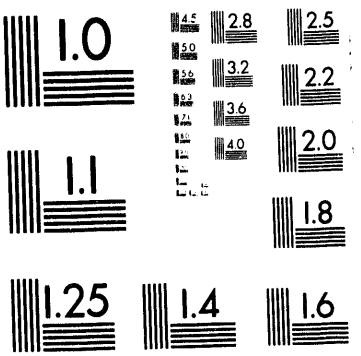
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DATABASE FOR WASTE GLASS COMPOSITION AND PROPERTIES

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ABSTRACT

A database of waste glass composition and properties, called the PNL Waste Glass Database, has been developed. The source of data is published literature and files from projects funded by the U.S. Department of Energy. The glass data have been organized into categories and corresponding data files have been prepared. These categories are glass chemical composition, thermal properties, leaching data, waste composition, glass radionuclide composition and crystallinity data. The data files are compatible with commercial database software. Glass compositions are linked to properties across the various files using a unique glass code. Programs have been written in database software language to permit searches and retrievals of data. The database provides easy access to the vast quantities of glass compositions and properties that have been studied. It will be a tool for researchers and others investigating vitrification and glass waste forms.

INTRODUCTION

Disposal of nuclear, mixed and hazardous wastes requires data on material properties and performance. The U.S. Department of Energy has a large investment in formulation and characterization of glass compositions for its wastes, and will need to invest further to accommodate new waste compositions. Readily available data from previous DOE programs is often difficult for regulators, researchers and interested parties to access even though about $100 million has been spent to obtain the data. Waste form developers, evaluators and basic scientists need access to this information so they can benefit from previous experience and avoid duplication of effort. The Pacific Northwest Laboratory (PNL) Waste Glass Database provides a comprehensive, convenient, and available resource to fulfill this information need.

The database serves as the primary source of electronically accessible data for radioactive, mixed and hazardous waste glasses. Data has been obtained from a variety of sources, such as published and unpublished reports, journal articles, project files and project data transmittals. Data are sought for waste compositions, glass compositions, process-related properties and disposal-related properties.

The purpose of this work is to collect the massive quantity of waste glass data in the literature and in laboratory files for compilation into a computerized file system. The DOE has spent several hundred million dollars on various waste management R&D programs to generate and report glass composition, characterization and performance data. The PNL Waste Glass Database collects and organizes this data so it is readily available for use in DOE's environmental restoration and waste management programs. Investigators in the DOE waste management system, government policy-makers, university researchers and other parties interested in waste treatment and disposal need a way to quickly access information on waste glasses pertaining to durability, radionuclide leaching, homogeneity, and thermal process properties. This information supports environmental analysis regulatory petitions, such as licensing, design of vitrification equipment and design of storage and disposal facilities. The need for such a tool will become more compelling as DOE proceeds with feasibility studies on vitrification treatment and disposal of the vast array of radioactive mixed waste (RMW) at various sites in the United States. The number of glass compositions to develop is large compared to the U.S. high-level waste program. Glasses for vitrification of mixed waste will also vary significantly from those for HLW. Borosilicate glass is used in the United States for HLW. For RMW, other glasses that are dominated by constituents such as calcia, alumina, calcium fluoride, magnesium fluoride, phosphate, and iron oxides as well as silica will likely be used.

Glass database activities at PNL originated with the Materials Characterization Center (MCC). Kindle and Kreiter (1987) describe the results of work to develop a comprehensive database for high-level nuclear waste package components, with the initial emphasis on waste glass and its properties. The activity ended in 1988 with the compilation of about 1300 records of glass leaching data and about 600 records of thermal and processing properties data. A relational database was developed, allowing users to conduct searches by glass

composition or by property. In 1991, it was decided to expand the database beyond high-level waste glasses to include all available glass data from reliable sources. The type of information to include has also been expanded. This continuing work is performed at PNL and funded by the DOE Office of Environmental Remediation and Waste Management.

The objective of the work is to extract data on glass composition, leaching and thermal properties from the literature and other reliable sources. This data will be transcribed and entered into a systematic computer file structure. The files will be made available to agencies and organizations in the United States through DOE.

The initial effort by the MCC was limited to only a certain portion of the literature and a time period of 1979-1988. The current effort will include all accessible and reliable data, published and unpublished. Unpublished data will be screened for completeness and flagged accordingly in the database to inform users of its origin. A commercial software language will be used to develop a relational database which allows users to search by glass composition, type of waste glass, thermal properties, and leaching properties.

DATABASE STRUCTURE

The waste glass database includes the following types of information regarding glasses and associated waste streams:

- Compositions of glasses and wastes
- Leaching data
- Thermal properties data
- Crystallinity data
- Data sources

Ten database file types capture these information categories. The 10 database files will be described in the following sections. All glass database file types begin with the fields shown in Table 1. These provide unique information on the source of the data and the identification of the glass. The first field in each file type is occupied by a 12 digit glass code that links a specific glass composition to all of the properties and information associated with that glass throughout the database. This identifier is a link that allows logical organization of the glass data into sub-units from which it is easier to query and extract data.

It is intended that users of the database will be able to discern between highly reliable data sources and data of uncertain or questionable quality. A Data Quality Category field (# 6 in Table 1), used to classify data, has thus been established using the codes Q1, Q2, and U. These codes are described as follows:

Q1 = High quality assurance (QA). This applies to data obtained under a QA program that has been approved by the US DOE Office of Civilian Radioactive Waste Management, the branch responsible for the HLW repository. Such data can potentially be used for repository licensing purposes.

Q2 = Moderate QA. This is for data obtained under a quality assurance program that is not as rigorous as in the Q1 category. Data found in refereed journals or in published and numbered DOE laboratory reports would have this designation by default unless a higher level of QA was indicated.

U = No QA. This is for data gathered where a QA program was absent, unknown or not reported.

Field # 6 in Table 1 is also used to identify data from DOE project files that have not yet been cleared for release; the letter L is used to denote such data.

1. Glass Composition

The type of glass is identified, i.e. defense HLW, low-level waste, etc. as shown in Table 2. The waste stream source is provided along with an indication of whether the glass that was evaluated contained radionuclides and whether the composition is based on batch or recipe information or on analysis. The next 52 fields of the file are for glass compounds in alphabetical order, such as alumina, boron oxide, silica, etc. The standard unit for entering composition data is weight percent with mole percent as an alternative.

TABLE 1 INITIAL FIELDS FOR GLASS DATABASE FILES.

Field or Column #	Field or Column Heading	Description of Data in Field
1	Glass Code	12 digit code: {first three letters of author} {yymmdd} {glass sequence number}
2	Reference Code	Last name of first author, or organization name, or acronym
4	Glass Specimen ID	Identifier of glass specimen indigenous to the reference or data source, e.g. "PNL-7668" or "SRL-211" or sequence number in data table.
5	S.ID+	Additional identification information for glass specimen, usually table or page number from reference.
6	Data Quality	Code such as Q1, Q2, U or L

TABLE 2 WASTE GLASS COMPOSITION FILE

Field or Column #	Field or Column Heading	Description of Data in Field
7	Glass Type	Type of waste glass, i.e. defense high level waste, commercial high level waste, low-level waste, transuranic waste, municipal solid waste, in-situ vitrification process glass, industrial glass, naturally occurring glassy mineral.
8	Waste Type	Waste stream source, e.g. Hanford high-level waste (HWVP), Savannah River high-level waste (DWPF), West Valley high-level waste (WVNS), etc.
9	Radioactive Y/N	Indicate whether glass was radioactive
10	Units	Glass composition units, M=mole% or W=wt%
11	Analy Y/N	Indicate whether or not composition data is from chemical analysis
12 - 77	Al2O3, SiO2, etc.	The next 56 fields are provisions for up to 56 various glass oxides and glass compounds such as silica, alumina, boron oxide, soda, calcium fluoride, etc.
78	Fe2/Fe3	Ratio of iron redox species in glass
79	melt temp	Melting temperature in Celsius
80	Melt Appearance	Description of appearance of glass melt
81	Comments	Additional descriptive information

2. Glass Radionuclide Composition

Another file contains radionuclide data as described in Table 3. This data is actually part of the glass composition data. However, since there are very few glasses with radionuclides, it was decided to establish a separate file type for such glasses. This approach avoids a large amount of empty fields which would occur if the radionuclide data were merged with the general composition file. The file has provision for 19 standard radionuclides and two other radionuclides which can be case-specific.

3., 4., 5. Static, Semi-Static and Flow Leach Tests

The static leach test file contains primarily results from the MCC-1 and MCC-3 procedures. It can also accommodate the Product Consistency Test (PCT) or alternative static tests. As shown in Table 4, key experimental set-up parameters are accommodated, such as glass sample condition (crushed or monolith), glass surface area to leachate volume, leachate type, test temperature, test duration and leach results in specified units. The standard leach result unit is normalized release, which is solution concentration of leached glass constituent normalized by its glass mass fraction and the glass surface area. The file has provision for leach data on 36 different elements of stable isotopes. Leaching of up to 23 radionuclides can be accommodated. Fields are available for the composition of the leach solution before and after the leach experiment.

The semi-static leach test files are the same as the static leach test file, except a leachate change interval and a leachate change volume are included. The tests in this file include the IAEA procedure where the entire leachate is periodically replaced. The flowing leach test file is the same as the semi-static file except that a continuous flow rate is added. It also includes tests in which the leachate is partially replaced as an approximation to continuous flow.

TABLE 3 WASTE GLASS RADIONUCLIDE COMPOSITION FILE

Field or Column #	Field or Column Heading	Description of Data in Field
7	Radioactivity Measurement Date	Date radioactivity measured or reported
8	Radioactivity Units	Reported units of radioactivity
9	Analy Y/N	Indicate whether radioactivity values are from analysis
10	Radionuclide symbol (19 fields)	Radionuclide composition (quantity or concentration) values.
30	Other radionuclide #1	Additional radionuclide symbol not in standard list of 19
31	Data value	Radionuclide composition (quantity or concentration) values.
32	Other radionuclide #2	Additional radionuclide symbol not in standard list of 19
33	Data value	Radionuclide composition (quantity or concentration) values.
34	Total	sum of radioactivity

TABLE 4 STATIC LEACH TEST FILE

Field or Column #	Field or Column Heading	Description of Data in Field
8	Glass sample condition	Indicate whether crushed or monolith, provide particle size data or size of monoliths
9	Initial Glass SA/V (1/m)	Initial glass surface area to leachate hold-up volume
10	Leachate volume, m3	Volume of leachate in leach container
11	V/M, m3/g	Initial volume to specimen mass, m3/g
12	SA / M, m2/g	Initial specimen specific surface area in units of m2/g
15	Agitation Condition	Type or description of leach container agitation during test
16	Standard test type	Name of recognized standard test, e.g. MCC-1, etc.
17	Leachate Type	Leachate Type, e.g. deionized water, brine, basalt ground water, etc.
18	Leachate Pretreatment/ Materials Added	Leachate pretreatment, e.g. saturate with silica. Indicates whether leaching done in the presence of other solid phases such as metals or host rock.
19	Temperature	Leaching temperature
20	Duration (d)	Duration of leach test in days
21	pH (Start)	Initial pH of leachate
22	pH (End)	Final pH of leachate
23	Filter Size	Filter size used on final leach solution, e.g. 0.45 micron
24	Weight change units	Units of weight change data, wt% or mass
25	Weight change	Weight change (typically loss) incurred by glass specimen during leaching
26	Stable elements leach units	Leach units for stable elements. Standard units are normalized release - g/m2.
27	Element symbols (36 fields)	Leach data for 36 glass constituent elements, including the most commonly reported leach results such as Si, B, Na, Li, K, etc.
63	Other	Leach data for other element not included in standard list of 36
64	Comments	Additional descriptive information
65	Radionuclide leach units	Leach units for radionuclides
66	Radionuclide symbols (21 fields)	Leach data for 21 radionuclides. Choice of radionuclides is based on reported values in the literature. Includes fission products such as Cs-137, Tc-99 and Sr-90. Includes transuranics such as Pu-239, Np-237, U-233 and Cm-244.
85	Other radionuclide #1	Additional radionuclide not in standard list of 21
86	Leach data value	Leach data for other radionuclide #1
87	Other radionuclide #2	Additional radionuclide not in standard list of 21
88	Leach data value	Leach data for other radionuclide #2
89	Species symbol (30 fields)	Nominal, pretest and final leachate compositions. 30 species are included. Units are parts per million (ppm) on a mass basis.

6. Toxic Characteristic Leach Procedure And Ep-Tox Tests

The toxic characteristic leach procedure (TCLP) and its predecessor, the extraction procedure toxicity (EP-tox) test, are tests prescribed by the U.S. Environmental Protection Agency to determine the toxicity characteristics of a material. The fields in the database file are shown in Table 5. The test results are generally given as less than a detection limit. Also, there is provision for a definitive value rather than a "< value."

7. Waste Composition

This file provides data on the composition of various wastes as shown in Table 6. The waste type and source is specified. The data entries are in the form of glass compounds. This file is useful to those who are seeking the equivalent glass composition of various waste streams. This is the starting point for developing a waste glass.

TABLE 5 FILE FOR TOXIC CHARACTERISTIC LEACH PROCEDURE AND EXTRACTION PROCEDURE TOXICITY TEST

Field or Column #	Field or Column Heading	Description of Data in Field
8	Glass sample condition . . .	Indicate whether crushed or monolith, size data
9	Initial Glass SA/V (L/m)	Initial glass surface area to leachate hold-up volume
10	Standard test type	Name of test used, e.g. TCLP or EP-Tox
11	Leachate Type	Description of leachate
12	Temperature	Leach test temperature
13	Duration (d)	Test duration in days
14	pH (Start)	Initial leachate pH
15	pH (End)	Final leachate pH
16	Test result units	Units for test results, i.e. mg/L, etc
17	Ag value	Extraction result for the element, Ag, silver
18	Ag <value	Extraction result for element expressed as < a maximum value
19 - 38	Elements	Extraction results expressed as definitive or < values for As, Ba, Cd, Cr, Hg, Se, Pb, and two additional specified elements.

TABLE 6 WASTE COMPOSITION FILE

Field or Column #	Field or Column Heading	Description of Data in Field
7	Waste Type	Type of radioactive waste, i.e. Hanford, Savannah River, WV, etc. Includes specific flowsheet reference, e.g. NCAW, etc.
8	Units	Units for waste composition, W = wt%
9	Glass oxide or compound symbols (46 fields)	Waste composition values expressed as stable oxides or glass compounds
10	Other	Other constituents not included in standard list of 46.

8. Thermal Properties

Thermal properties are defined here as those measured at melting temperatures or temperatures near and above the glass transition. This file includes such properties as viscosity, electrical conductivity, thermal conductivity, density, and heat capacity. Fields are provided for transition temperature and softening temperature as in Table 7. Other properties are specified along with the units as found in the source. Temperature dependence can be accommodated. Up to 12 property values and the associated temperatures can be entered for each record.

TABLE 7 THERMAL PROPERTIES FILE

Field or Column #	Field or Column Heading	Description of Data in Field
7	Tg, C	Glass transition temperature
8	Ts, C	Glass softening temperature
9	Property	Type of glass property data, i.e. viscosity, electrical conductivity, thermal conductivity, thermal expansion coefficient, etc.
10	Temp Units	Temperature units associated with property, usually Celsius
11	Units	Units of glass property, e.g. poise (P), ohm-cm, etc
	Temperature and Property Data Pairs.	12 such pairs are provided per glass composition and thermal property. The format is as shown:
12	T1	Temperature 1
13	V1	Property 1

9. Crystallinity

Fabricated waste glasses often contain a small percentage of crystallinity. This file provides a means to catalog this information. As shown in Table 8, a field for describing how the crystallinity was determined is provided. Other fields are for describing the crystal and its weight or volume percent in the glass. The six fields shown are repeated five more times to accommodate up to six crystalline phases for each glass.

TABLE 8 CRYSTALLINITY FILE

Field or Column #	Field or Column Heading	Description of Data in Field
6	Analy. Method	Method for analyzing crystallinity, e.g. X-ray diffraction, etc.
7	Crys. Species No.1	Indicate type, e.g. spinel, etc
8	Formula	Molecular formula for crystals
9	Wt%	Weight % as crystals in the host glass
10	Vol%	Volume % as crystals in the host glass
11	Description of Crys. Species	Descriptive information, size, etc.

10. REFERENCE INFORMATION

A file to uniquely describe the source of data is part of the database and is shown in Table 9. The 9 characters of the reference code are identical to the first 9 characters of the glass code for the glasses found in the reference. This linkage assures that all glasses can be tied back to a reference. The format of Table 9 allows descriptions of references from journals, government reports, and project files.

DATA RETRIEVAL APPROACH

The database occupies approximately twelve megabytes of disk space; it is separated into ten files that relate to different aspects of glass properties. Each of these files has one common field, Glass Code. The Glass Code field acts as the primary key; it is used to relate each of the files when queries are performed. Some of the files contain unique entries in the Glass Code field, allowing the field to serve as an independent primary key for that file. Other files have multiple key fields to serve as the link between files. This is necessary because some of the files contain information on the same glass composition - run with different test variables. This forced the use of a second, and in some instances, even a third indexed field. The combination of these fields is called a compound key. The result remains the same: a direct relationship between files based upon Glass Code. Any variable may be used to query the files; the purpose of the key field(s) is only to relate the records based upon the Glass Code.

The user interface allows the database to be used to its full potential by the largest possible audience. The user interface will be menu driven. It can support queries upon any field contained in the files, as well as multiple field queries. The

TABLE 9 DATA REFERENCES FILE

Field or Column #	Field or Column Heading	Description of Data in Field
1	Reference Code	9-digit code identical to the glass code used in other data files except the 3 digit glass sequence number is omitted
2	First Author	First author name if report or article. Principle investigator name if unpublished data from project files.
3	Second Author or et al	Second author or co-investigator
4	Year	Year of publication or preparation of source file
5	Month	Month of publication
6	Day	Day of publication
7	Article Title	Title of journal article
8	Publication, Report, or Journal	Title of publication in which journal article appeared, or title of topical report
9	Pages	Page numbers for journal article
10	Internal DOE Transmittal Description	For information from DOE project files, description of transmittal letter (sender, receiver, date)
11	Publisher, issuing organization or DOE program	Publisher name or organization issuing topical report (e.g. PNL, etc)
12	Report #	Publication number of topical report (e.g. PNL-6723)
13	Other Information	Space for other descriptive information for reference

fields can be selected through a dialog box that will display the available fields. Once the fields are selected and the query is performed, the result is displayed in a tabular form. This table can be saved and is a valid file upon which subsequent searches may be made. The use of a control value is also supported. An example of this would be, 'Search by Al2O3.CMP >=10", which tells the database to find all instances of alumina with a composition than or equal to 10 wt %. Report formats will also be incorporated into the project. This will allow the user to create a query, and send the result in a report form to the printer, screen, text file or spreadsheet file.

CONCLUSION

A database with waste glass composition and properties has been developed. Properties include leaching, thermal parameters, crystallinity, and radionuclide content. The database includes reference information so users can access the original data documents. About 2000 glasses are in the database and there are about 8000 records for these glasses. Each record contains up to 100 entries. The data has been extracted from over 100 sources including published reports, journal articles, books and project files. Commercial database software has been used to develop a method for conducting queries and retrieving selected information. The database will be a tool that allows investigators ready access to waste glass properties for such purposes as disposal evaluation, glass development, and vitrification system design.

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