 square meter (1 square foot) of melt surface. This is a typical size for laboratory waste melters, but is about the largest size that can be easily installed and maintained in a glove box, and is therefore production scale for glove box waste disposal. With increased melt rates, this size melter can be considered a pilot scale melter comparable to the Scale Glass Melter for facilities such as the DWPF.

A melter was designed and constructed to these criteria, and tested with simulated HLW slurry.(7) To minimize costs, the melter did not attempt to include mechanical design details required to seal the melter for radioactive service, or to make the melter easily repaired. The design focused on demonstration of the combination of slurry feeding and stirred melting with HLW glass components and melter materials.

The measured slurry melt rates with the new design are 155 kg m^{-2} h^{-1} with agitation, and 19.5 kg m^{-2} h^{-1} without agitation. This demonstrates an increased melt rate by a factor of 8, similar to the factor calculated from the Owens-Illinois stirred melter tests of commercial glass from raw materials.(6) The demonstrated melt rate was limited by the ability of agitation to disperse the slurry as it was sheared by the underlying foam. Estimated electrode current densities indicate that an additional factor of 2 increase is possible before electrode current is limiting melt rate. It is therefore concluded that melt rates of about 290 kg m^{-2} h^{-1} are possible through design optimization. Glass was homogeneous and essentially fully dense. Its durability was the same as would be expected for glass held for longer than four hours at temperature.

Since this design uses all metal glass contact materials, only two melter pieces contact the molten glass. Replacement of the tank or agitator is possible, minimizing disposal volumes and costs. Thus, this design combines the most desirable features of the continuous pot melters and the slurry-fed ceramic-lined melters.

Identification of Non-High Level Waste Types Suitable for Vitrification

Low-Level, beta-gamma wastes are generally disposed of in cement waste forms, or deep burial of stabilized waste, because their hazards are similar to those of the uranium ore from which these wastes originated. These wastes result primarily from uranium operations which produce reactor fuel and targets for plutonium production. At this time, the additional expense of general vitrification relative to cement stabilization does not appear to be justifiable for simple hazardous or LLW. If the Low Level wastes are contaminated with alpha isotopes, have unusually high activities, or contain hazardous chemicals converting them to mixed wastes, then the added stability and leach resistance of vitreous wasteforms is very desirable, and should be economically justifiable (see Table I).

Mixed wastes, heavy metals, inorganics, asbestos, and organic wastes share many of the characteristics of beta-gamma wastes, but they contain RCRA listed chemical elements or compounds which require permanent isolation

from the environment. The high temperature melting process destroys the chemical compounds associated with such wastes, and ties poisonous elements up into a durable matrix.(11,12) It has been demonstrated that soda-lime-silicate and borosilicate glasses bind hazardous elements so that these wastes can be delisted based on TCLP leach test results,(8,11,12) allowing them to be treated as Low Level Waste.

Alpha wastes, (TRU) wastes are comparable to High Level Waste in biologic risk, but are usually small scale operations. Generally Pu^{238} and Pu^{241} are considered separately from other transuranic isotopes.(18) For this discussion Pu^{238} and Pu^{241} will be categorized as alpha wastes, and all other transuranics as TRU wastes. In these waste types major goals are to convert small volume, low activity and often flammable wastes into small waste forms that are easy to handle, but provide a high degree of contamination control (isolation). Waste glass is essentially a sealed source, which will reduce the dispersibility of alpha wastes during accidents, and transfers low levels of activity by contact. The high temperature melting process combusts organics, and reduces the volume of this waste category, which primarily consists of contaminated plastic, cloth and paper products.

TABLE I

SRS Mixed Wastes Suitable for Vitrification Purposes

Type of Waste	Hazardous Species	Amount Cubic Meters Inventory(annual)
Laboratory and Miscellaneous Wastes		-
Treatability Study Wastes		
Soil Borings and CMP Pit Soils	Pesticides	-
Waste Site Soils	Pb	
Soil Borings TNX Seepage Basin	Cr, Ag, CN, Hg	-
Ceramic Filter Elements and Beads	Cr,Pb,Hg	
IDMS Mixed Waste Waste Melter	Hg -	
Evaporator Solids	Cr, Pb, Hg	-
Influent Strainer Solids	Cr, Pb, Hg	-
Ion Exchange Resins	Cs, Pb, Hg	-
CIF Blowdown	Cd, Cr, Pb, Ni, Ag	(960)
CIF Ash	Cd, Cr, Pb, Ni, Ag	(170)
		230
M Area LETF Filter Paper	Cd,Cr,Pb,Ni,Ag,CN	(61)
		70
M Area LETF Sludge	Cd,Cr,Pb,Ni,Ag,CN	(20)
		640
M Area Sludge	Cd,Cr,Pb,Ni,Ag,CN	(125)
Low Level Alpha waste (< 100nCi/g)		3730
		(7)
Mixed TRU Waste		3040
		(21)
Soil Melter		(5775)

TABLE II

Determination of Preferred Melter System for Beta-Gamma (U, UE), LLMW, Inorganics (Heavy Metals), Asbestos, Organics, and Soils Wastes.

	Characteristics																
Melter Type	Raw Materials Cost	Waste Loading	Homogeneity	Melter Life	Melter Repair	Waste Variation	Product Sampling	Small Scale	Large Scale	Portability	Drain/Restart	Generic Licensing	Melter Costs	Melter Disposal	Various Feed Connections	Various Off-Gas Connections	**Total Rating**
Relative Importance	6	6	8	10	4	10	10	1	10	5	7	8	8	2	10	8	113
Calciner/Pot Melters	3	5	4	4	3	4	2	0	3	3	6	4	4	1	3	2	51
(Modified Commercial)	6	6	6	10	2	8	8	0	10	2	2	6	8	1	10	6	(91)
HLW Joule Heated Melter	3	5	6	10	1	6	8	0	8	3	2	6	3	1	8	4	74
(Stirred Melter)	5	6	8	4	4	7	9	1	6	5	7	6	6	2	6	4	(86)

(Indicates Preferred System)

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TABLE III

Determination of Preferred Melter System for Alpha, TRU, Mixed Alpha, Disarmament/Decommissioning, and Incinerator Wastes.

	Characteristics																
Melter Type	Raw Materials Cost	Waste Loading	Homogeneity	Melter Life	Melter Repair	Waste Variation	Product Sampling	Small Scale	Large Scale	Portability	Drain/Restart	Generic Licensing	Melter Costs	Melter Disposal	Various Feed Connections	Various Off-Gas Connections	**Total Rating**
Relative Importance	1	8	6	8	10	4	8	10	2	0	10	6	4	10	4	4	95
Calciner/Pot Melters	0	4	4	3	5	1	2	2	2	0	10	3	3	8	1	1	49
Modified Commercial	1	8	6	4	2	3	6	2	2	0	2	5	2	2	4	4	53
HLW Joule Heated Melter	0	6	6	6	2	3	7	4	2	0	2	5	1	2	3	3	52
(Stirred Melter)	1	6	6	6	8	3	7	10	2	0	10	6	4	10	3	3	(85)

(Indicates Preferred System)

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HLW laboratory melters are full scale demonstrations for these waste types.

If the nation's inventory of nuclear weapons is to be permanently reduced, then a means of denaturing and disposing of the weapon grade materials (Pu^{239} and Enriched U) must be found. One possible means of controlling these materials is to vitrify in a glass containing neutron poisons. Such a waste form would permit high Pu and U concentrations, permitting inventory and verification by calorimetry or radiochemical methods. Pu in such waste forms could be reconverted, but conversion would be slow and difficult making it unlikely that arms control agreements would be violated by reprocessing such materials. In this application nuclear criticality control and materials accountability would be major requirements, so a very small melter that is well mixed and totally drained between batches would be necessary.

Contaminated soils and similar contaminated concrete wastes result from the excavation, repair and decommissioning of Separations and Interim Waste Storage facilities. Contamination can be the result of hazardous chemical spills, or low levels of TRU or mixed fission products. Currently, these wastes are disposed of without further processing by shallow burial. Vitrification of these materials will bind the hazardous and radioactive species, reducing the risk of ground water contamination. Examination of in situ vitrification of SRS soils has indicated that the soils require the mixing in of fluxing agents, such as Na_2O.(19) Homogeneous addition and mixing is a simple task as a part of the addition of solids to a commercial style melter,(20) or as a separate stream to a stirred melter.(6)

Radioactively contaminated salts would result from the transuranic decontamination of metallic components via such techniques as molten salt fluxing followed by acid or steam cleaning. These soluble salts could then be incorporated in melter feed for final disposition in a waste glass.

Evaluation of TRU, Mixed and LLW Melter System Design Concepts

As with any program, costs have to be minimized. Maximum melter reliability, ability to operate at high rates to recover lost production time, and increased waste loading should be Waste-Melter goals. Cost savings can be achieved by either decreasing the frequency and length of equipment outages, or by increasing the instantaneous production rate. Increased waste loading not only increases utilization, it also reduces the amount of repository space required for waste disposal. This increased waste loading has higher potential cost savings than any of the other goals considered.

A calciner/pot melter system, a modified commercial melter, a modified HLW Joule heated melter, and an Advanced Stirred Melter system were considered for each of the waste types. Each characteristic was rated for its relative importance on a scale from 0 to 10. Each melter system was rated for all of the characteristics on a scale of 1 to 10, and then each melter system characteristic rating was multiplied by the relative importance factor to get the final system characteristic rating shown in the attached tables

CONCLUSIONS

Application of HLW technology to other waste types is expected to have a large beneficial impact, with a high expectation of success. The most beneficial programs would include:

1. A Laboratory Stirred Waste Melter - could be built, capable of processing depleted uranium. This would be required for treatability studies to support the following recommended programs. It could serve as the basis for a laboratory sized unit to dispose of miscellaneous mixed and hazardous laboratory wastes.
2. A TRU Waste Glass System - could be designed to be integrated with the TRU Waste Facility's Pu waste incinerator system. This would also provide the operating experience and detailed design required for installation of smaller melters in Pu facilities, so that Pu wastes could be size reduced, made nonflammable, and immobilized before they leave the production facilities. The melter should be designed based on a stirred melter.
3. A Mixed Low-Level Waste Melter Facility - could be created by modification of the existing Integrated Demonstration Melter System (IDMS). Initial operations would include the vitrification of sludges from SRSs M Area, a homogeneous mixed low-level waste. This would provide the necessary operating experience for subsequent treatment of heterogeneous low-level and mixed waste incinerator ashes from the Consolidated Incinerator Facility (CIF). The existing Scale Glass Melter might be used in this program, with eventual replacement by either an adaption of commercial melter systems, or a stirred melter.
4. A Soil Melter System - could be developed to convert, at high rates, the contaminated clays, sands, and concrete being stored in the burial ground into an inexpensive, high durability wasteform for onsite disposal. This melter system would be based on either an adaption of commercial melters, or a stirred melter.
5. Fused Salt Decontamination Method Development - would result in processes that could rapidly remove transuranic contamination from metallic components. Several salts should be regerable, with final disposition in waste glass. Demonstration of decontamination efficacy, salt composition, salt regeneration, and salt vitrification should be demonstrated on glove box scale units.

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