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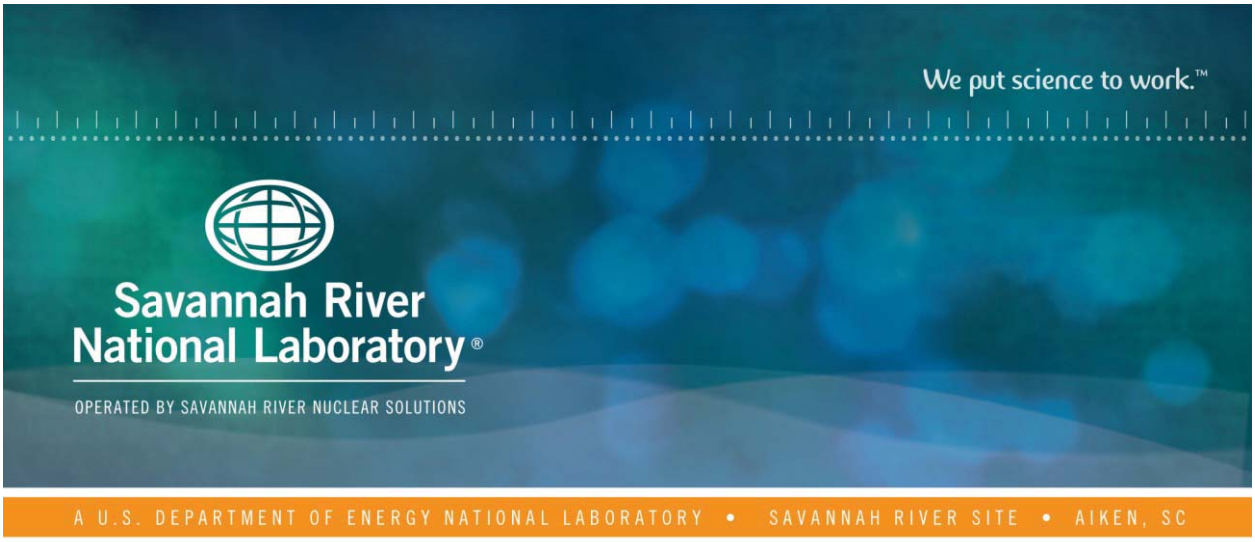

Summary of Information Available and Recommendations for a DOE Complex-Wide Waste Glass Database

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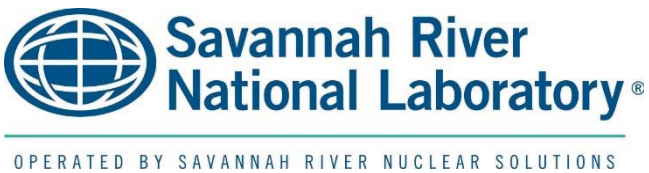

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EXECUTIVE SUMMARY

The purpose of this document is to summarize the existing repositories of nuclear waste glass property-composition data produced for DOE. A framework is described for consolidation of the data into a single database. Recommended attributes of the consolidated database are provided, including quality assurance, the user interface, and means for updates and maintenance. Future work will utilize the framework described herein to produce the complex-wide database and establish the user interface.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF ABBREVIATIONS

C_C	crystallization after the CCC heat treatment
CCC	Canister Centerline Cooled
C_T	crystallization type and amount as a function of temperature
CVS	Composition Variation Study
CUA	Catholic University of America
DOE	U.S. Department of Energy
DWPF	Defense Waste Processing Facility
EM	DOE Office of Environmental Management
HLW	High-Level Waste
HWVP	Hanford Waste Vitrification Project
INEEL	Idaho National Engineering and Environmental Laboratory
k3	K-3 refractory corrosion rates
LAW	Low-Activity Waste
MCC-1	Materials Characterization Center test one
NQAP	Nuclear Quality Assurance Program
PCT	ASTM Product Consistency Test
PNNL	Pacific Northwest National Laboratory
QA	Quality Assurance
SRNL	Savannah River National Laboratory
TCR	Task Change Request
TDO	Technology Development Office
T_g	glass transition temperature
VHT	Vapor Hydration Test
VSL	Vitreous State Laboratory
w_{SO3}	sulfur solubility
WTP	Hanford Tank Waste Treatment and Immobilization Plant
WVDP	West Valley Demonstration Project
XRD	X-ray Diffraction
ε_T	electrical conductivity as a function of temperature
η_T	viscosity as a function of temperature
ρ	density

1.0 Introduction

The U.S. Department of Energy (DOE) utilizes vitrification technology for the safe and permanent immobilization of legacy radioactive wastes resulting from nuclear materials production. DOE vitrification facilities employ complex glass property-composition models to ensure processability of waste streams and acceptability of the final glass products. Large amounts of glass property-composition data have been produced by Savannah River National Laboratory (SRNL), Pacific Northwest National Laboratory (PNNL), the Catholic University of America (CUA), and other institutions in support of the development, validation, and continuous improvement of these models. The data exist in multiple repositories and there is currently no way to access the data as a whole to support continuing studies.

The purpose of this document is to summarize the existing repositories of nuclear waste glass property-composition data produced for DOE. A framework is described for consolidation of the data into a single database. Recommended attributes of the consolidated database are provided, including quality assurance, the user interface, and means for updates and maintenance. Future work will utilize the framework described herein to produce the complex-wide database and establish the user interface.

Many fields of science and engineering have experienced significant growth in understanding and capability by opening data to all potential developers. This effort will encourage practitioners from U.S. and foreign universities and laboratories to perform analyses, fit models, and develop innovative approaches to property prediction and glass formulation. The DOE-EM mission stands to significantly accelerate innovation in this area with only the minimal cost of database compilation and maintenance. These innovations are likely to reduce cleanup cost and duration with the development of new, more efficient cleanup strategies as a result.

2.0 Quality Assurance

This task is part of the SRNL Technical Assistance Program in support of the DOE Office of Environmental Management (EM) Technology Development Office (TDO). The task is authorized via Task Change Request (TCR) SR031102 and is designated Task 8 of the TCR. Work was performed following Task Plan SRNL-L3310-2020-00017. Requirements for performing reviews of technical reports and the extent of review are established in Savannah River Site Manual E7, Procedure 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2.

Work performed at PNNL will be done according to the Nuclear Quality Assurance Program (NQAP) Manual (NQAP-2012, Rev. 4.0, 2019, PNNL-SA-115260) and associated implementing procedures including: *IP-0601 Document Preparation and Control, IP-0602 Independent Technical Review, IP-1102 Calculation Control, IP-1103 Qualification of Existing Data, IP-1106 Data Management, and IP-1707 Records Management* (among others). Under this program, data will be collected, checked, qualified if necessary, and converted to appropriate database format, assembled into database, checked and documented. The qualifications of staff performing the work will be verified and appropriate training will be performed and documented.

3.0 Existing Collections of DOE Nuclear Waste Glass Data

Several glass property-composition databases have been developed in support of DOE nuclear waste vitrification activities, including those at: West Valley Demonstration Project (WVDP); Defense Waste Processing Facility (DWPF); Hanford Waste Vitrification Project (HWVP); Idaho National Engineering and Environmental Laboratory (INEEL) Vitrification Projects; the Hanford Tank Waste Treatment and Immobilization Plant (WTP) Low-Activity Waste (LAW) Vitrification and WTP High-Level Waste (HLW) Vitrification; and others. These data are compiled in a number of formal and informal databases described

in this document. Each of these databases serves or served a specific purpose and as such, they vary in the type of data captured, the properties measured, the quality assurance (QA) programs used to generate, tabulate, and verify the databases, and the format or software used to store and access the data. Most of these databases are described in more detail below, although this is not intended to be an exhaustive list.

3.1 ComPro

SRNL maintains ComPro™, a database of nuclear waste glass properties and compositions,[1] as part of its role in supporting the DWPF. ComPro contains 13,479 rows of glass data. Each row consists of a glass composition and its associated properties. Note that, for some glasses, multiple compositional "views" are included as individual rows in the database (e.g., targeted glass composition, measured glass composition, measured glass composition with bias-correction, etc.).

The data in each row of ComPro generally include the glass identifier, quality assurance level, source information (report title, document number, author, etc.), type of heat treatment performed, composition (oxide basis), and ASTM Product Consistency Test (PCT)[2] results (as normalized concentrations for select leachate species). For some glasses in the database, information on temperature used for melting, visual observations of the glass, microscopy information, X-ray diffraction (XRD) data, and liquidus temperature data are included. Note that extensive error checking for the data included in ComPro was completed only for the composition and PCT data.[3, 4]

The data in ComPro are categorized according to the QA practices under which they were collected.[3] As a result of this process, 7,871 rows in ComPro are designated as "model" data, meaning that the data were collected in accordance with DOE/RW-0333P Quality Assurance Requirements and Description (DOE/RW-0333P QARD)[a] or its predecessor RW-0214.

ComPro was last updated in the Fall of 2013 (Revision 2).[1] Since that time additional studies have been completed in support of the DWPF. Glass composition and property data from these studies are maintained in informal databases at SRNL, with the intent of incorporating them into ComPro as part of the next revision. Many of these glass compositions were fabricated and characterized in cooperation with the Vitreous State Laboratory at the Catholic University of America (VSL). A search identified the following documents with data that are appropriate for inclusion in the next ComPro revision:

- SRNL-STI-2013-00462, Rev. 0
- VSL-13R2580-1
- SRNL-TR-2016-00094, Rev. 0
- SRNL-STI-2016-00115 Rev. 0
- SRNL-STI-2016-00372 Rev. 0
- VSL-16R3370-1
- SRNL-STI-2017-00016, Rev. 0
- SRNL-STI-2018-00699, Rev. 0
- VSL-19R4680-1
- "Nepheline Crystallization and the Residual Glass Composition: Understanding Waste Glass Durability," IJAGS (*accepted for publication*)

3.2 WTP LAW Glass Properties Database

PNNL has developed a lengthy database containing most if not all of the LAW glass data collected in support of the WTP. Two versions of this database exist: a full version without a controlled QA pedigree and a subset with a controlled QA pedigree. The full version is the most complete database, last updated

[a] U.S. Department of Energy Office of Civilian Radioactive Waste, Quality Assurance Requirements and Description, DOE/RW-0333P.

in 2016 in support of WTP glass property models and constraints development.[5] The information in this database includes: PCT response of quenched and canister centerline cooled (CCC) glasses; Vapor Hydration Test (VHT) results of quenched and CCC glasses, viscosity as a function of temperature (η_T), electrical conductivity as a function of temperature (ε_T), crystallization type and amount as a function of temperature (C_T), crystallization after the CCC heat treatment (C_C), sulfur solubility (w_{SO3}), density (ρ), analyzed and targeted compositions, K-3 refractory corrosion rates (k3), selected glass transition temperatures (T_g), and general observations (melting temperature, appearance of as-melted glass, indications of salt segregation, etc.). This database includes data from a number of different studies, some of which were not performed under the applied research aspects of the NQA-1 program while others were performed under NQA-1 applied research standards. The database itself was not reviewed following NQA-1.

3.3 WTP LAW Glass Properties Database with QA Pedigree

The WTP LAW Glass Properties Database described in Section 3.2 was screened to select only those data that were generated under the PNNL Nuclear Quality Assurance Program (NQAP)[6] or an equivalent QA program under applied research. This database includes the same properties as the full version, but with fewer entries. This database was last issued in April 2020.[7] During the QA reviews of this database, errors were identified in the full (without a controlled QA pedigree) version of the database. In addition, the data of three studies, not available at the time of the last release of the full version, were added. The full version has not been updated to reflect these changes.

3.4 WTP HLW Glass Properties Database

PNNL has developed a lengthy database containing most if not all of the HLW data collected in support of the WTP, INEEL, WVDP, DWPF, and others. The full version is the most complete HLW database currently available and was last updated in 2016 in support of WTP glass property models and constraints development.[5] The information in this database includes: PCT response of quenched and CCC glasses, η_T, ε_T, C_T, C_C, T_L, w_{SO3}, ρ, T_g, analyzed and targeted compositions, and general observations (melting temperature, appearance of as-melted glass, indications of salt segregation, etc.). This database includes data from a number of different studies, some of which were not performed under the applied research aspects of the NQA-1 program or RW-0333P, while others were performed under NQA-1 applied research and/or RW-0333P (and its predecessor) standards. The database itself was not reviewed following NQA-1 or RW-0333P. There is undoubtedly overlap between this database and the ComPro database managed by SRNL. The degree of overlap and the presence of unique data entries will be identified as part of the consolidation effort.

3.5 WTP HLW Glass Properties Database with QA Pedigree

A database with a controlled QA pedigree was generated by PNNL and VSL under the WTP project for support of HLW glass qualification activities.[8] This database is fully compliant with NQA-1 applied research and RW-0333P requirements. It contains measured and targeted glass compositions, PCT responses for quenched and selected CCC heat treated glasses, C_C, T_L, C_T, η_T, ε_T, ρ, and selected T_g.

3.6 Accelerated Leach Testing of Glass (ALTGLASS) Database

SRNL maintains the ALTGLASS database[9], a collection of data from short- and long-term PCTs (ASTM C1285 A and B) on HLW and LAW glasses. The database provides an archive of experimental data for the purpose of studying, modeling, or validating existing models of nuclear waste glass corrosion. ALTGLASS provides the leachate compositions for corroding solutions including data for: Si, B, Ca, Li, Na, K, Al, Fe, Mo, Mg, Ti, Zn, and Zr. The database provides additional experimental information about each of the reported PCTs including: glass type (HLW or LAW) and composition; ρ; glass mass and particle size; type and volume of leachant; surface area-to-volume ratio; test vessel type; test duration and

temperature; final pH of leachant solution; the secondary mineral phases formed during corrosion and the method used for identification.

There was no tracking of QA pedigree in the development of ALTGLASS. The current version, ALTGLASS 4.0,[10] has been updated with an additional 100 rows of data representing PCT results from corrosion experiments conducted in the United States by SRNL, PNNL, Argonne National Laboratory, and VSL, as well as the National Nuclear Laboratory (NNL) in the United Kingdom and the CEA in France.

3.7 ALTGLASS Database with QA Pedigree

A subset of the data contained in ALTGLASS was obtained in programs performed under NQA-1 applied research, RW-0333P, or both. Data for those LAW glasses that fit these criteria were selected for a version of ALTGLASS with a QA pedigree.[11] This database also includes new static leach test data obtained after the last issuance of the full version of ALTGLASS.

3.8 Composition Variation Study (CVS) Database

PNNL performed a composition variation study in support of the Hanford Waste Vitrification Plant (HWVP, a predecessor to the WTP).[12] A total of 124 simulated HLW glasses were statistically designed to efficiently cover composition space. Glasses were tested for: PCT response of quenched and CCC glasses, η_T, ε_T, C_C, T_L, ρ, T_g, analyzed and targeted compositions, coefficient of thermal expansion, 28-day Materials Characterization Center test one (MCC-1), and general observations (melting temperature, appearance of as-melted glass, indications of salt segregation, etc.). The experimental design, glass fabrication and testing, and reporting were all performed under the QA program DOE/RW-0214 (a predecessor to RW-0333P and generally consistent with NQA-1).

3.9 Washington State University and Rutgers Data

Washington State University (Prof. John McCloy) and Rutgers (Prof. Ashutosh Goel) have collaborated since 2014 on fundamental studies of nepheline crystallization from simplified glass compositions, generally variations from $NaAlSiO_4$ stoichiometric nepheline glass adding or substituting Li, Ca, Fe, Ti, Sn, Zr, B, and P in various ways. For the most part, the data are published in journal papers but have not yet been collated together in one place. Included in these data are the following (note that not all datasets contain all items):

- X-ray diffraction results, usually quantitative, on phase formation as a function of heat treatment (heat treatments vary and can be CCC or other isothermal heat treatments designed to maximize crystallization, or non-isothermal heat treatments designed to identify the temperature of formation of certain phases).
- Glass transition temperatures and crystallization temperatures, from thermal analysis data
- Raman spectra of glasses and crystallized glasses
- Nuclear magnetic resonance (e.g., Si-29, Al-27, Na-23, B-11, P-31) spectra of glasses and crystallized glasses
- Electron microprobe measurements of crystal phases and glasses
- Mossbauer measurements of Fe-containing glasses and crystallized glasses
- Vibrating sample magnetometry measurements of Fe-containing glasses and crystallized glasses
- Pair distribution function analyses from X-ray and neutron total scattering of selected glasses and liquids
- X-ray absorption of selected glasses
- Chemical durability of selected glasses

3.10 Additional Glass Databases and Data Sources

Additional, valuable databases exist in the glass research community that would add significant value to the EM mission. Examples of such databases include but are not limited to: SciGlass database (a commercial database of glass properties including those important to waste glass), Fluegel database (a commercial and waste glass database collected by Alex Fluegel ca. 2010 and summarized at www.glassproperties.com), VSL long-term static corrosion database (a privately funded collection of glass static corrosion test results with over 10,000 unpublished data points), and international waste glass property databases maintained by NNL, Sheffield University, JAEA, JNFL, CEA, Orano, etc.

PNNL, SRNL, and VSL have produced many reports over the past several years describing a host of waste glass properties as part of DOE and industrial sponsored activities (see for example the tasks guided by the DOE-ORP research and development plans by Peeler, et al.[13]) that have not yet been incorporated into any of the existing databases.

4.0 Recommendations for Complex-Wide Database

The following sections suggest the framework for creation of a DOE complex-wide open database (CWOD) of waste glass properties.

4.1 Selection of Glass Properties Included in Database

The specific datasets and properties to include in the CWOD will be prioritized. Table 4-1 lists the initial priorities for both the sources and types of data to be included. The prioritization of individual glass properties will be revisited after the initial database is assembled.

Table 4-1. Prioritization for Data Sources and Types

Priority	Databases	Properties
High	Those databases with full QA pedigree and with data focused on WTP and DWPF glasses (e.g., ComPro, WTP-LAW-QA, WTP-HLW-QA, ALTGLASS-QA, CVS)	Those properties currently constrained for plant operation or glass qualification (e.g., PCT-Q, PCT-CCC, VHT-Q, VHT-CCC, CC, T_L, C_T, η_T, ε_T)
Medium	Those databases without full QA pedigree with data focused on WTP and DWPF glasses (e.g., WTP-HLW-noQA, WTP-LAW-noQA, ALTGLASS)	Properties that either directly or indirectly influence the success of WTP or DWPF without formal constraints (e.g., Long-term PCT, Single-Pass Flow-Through, ρ, T_g, TCLP, Cp, k, CTE, T_S, radiation stability)
Low	Those databases for other waste glass applications (e.g., INEEL, WVDP, LaHague, Sellafield, Tokai, Rokkasho, etc.)	Properties currently unrelated to waste vitrification (e.g., optical properties, mechanical properties, radiation stability, surface tension, etc.)
Very Low	Commercial glasses and waste glasses not of the traditional alkali-borosilicate family (e.g., phosphate, tellurite/tellurate).	To be determined

4.2 Quality Assurance

Compilation of the database will be performed according to SRNL and PNNL nuclear quality assurance programs. The QA pedigree of the original data will be marked as a characteristic of the datasets in the COWD. Those data with NQA-1 and RW-0333P QA will be entered in the database and checked according to appropriate procedures for those QA levels. Other glasses will be marked with *For Information Only* QA level and associated entry and validation will be performed. It is likely that some datasets will be deemed worthy to qualify under a more rigorous QA standard. In those cases, a qualification plan will be developed, approved, and followed to qualify data for inclusion at an elevated QA level.

4.3 Structure and Format

The structure and format of the database will be determined after a preliminary assembly of the highest priority data sources is completed. SRNL and PNNL researchers will combine existing data sets in the high priority category (Table 4-1). The result will be screened for the removal of overlapping data. The new database will then be reviewed to determine how best to utilize the information for continuing modeling efforts, and for the addition of new and lower priority data sets. The outcome of this process will determine the appropriate structure and format for the CWOD.

4.3.1 Software

The database will be hosted by SRNL computing resources. The specific database software, or backend, to be used will be determined after the preliminary database of high priority information is assembled.

Discussions with SRNL Information Technology staff have indicated that it is better to select the appropriate software after the preliminary database is assembled, rather than select a software platform *a priori*. Consideration will be made for the amount and types of data to be stored, efficient access methods, appropriate ties to the user interface, and upgradeability.

4.3.2 User Interface

A user interface, or frontend, for the database will be developed once the backend software platform is identified. The user interface will be hosted online by SRNL computing resources to allow for accessibility to the broadest range of interested users. The interface will allow users to search the database using multiple fields, including glass compositions and individual properties. The interface will allow for downloading of requested data in multiple formats to allow users to manipulate data in support of their own modelling efforts. Users will be required to cite use of the CWOD in any derivative works. The interface will also provide users with the ability to submit new data for review and potential inclusion in the CWOD.

4.3.3 Availability and User Permissions

Full database control will be limited to specifically trained individuals at SRNL and PNNL, deemed the CWOD administrators. Decisions on access to the database will be made by the CWOD administrators under the guidance of DOE-EM. It is the intent to have as broad a user base as possible. Interested parties will be able to apply for permission to access the open portion of the database and will be assigned user access credentials. Users will be able to submit their own datasets for inclusion in the database. These submissions will be screened by the CWOD administrators, verified, classified by quality assurance pedigree, and incorporated into the open portion of the database for all users to access. Newly added datasets will be available only to program staff at SRNL and PNNL until screening and reviews are complete, after which they will be transferred to the open access portion of the database.

4.4 Updates and Maintenance

As new data sets are loaded by users or by the CWOD administrators they will be checked, screened, and qualified as necessary by the SRNL/PNNL project team and then made available to all users. Maintaining the database will require some continued, although relatively modest, funding from DOE.

4.5 Initial Trial and Feedback

Selected individuals from the international waste glass community will be invited to participate in a trial program to review the initial database. They will be asked to give written or oral feedback to the CWOD administrators who will disposition the comments. Updates to the database and software interface will be made and a decision as to immediate roll-out or an additional iteration of beta testing will be made.

4.6 Final Rollout

It is anticipated that the initial database will be rolled out with the highest priority datasets and highest priority properties. Additional datasets and properties will be added and made available on-line as the project progresses. Users (including those from the major waste glass testing organizations in the US) will continue to add datasets to the database. Those will be screened, verified, and qualified as necessary and included in the on-line portion of the database for all users to access.

5.0 Summary

To accelerate innovation in the area of nuclear waste glass property models and glass formulation, DOE-EM is embarking on the development of an open database of waste glass properties and compositions. This database will bring together many smaller datasets developed at PNNL, SRNL, and other institutions working in the area of glass testing and formulation. The data and property sets will be prioritized and incorporated into a single database in stages. The initial stage will incorporate those datasets with appropriate quality assurance pedigree for glass compositions and properties of direct influence on glass

formulations at Hanford and Savannah River sites. Additional datasets and properties will be added on a prioritized basis. The database will be open for all registered and approved users who will have immediate access to the current public version of the database. Each user will be able to upload datasets for project staff to screen, evaluate, and incorporate into the public database.

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