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Waste Glass Property Database and Data Qualification Plan

June 2023

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Abstract

The U.S. Department of Energy vitrification facilities currently employ glass property-composition models to ensure processability of waste streams and acceptability of the final glass products. The efficiency (e.g., process flexibility, reduced down time, and reduced cost) of waste vitrification is directly tied to the size of the waste processing envelope. To increase the size of the processing envelope, a larger database was developed. The database will improve the prediction accuracy of glass properties, leading to higher waste loadings, higher waste throughput, broader process flexibility, and reduced mission life.

Some of the data in this expanded database has insufficient quality assurance (QA) rigor for use in nuclear facility operation and glass qualification. A plan was developed to qualify high-value data under the appropriate QA rigor for use at the Hanford Waste Treatment and Immobilization Plant's High-Level Waste (HLW) Facility. This qualification plan first compares the data coverage over the composition region of interest to Hanford HLW by property. Those studies that significantly improve the data coverage in the Hanford HLW glass composition region were prioritized for qualification activities. Qualification methods, including QA equivalence, peer review, data corroboration, and confirmatory testing, have been assigned to each high-priority study.

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Acronyms and Abbreviations

CCC	container centerline cooled
CUA	Catholic University of America
DFHLW	Direct Feed High-Level Waste
DOE	U.S. Department of Energy
EC	electrical conductivity
EWG	enhanced waste glass
GLP	good laboratory practices
HASQARD	Hanford Analytical Services Quality Assurance Requirements Document
HLW	high-level waste
K-3	K-3 refractory corrosion
LAW	low-activity waste
mf	mass fraction
NQA	Nuclear Quality Assurance
PCT	Product Consistency Test
PNNL	Pacific Northwest National Laboratory
QA	quality assurance
SOL	undissolved solids loading
SRNL	Savannah River National Laboratory
TCLP	Toxicity Characteristic Leaching Procedure
TRL	technology readiness level
WTP	Waste Treatment and Immobilization Plant

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1.0 Introduction

The U.S. Department of Energy (DOE) vitrification facilities currently employ glass property-composition models to ensure processability of waste streams and acceptability of the final glass products. The efficiency (e.g., process flexibility, reduced down time, and reduced cost) of waste vitrification is directly tied to the size of the waste processing envelope. To increase the size of the processing envelope, a larger database is needed. Such a database can improve the prediction accuracy glass properties, leading to higher waste loadings, higher waste throughput, broader process flexibility, and reduced mission duration.

Large amounts of glass property-composition data have been produced by Pacific Northwest National Laboratory (PNNL), the Catholic University of America (CUA), Savannah River National Laboratory (SRNL), Idaho National Laboratory (INL), and other institutions in support of the development, validation, and continuous improvement of waste glass formulations and property-composition models. The data exist in multiple sets, and there is currently no way to access the data as a whole to support continued development of glass property-composition models for future vitrification facility process improvement. A plan to compile these data into an open access database was developed in 2020 [1]. This project aims to compile these data in a single database useful to the DOE waste vitrification facilities, including the Hanford Waste Treatment and Immobilization Plant (WTP), the Defense Waste Processing Facility (DWPF) at the Savannah River Site, and potentially the Idaho Calcine Disposition Project. The database compilation method is presented in Section 2.0, and the compiled glass database is reported in Section 2.2.

Some of the data in this expanded database have insufficient quality assurance (QA) rigor for use in nuclear facility operation and glass qualification. A plan was developed to qualify high-value data under the appropriate QA rigor for use at the WTP High-Level Waste (HLW) Facility (Section 3.0). The compiled database (from Section 2.2) was compared to the glasses formulated using the enhanced waste glass (EWG) method developed for Hanford HLW (Sections 3.1 and 3.2). The composition gaps (Sections 3.3 to 3.5) for different properties were identified, including (1) the regions where new glasses/measurements are needed, and (2) the publications developed under insufficient QA programs that contain the data of interest (i.e., publications that need future QA qualification). A data qualification plan with a priority order of datasets and properties was then developed (Section 3.6). In the plan, methods including QA equivalence, peer review, data corroboration, and confirmatory testing are assigned to each study for future qualification efforts.

To summarize, objectives of this project are as follows:

- Gather waste glass property data and compile it into a single database.
- Identify potential compositional gaps for Hanford HLW treatment by comparing the database to the glasses formulated using the EWG method.
- Draft a plan to qualify high-value data with insufficient QA rigor for use in Hanford HLW treatment.

2.0 Glass Database

2.1 Compilation Method

Previously compiled databases and individual data reports were collected, which included a total of 134 datasets. Table 2.1 lists a few examples of the previous data files and their number of data points.

Table 2.1. Previous databases and number of data points.

Database/File Name	Lab/Study	Number of Data Points
ComPro Revision 2 Spreadsheet.xlsx	SRNL database	13477
Database (4-22-09).xlsx	PNNL database	3769
ORP HLW Dataset 2012.xls	CUA HLW [2]	276
ORP LAW Dataset 2012.xls	CUA LAW [2]	231
EWG-CCP-078 Rev5.xlsx	2022 LAW property models (PNNL-30932) [3]	1186
Np Database 10-05-2020, CCP-130.xlsx	Lu et al. 2021 [4]	955
HLW SO3 Solubility Final model with Var-Covar Matrix.xlsx	2016 property models (PNNL-25835) [5]	68
HLW+LAW Visc Comp for JMP_v1a.xlsx	PNNL Viscosity	2037
Spinel Model Data_8-22-16.xlsx	PNNL Spinel	858
High-alumina matrix summary 12-15-15 with partition rules.xlsx	PNNL-27138 [6]	94

A Python programming code was developed to compile previous database files (in Excel .xlsx format). The Python code can (1) read the Excel files, (2) reformat the files to desired format (e.g., separate target and analyzed compositions), (3) merge the cells based on column headers and glass IDs (e.g., the code first determines if a glass ID and composition already exist, and then decides to either add a new row for the glass or merge the new property data to the existing glass ID), and (4) generate a single, compiled database in Excel format.

In developing the data qualification plan, any available original reports and papers were first gathered and organized, and then the QA information for each study was acquired and consolidated into the database. Table 2.2 lists each property along with its abbreviation and corresponding QA requirement. Generally, U.S. nuclear waste glass property data have been collected under one or more of four general QA levels:

1. Good laboratory practices (GLP) is a catch-all category for QA rigor that is sufficient to publish government reports, conference proceedings, dissertations/theses, and journal articles. Some studies associated with GLP QA category in this report were performed under the American Society of Mechanical Engineers (ASME) *Quality Assurance Requirements for Nuclear Facility Applications* (NQA-1) [7] under Basic Research (or technology readiness level [TRL] 1-3). Although data records, reviews, and checks have been performed and documented, these data are not intended for quality-impacting work such as design basis input, plant operations, or waste form compliance.
2. NQA-1 under Applied Research (or TRL > 3; referred to as NQA in this report) satisfies the requirements for the establishment and execution of QA programs during siting, design, construction, operation, and decommissioning of nuclear facilities.

3. The *Quality Assurance Requirements and Description* (QARD) (often referred to as RW-0333P) [8] is the QA program for the Yucca Mountain repository and is used to control the quality of data used in HLW glass compliance.
4. The *Waste Management System; Testing and Monitoring Activities; Final Rule: Methods Innovation Rule and SW-846 Final Update IIIB* (SW-846) [9] includes the QA requirements necessary to evaluate the hazardous or nonhazardous nature of solid wastes under the *Resource Conservation and Recovery Act* (RCRA) [10], for example, delisting of Hanford HLW glass or satisfying land-disposal restrictions for Hanford low-activity waste (LAW) glass. The Hanford-specific QA program that is set to satisfy the SW-846 requirements is the *Hanford Analytical Services Quality Assurance Requirements Document* (HASQARD) [11]. So, for data related to RCRA hazard evaluation, either SW-846 or HASQARD is required.

Table 2.2. Property, abbreviation/symbol, and QA requirement for Hanford HLW treatment.

Property	Abbreviation/Symbol	QA requirement
Product Consistency Test response	PCT	QARD (RW-0333P) [8]
Toxicity Characteristic Leaching Procedure	TCLP	SW846 [9] or HASQARD [11]
Nepheline formation	Nepheline	QARD (RW-0333P) [8] (impact PCT)
Viscosity	Visc. or η	
Electrical conductivity	EC or ϵ	
Spinel formation at 950 °C	Sp950	
Glass transition temperature	T_g	NQA-1 [7] (TRL > 3 or Applied Research) or RW-0333P
SO ₃ solubility	SO ₃	
K-3 refractory corrosion	K-3	
Density	Density or ρ	
Liquidus temperature	T_L	

2.2 Compiled Glass Database

The current version of the database is in an Excel file (.xlsx) format with 17,006 rows and 305 columns. In several cases, the same glass with different compositional views (target or analyzed) and heat treatments (quenched or canister centerline cooled [CCC]) were compiled separately as different rows. In the database, there are 6718 quenched glasses with the target composition.

Table 2.3 lists the total number of data points, including QA qualified and insufficient QA data by each property. All the available K-3 and SO₃ solubility data satisfy the QA requirement, while more than half of PCT, T_L , and nepheline data do not satisfy the QA requirements. Table 2.4 summarizes the number of glasses and property measurements by study.

Table 2.3. Number of data points by property.

Property	QA Requirement	Total Num of Data	QA Data	Insufficient QA Data
PCT	QARD	4526	1845	2681
TCLP Cd	SW846, or HASQARD	355	187	168
Nepheline	QARD	1286	286	1000
T _L	NQA	850	364	486
Viscosity	NQA	2389	2017	372
EC	NQA	846	841	5
Spinel	NQA	877	665	212
T _g	NQA	177	162	15
SO ₃ solubility	NQA	761	761	0
K-3	NQA	349	349	0
Density	NQA	462	361	101

Table 2.4. Glass studies with ≥ 20 glasses (unique glass IDs) by property. Number of property measurements includes quenched and CCC samples. The studies with < 20 glasses are grouped into data from PNNL, SRNL, CUA, and others (shown at the end of the table).

Reference	Number of Unique Glass IDs	QA	PCT	TCLP	Nepheline	T _L	Viscosity	EC	Sp950	T _g	SO ₃	Density	K-3
VSL-12R2470-1 [2]	497	NQA	304	0	0	0	244	0	252	0	11	0	0
VSL-01R2540-2 [12]	181	QARD ^(a)	28	140	0	142	23	2	0	2	0	20	0
WSRC-TR-93-672 [13]	175	Unknown	177	0	0	0	0	0	0	0	0	0	0
VSL-07R1240-04 [14]	147	NQA and QARD ^(b)	131	114	0	11	119	118	102	11	15	13	0
PNL-10359 [15]	146	QARD	290	0	121	0	122	0	0	123	0	143	0
VSL-04R4480-1 [16]	145	NQA	346	2	0	0	334	34	0	0	100	12	0
VSL-01R3560-2 [17]	115	NQA	109	0	1	0	108	95	0	0	81	22	0
DP-MS-81-108 [18]	104	GLP	0	0	0	35	0	0	0	0	0	0	0
PNNL-20184 [19]	96	NQA	0	0	128	0	0	0	0	0	0	0	0
PNNL-27138 [6]	94	NQA	0	0	45	0	11	0	1	0	0	0	0
VSL-07R1010-1 [20]	89	NQA and QARD	68	0	0	0	33	0	55	0	7	0	0
PNNL-28838 [21]	82	NQA	82	0	41	0	41	41	0	0	41	0	0
PNNL-10918 [22]	79	GLP	78	0	0	0	39	0	0	0	0	0	0
PNNL-13744 [23]	78	NQA	75	0	0	0	0	0	0	0	0	63	0
VSL-07R1130-1 [24]	78	NQA	45	0	0	0	37	37	0	0	48	0	39
PNNL-26630 [25]	75	NQA	72	0	36	0	36	36	0	0	38	0	0
SRNS-STI-2008-00057 [26]	75	QARD	90	0	45	0	2	0	31	0	0	0	0
VSL-03R3460-1 [27]	71	NQA	96	0	0	0	94	76	0	0	66	0	59
PNNL-17950 [28]	69	GLP	72	0	66	40	35	0	37	0	0	0	0
INEEL-EXT-99-01322 [29]	67	GLP	67	0	0	50	64	0	0	0	0	67	0
VSL-05R5780-2 [30]	65	QARD	35	0	0	0	9	0	65	0	6	0	0
INEEL/EXT-02-00386 [31]	64	QARD	63	0	0	62	62	0	0	0	0	63	0
WSRC-STI-2007-00477 [32]	64	QARD	124	0	0	0	0	0	0	0	0	0	0
PNNL-11790 [33]	63	QARD	0	0	0	63	0	0	0	0	0	0	0

Reference	Number of Unique Glass IDs	QA	PCT	TCLP	Nepheline	T _L	Viscosity	EC	Sp950	T _g	SO ₃	Density	K-3
Fu and Pegg 1998 [34]	62	Unknown	17	38	0	58	15	13	0	16	0	6	0
SRNL-STI-2009-00778 [35]	60	QARD	234	0	150	0	0	0	0	0	0	0	0
WSRC-TR-2000-00351 [36]	60	QARD	87	0	0	0	0	0	0	0	0	0	0
Vienna, 2002, Dissertation [37]	59	GLP	0	0	0	59	0	0	53	0	0	0	0
PNNL-31556 [38]	50	NQA	0	0	0	0	25	25	0	0	25	0	0
VSL-09R1510-2 [39]	50	NQA	46	0	0	0	31	31	0	0	41	0	26
Bulkley and Vienna 1997 [40]	45	GLP	39	0	0	0	35	0	0	0	0	0	0
PNNL-26057 [41]	45	NQA	0	0	45	0	0	0	0	0	0	0	0
WSRC-TR-2004-00313 [42]	45	QARD	45	0	0	0	0	0	0	0	0	0	0
Crum et al. 1997 [43]	44	Unknown	0	0	0	42	0	0	0	0	0	0	0
INEEL-EXT-98-00970 [44]	44	GLP	44	0	0	28	36	0	0	0	0	0	0
VSL-06R6900-1 [45]	44	NQA	40	0	0	0	36	36	0	0	36	0	19
PNNL-29847 [46]	42	NQA	0	0	20	0	20	11	0	0	20	0	0
WSRC-TR-2002-00549 [47]	42	QARD	84	0	0	0	0	0	0	0	0	0	0
Mika et al. 1997 [48]	41	GLP	0	0	0	47	0	0	20	0	0	0	0
PNNL-10987 [49]	40	GLP	76	0	39	32	25	0	0	0	0	0	0
VSL-06R1240-1 [50]	40	QARD	50	0	0	0	40	0	40	0	1	0	0
VSL-06R6780-2 [51]	40	QARD	40	0	0	0	38	0	40	0	0	0	0
WSRC-STI-2007-00659 [52]	40	Unknown	154	0	69	0	0	0	0	0	0	0	0
WSRC-TR-2002-00482 [53]l	40	QARD	80	0	0	0	0	0	0	0	0	0	0
PNNL-20774 [54]	39	GLP	28	0	17	9	0	4	14	0	0	0	0
VSL-03R3760-2 [55]	39	NQA and QARD	6	9	0	18	3	3	0	7	0	18	0
SRNL-L3100-2011-00169 [56]	38	GLP	0	0	38	0	0	0	0	0	0	0	0
SRNL-STI-2010-00566 [57]	38	QARD	76	0	0	0	0	0	0	0	0	0	0
SRNS-STI-2008-00056 [58]	38	QARD	76	0	0	0	36	0	38	0	0	0	0
VSL-05R5460-1 [59]	37	NQA	47	3	0	0	10	2	0	0	0	0	0
WSRC-RP-95-0539 [60]	37	QARD	37	0	0	0	0	0	0	0	0	0	0
SRNL-STI-2011-00440 [61]	34	QARD	68	0	0	0	0	0	0	0	0	0	0

Reference	Number of Unique Glass IDs	QA	PCT	TCLP	Nepheline	T _L	Viscosity	EC	Sp950	T _g	SO ₃	Density	K-3
WSRC-TR-2002-00120 [62]	34	QARD	68	0	0	0	0	0	0	0	0	0	0
SRNL-STI-2009-00465 [63]	32	QARD	64	0	0	0	0	0	0	0	0	0	0
VSL-10R1790-1 [64]	32	NQA	32	0	0	0	9	9	0	0	30	0	4
VSL-16R4000-1 [65]	32	NQA	32	0	0	0	32	32	0	0	28	0	26
WTP-RPT-085 [66]	31	QARD	0	0	0	0	0	0	11	0	0	0	0
Johnston et al. 1990 [67]	30	GLP	30	0	0	0	0	0	0	0	0	0	0
Jiricka et al. 2003 [68]	30	GLP	0	0	0	0	0	0	29	0	0	0	0
WSRC-MS-93-222 [69]	30	GLP	30	0	0	0	0	0	0	0	0	0	0
WSRC-STI-2007-00616 [70]	30	GLP	60	0	0	0	0	0	0	0	0	0	0
SRT-SCS-98-017 [71]	30	QARD	30	0	0	0	0	0	0	0	0	0	0
SRNL-STI-2009-00608 [72]	29	Unknown	58	0	29	0	0	0	0	0	0	0	0
PNNL-12202 [73]	29	GLP	28	0	0	29	0	0	0	0	0	0	0
WSRC-TR-2003-00287 [74]	29	QARD	58	0	0	0	0	0	0	0	0	0	0
INEEL-EXT-2000-00110 [75]	28	QARD	8	0	0	6	0	0	0	0	0	0	0
PNNL-12208 [76]	28	GLP	0	0	0	31	0	0	4	0	0	0	0
SRNL-STI-2011-00063 [77]	28	QARD	56	0	0	0	0	0	0	0	0	0	0
SRNS-STI-2008-00055 [78]	28	GLP	56	0	0	0	0	0	0	0	0	0	0
WSRC-TR-2006-00006 [79]	28	GLP	28	0	28	0	0	0	0	0	0	0	0
PNNL-14050 [80]	27	GLP	50	0	0	0	14	0	0	0	0	0	0
VSL-06R6480-3 [81]	27	NQA	68	0	0	0	48	26	0	0	0	0	37
WSRC-TR-2001-00166 [82]	27	QARD	54	0	0	0	0	0	0	0	0	0	0
WSRC-TR-99-00332 [83]	27	QARD	27	0	0	0	0	0	0	0	0	0	0
PNNL-27555 [84]	26	NQA	0	0	26	0	0	0	0	0	0	0	0
SRNS-STI-2008-00060 [85]	26	QARD	52	0	0	0	0	0	0	0	0	0	0
VSL-08R1410-1 [86]	26	NQA	15	0	2	0	0	0	0	0	0	0	0
WSRC-TR-2001-00131 [87]	26	QARD	52	0	0	0	0	0	0	0	0	0	0
SRNS-STI-2008-00099 [88]	25	Unknown	75	0	25	0	0	0	0	0	0	0	0
PNNL-13483 [89]	24	GLP	48	0	0	0	0	0	0	0	0	0	0
PNNL-6320-4 [90]	24	Unknown	24	0	0	0	0	0	0	0	0	0	0

Reference	Number of Unique Glass IDs	QA	PCT	TCLP	Nepheline	T _L	Viscosity	EC	Sp950	T _g	SO ₃	Density	K-3
WSRC-TR-2000-00358 [91]	24	QARD	48	0	0	0	0	0	0	0	0	0	0
VSL-03R3460-2 [92]	22	NQA	53	0	0	0	45	28	0	0	0	0	12
VSL-15R3290-1 [93]	22	NQA	10	0	0	0	11	11	0	0	20	0	24
VSL-12R2540-1 [94]	21	NQA	0	0	0	0	0	0	0	0	23	0	0
VSL-14R3050-1 [95]	21	NQA	21	0	0	0	15	20	0	0	20	0	10
WSRC-TR-2001-00511 [96]	21	QARD	42	0	0	0	0	0	0	0	0	0	0
Li et al. 1997 [97]	20	NQA	26	0	20	0	0	0	0	0	0	0	0
PNL-10191 [98]	20	Unknown	20	0	0	0	20	0	0	0	0	0	0
PNNL-29023 [99]	20	NQA	0	0	20	0	0	0	0	0	0	0	0
SRNL-STI-2011-00178 [100]	20	QARD	40	0	0	0	0	0	0	0	0	0	0
VSL-08R1360-1 [101]	20	QARD and NQA	13	0	30	0	7	0	11	0	2	0	0
VSL-13R2940-1 [102]	20	NQA	20	0	0	0	18	20	0	0	20	0	7
PNNL (20) ^(c)	227	Multiple	61	0	3	1	1	0	44	0	0	5	0
CUA (52)	337	Multiple	209	27	45	1	208	119	10	3	81	2	77
SRNL (26)	322	Multiple	487	4	43	0	1	0	10	0	0	0	0
Others (20)	102	Multiple	36	0	65	7	8	1	8	15	0	39	0

(a) QARD or QARD equivalent program.

(b) Different QA programs were used for different properties.

(c) Value in parenthesis is the total number of studies.

3.0 Data Qualification Plan

In this section, one potential Hanford HLW flowsheet – the Direct Feed High-Level Waste (DFHLW) flowsheet – and the EWG formulation method are introduced (Section 3.1). Note that no decision has been made to implement a DFHLW flowsheet; however, this case represents the largest anticipated data gaps for current DOE vitrification projects.

Section 3.2 presents the glasses formulated by the EWG method, showing the glass composition region of potential interest for Hanford DFHLW treatment. These formulated glasses are then compared to the compiled database to identify the potential compositional gaps to fill for DFHLW.

Data gaps and coverage were evaluated by comparing the single- and double-glass-component concentration distributions for existing data with the compositions of glasses anticipated for the Hanford DFHLW flowsheet. Because the database has multiple dimensions, including more than 20 significant glass composition terms, over 10 properties, and various QA program requirements, two different plotting methods were used to illustrate data gaps as functions of composition with QA qualification and property parameters. These plotting methods are violin plots (Section 3.3) and heatmaps with scatter plots (Section 3.4). The violin plots show the distribution of glass components in one composition dimension while the scatterplots show distribution of glass components in two composition dimensions. Each plot is then evaluated to address the following questions:

- What compositional spaces are potential regions of interest for Hanford DFHLW treatment?
- Are existing data available to cover these regions by each property? Is there a need to make new glasses and/or new measurements for different properties?
- If data are available, were they collected under a sufficient QA program for Hanford HLW treatment? If not, how should the insufficient-QA data be qualified?

Lastly, Section 3.5 presents a table summarizing the compositional gaps by property and Section 3.6 proposes a plan for qualifying insufficient-QA data for Hanford HLW treatment.

3.1 Glass Formulation by EWG Method

Glass compositions were formulated to immobilize estimated Hanford DFHLW waste feed compositions developed by Britton (2023) [103]. The waste composition estimate assumed that sludge and insoluble fraction of salt cake from the south-east quadrant of Hanford tanks (A, AN, AP, AX, AW, AY, and AZ farms) would be blended and retrieved in roughly 500,000-gal batches. The waste would be delivered using average A-Complex supernate with a carrier fluid density of 1.2 g/mL (roughly 4 molar Na⁺). As there is uncertainty in how high of a solids loading could be effectively mobilized, mixed, sampled, and transported, four sets of waste feed were estimated with different prospective undissolved solids loadings (SOLs = 5, 10, 15, and 20 wt%). This waste feed projection includes a total of 154 batches of feed. These batches of feed assume four different SOLs to cover the likely range (78 batches for 5 wt% SOL, 35 batches for 10 wt% SOL, 23 batches for 15 wt% SOL, and 18 batches for 20 wt% SOL).

Figure 3.1 shows the non-volatile waste component concentration on an oxide and halogen mass fraction basis (*mf*) distributions from a DFHLW washing and blending study campaign inventory.

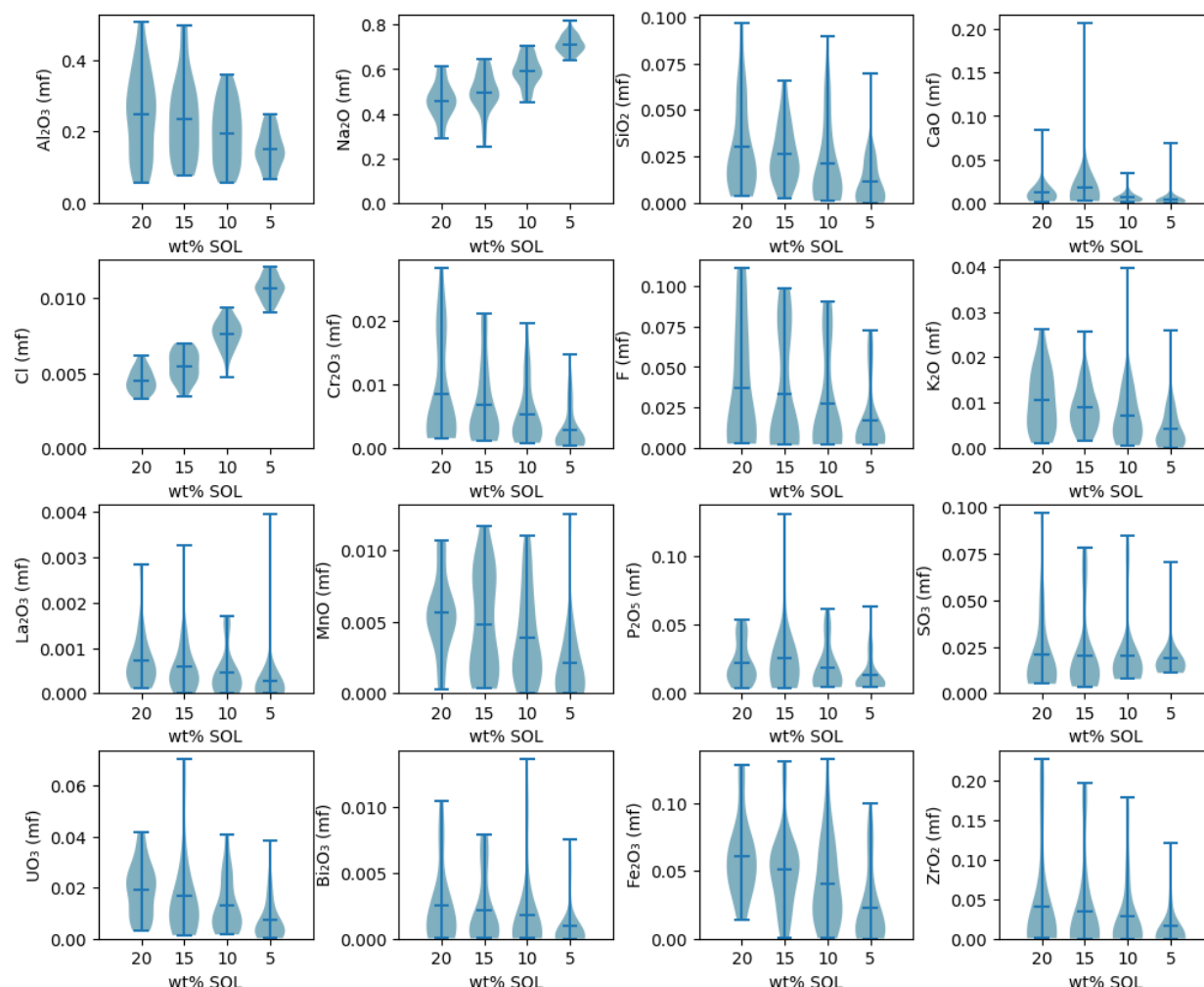


Figure 3.1. Waste compositions (mass fraction) from revised DFHLW washing and blending study campaign inventory.

The EWG property models and constraints [5], summarized in Table 3.1 and Table 3.2, were used to formulate glasses with two minor adjustments [104]:

1. Component constraint for F was increased from 2.5 to 4.5 wt% according to a study by Jin et al. [105], where $F \leq 4.5$ wt% was recommended as a relatively conservative limit for HLW glass formulation based on crystallinity and PCT data.
2. A model validity limit from the HLW SO_3 solubility model in Vienna et al. [5] for V_2O_5 (≤ 4.056 wt%) was added due to concern about V_2O_5 solubility in borosilicate glasses.

Table 3.1. Glass property constraints [5].

Constraint	Limit
PCT responses	$\ln[r_B \text{ (g/m}^2\text{)}] \leq 1.386^{(a)}$
	$\ln[r_{Na} \text{ (g/m}^2\text{)}] \leq 1.386^{(a)}$
	$\ln[r_{Li} \text{ (g/m}^2\text{)}] \leq 1.386^{(a)}$
Nepheline in CCC glass ^(b)	$p \leq 0.3$ (probability)
Spinel	$T_{2\%} \leq 950^\circ\text{C}$
Zirconium-containing phases	$T_L\text{-Zr} \leq 1050^\circ\text{C}$ if $g_{ZrO_2} > 0.04$
Viscosity at 1150°C	$1.386 \leq \ln(\eta_{1150}, \text{Pa}\cdot\text{s}) \leq 1.792^{(c)}$
P ₂ O ₅ and CaO concentrations	$g_{P_2O_5} \times g_{CaO} \leq 0.00065$ (mass fraction) ²
Salt, SO ₃ concentration	$g_{SO_3} \leq g_{SO_3}^{Limit}$
Eskolaite formation	$g_{Cr_2O_3} \leq 0.03$
B ₂ O ₃ and SiO ₂ concentrations	$g_{SiO_2} + g_{B_2O_3} \geq 0.32$
Noble metal	$g_{PdO} + g_{Rh_2O_3} + g_{RuO_2} \leq 0.0025$

(a) This corresponds to $r_a \leq 4 \text{ g/m}^2$ (or 8 g/L in normalized loss units).
 (b) CCC is container centerline cooled glass sample.
 (c) This corresponds to $4 \leq \eta_{1150} \leq 6 \text{ Pa}\cdot\text{s}$.

Table 3.2. Glass component concentration constraints, in mass fractions [5].

Component	Min	Max
Al ₂ O ₃	0.019	0.300
B ₂ O ₃	0.040	0.220
Bi ₂ O ₃	0	0.070
CaO	0	0.100
CdO	0	0.015
Cr ₂ O ₃	0	0.030
F	0	0.045 ^(a)
Fe ₂ O ₃	0	0.200
K ₂ O	0	0.060
Li ₂ O	0	0.060
MgO	0	0.060
MnO	0	0.080
Na ₂ O	0.041	0.240
NiO	0	0.030
P ₂ O ₅	0	0.045
SiO ₂	0.220	0.530
SrO	0	0.101
ThO ₂	0	0.060
TiO ₂	0	0.050
UO ₃	0	0.063
V ₂ O ₅	0	0.04056 ^(a)
ZnO	0	0.040
ZrO ₂	0	0.135

(a) Upper limits for F and V₂O₅ differ from Vienna et al. 2016 for reasons described in the text.

A computer program, written in Python, was developed with an optimization package GEKKO [106] to optimize glass formulations with maximum waste loadings while simultaneously satisfying all the constraints in Table 3.1 and Table 3.2. The 12 current Hanford glass-forming chemicals plus V_2O_5 were varied – kyanite, boric acid, wollastonite, lithium carbonate, olivine, sodium carbonate, vanadium oxide, rutile, zincite, zircon, and sucrose). PyCharm 2022.3 (Community Edition, developed by JetBrains) was used as an integrated development environment. Calculation sheets using Microsoft Excel Solver were generated to spot check (10% of batches) and confirm the results obtained from the Python codes.

3.2 EWG Glasses

Figure 3.2 and Figure 3.3 show the waste loading ranges and glass compositions formulated with different SOLs, respectively. Waste loading decreases with decreasing SOL, mostly due to the increasing Na_2O concentration in the wastes. Since the volume is the same for all the batches shown in Figure 3.3, lower SOL results in more batches. Figure 3.4 shows the waste loading limiting factors of these glasses, where nepheline, viscosity, and SO_3 solubility are the most limiting.

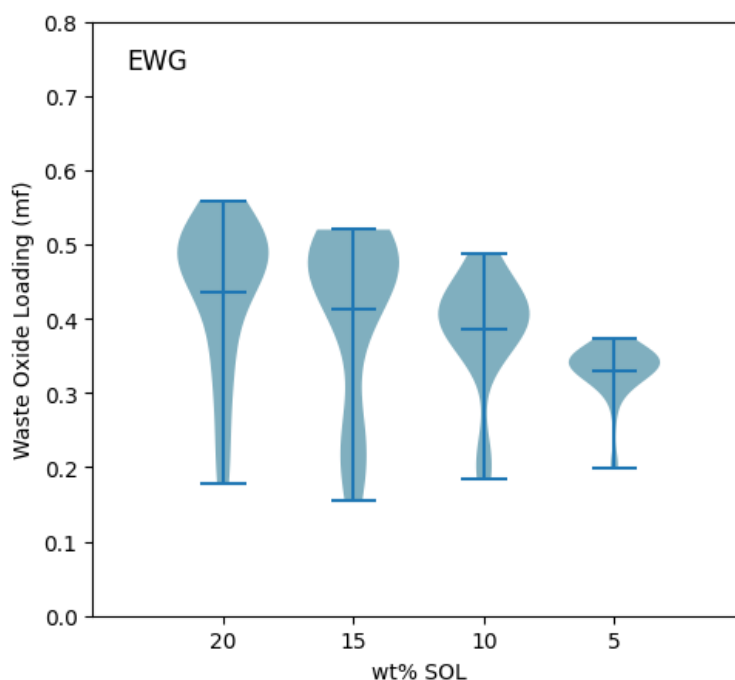


Figure 3.2. Waste oxide loading (mass fraction) of the glasses.

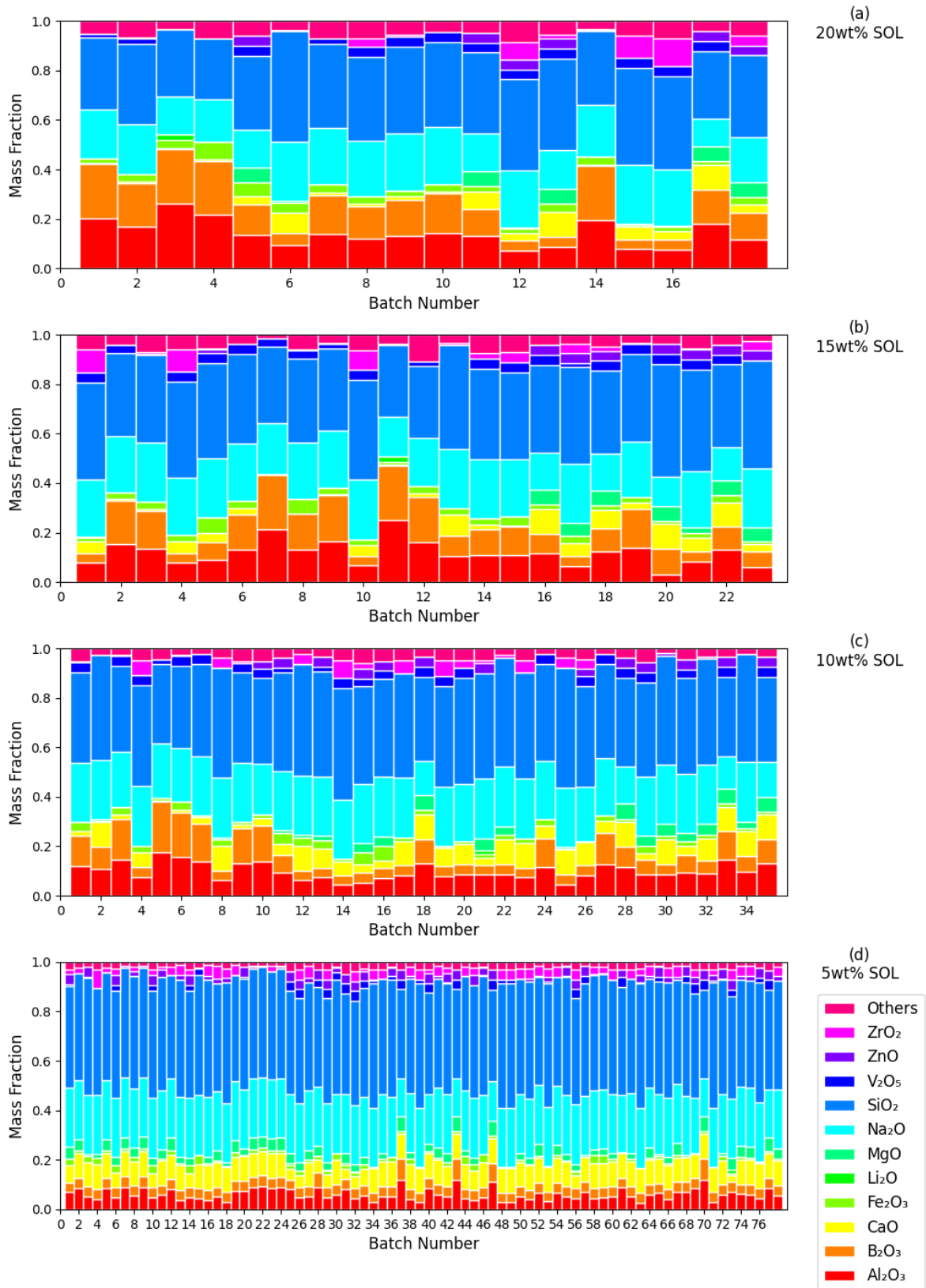


Figure 3.3. Glass compositions (mass fraction).

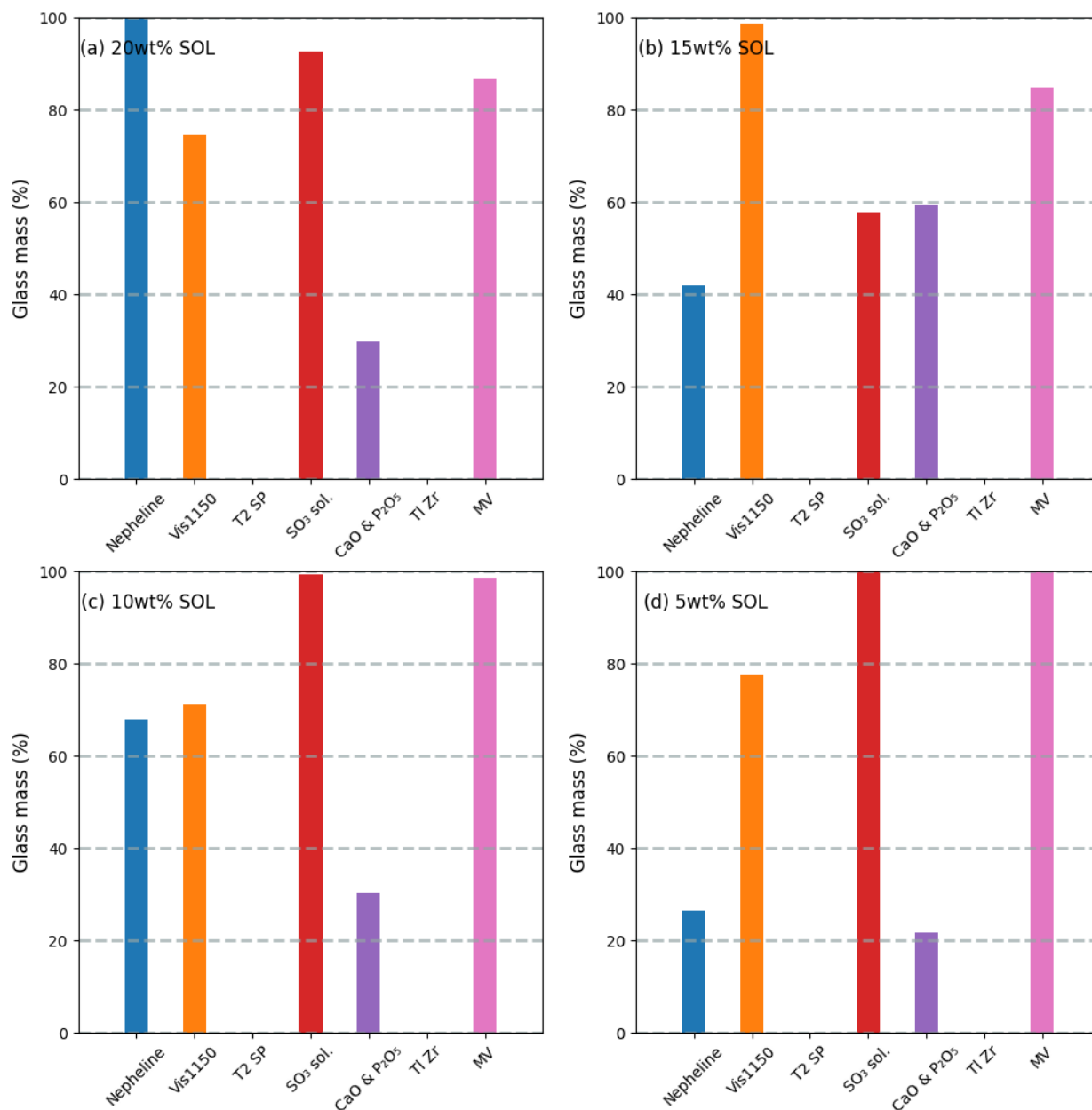


Figure 3.4. Waste loading limiting factors (nepheline formation, viscosity at 1150 °C, temperature at two vol% spinel with the melt, SO₃ solubility, CaO×P₂O₅ constraint, liquidus temperature of zirconium-containing phases, and model validity limits) of the glass masses.

3.3 One-Dimensional Composition Comparisons

Figure 3.5 shows an example violin plot and the type of information it provides (e.g., distribution of the number of glasses at a component concentration, minimum, median, average, and maximum values, as well as the range for 20 to 80 percentile). Figure 3.6 shows a violin plot of PCT data in the database (pass QA requirement and need QA qualification) compared to the target DFHLW glasses. Violin plots of the other properties can be found in Appendix A.

These violin plots present a statistical comparison of each individual component, providing information on whether compositional gaps exist in one-dimensional space. For example, in Figure 3.6, the existing data in the compiled database cover the projected compositional space (or range) of Al_2O_3 in the current target DFHLW glasses, indicating no composition gap in terms of Al_2O_3 . Note that the current target glass compositions reported here might change slightly due to updated waste feed inputs in the future. On the other hand, the existing data in the compiled database, especially the data with sufficient QA program, did not cover the whole compositional spaces of V_2O_5 in the target DFHLW glasses, indicating a composition gap at high V_2O_5 concentrations. Section 3.5 summarizes the composition gaps identified.

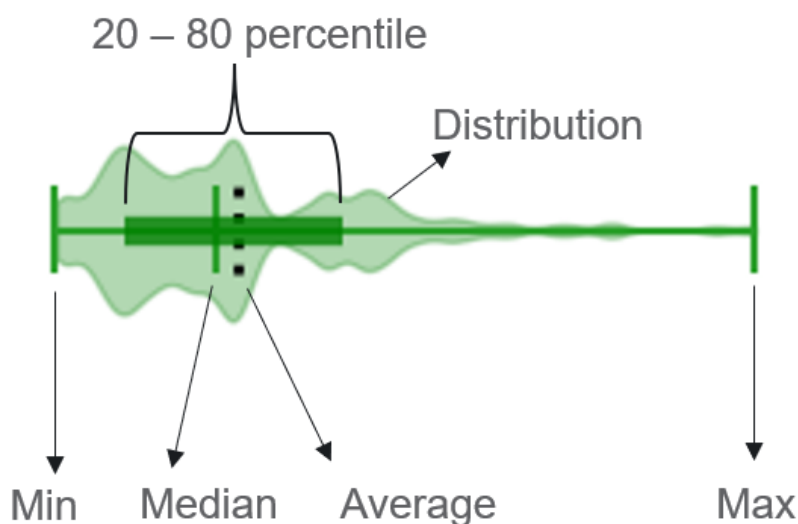


Figure 3.5. Example violin plot and the information it provides.

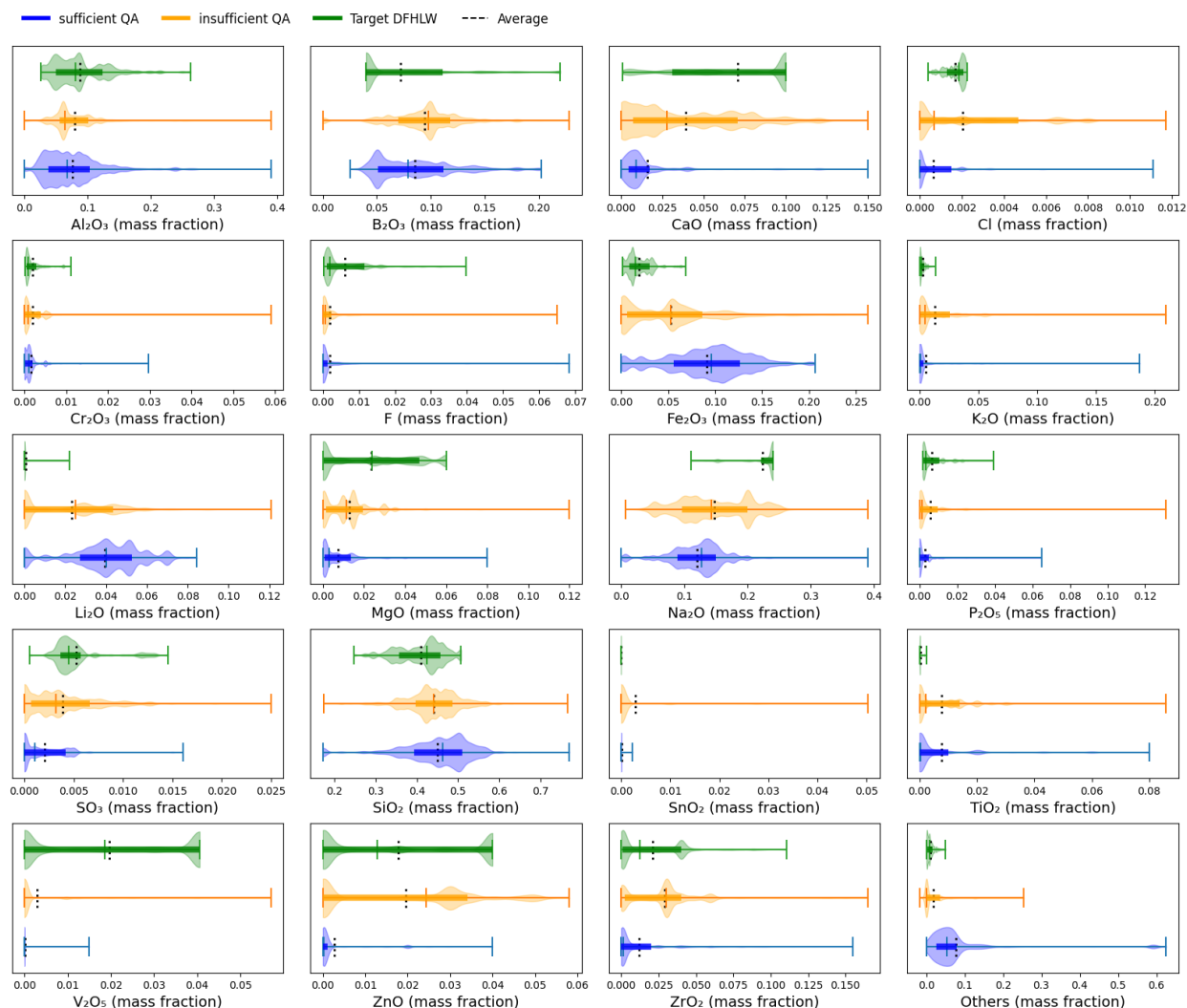


Figure 3.6. Violin plots of PCT Na data in the database (sufficient-QA and insufficient-QA data), along with target DFHLW glasses.

3.4 Two-Dimensional Composition Comparisons

To examine paired compositional spaces, heatmaps with scatter plots were generated for every possible combination of two components from the following major components: Al_2O_3 , B_2O_3 , CaO , Fe_2O_3 , K_2O , Li_2O , MgO , Na_2O , SiO_2 , ZnO , and ZrO_2 , and “Others” (i.e., components other than those listed here). Due to the total number of figures (over 2000), only example heatmap figures are presented in this section; other figures can be found in Appendix A. A summary of the compositional gaps identified can be found in Section 3.5.

Example 1: Determination of whether qualification of insufficient-QA data is needed. Figure 3.7 shows the ZnO versus CaO of PCT data in the database (heatmap) compared to the target DFHLW glasses (scatter points). Comparing the figures of the whole dataset (Figure 3.7a) and QA data only (Figure 3.7b), shows that the high ZnO and/or high CaO regions are only covered by the insufficient-QA data. Therefore, qualification of these insufficient-QA data would allow Hanford to process the targeted DFHLW glasses.

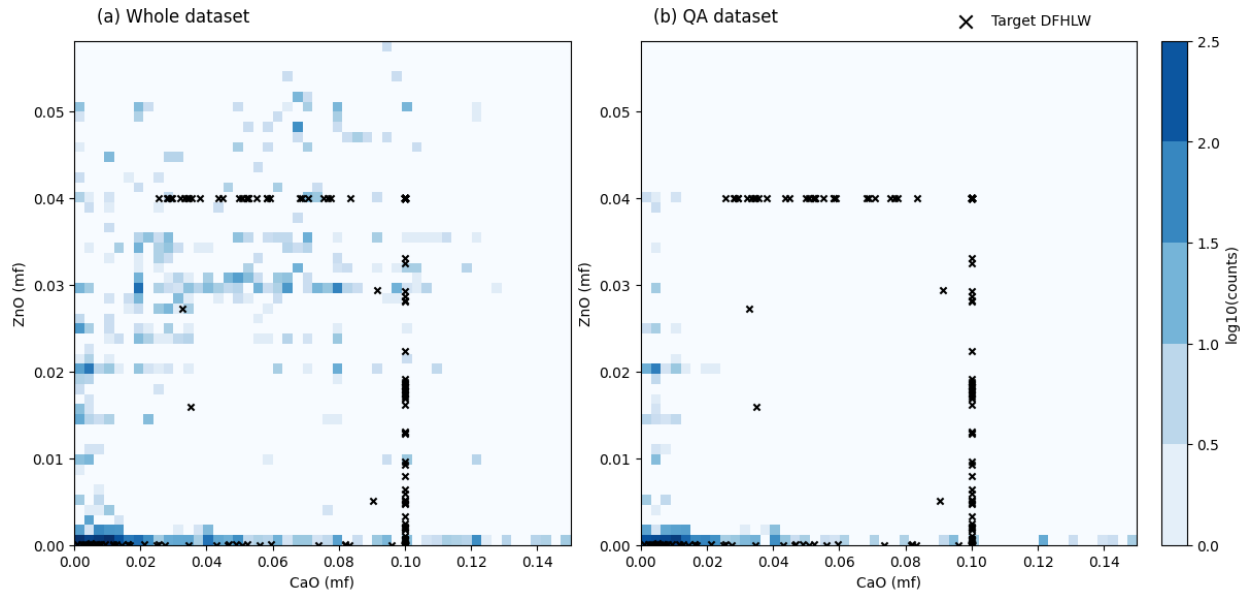


Figure 3.7. ZnO versus CaO of PCT B data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

Example 2: Determination of whether new glasses and/or measurements are needed. Figure 3.8 shows B_2O_3 versus Al_2O_3 of K-3 data in the database compared to the target DFHLW glasses. Since many EWG glass compositional regions are not covered by any data in the database, more glasses and tests on K-3 refractory corrosion would allow Hanford to process in the target DFHLW composition region.

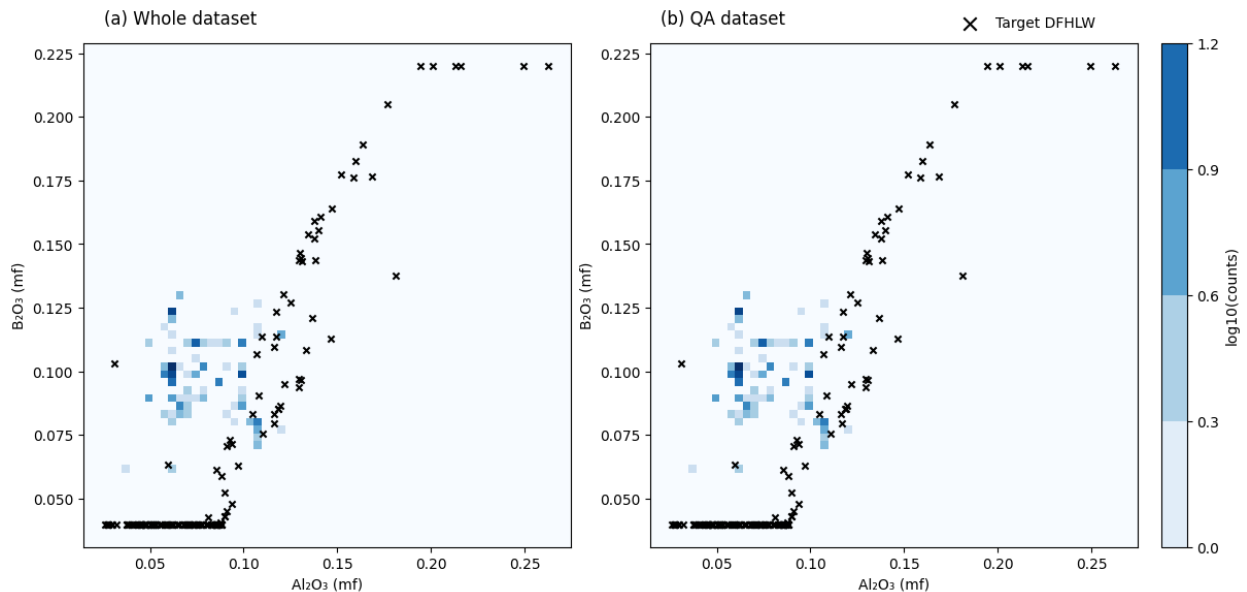


Figure 3.8. B_2O_3 versus Al_2O_3 of K-3 data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

Example 3: Determination of whether any waste loading limiting constraints need to be revised. Figure 3.9 shows P_2O_5 versus CaO of nepheline data in the database compared to the target DFHLW glasses. As shown in the figure, many target DFHLW glasses are limited by the $P_2O_5 \times CaO$ constraint, which was constructed to avoid immiscible liquid or crystalline phosphate phase separation [107]. This constraint was primarily obtained based on the data available at the time, which could be revised after qualifying the insufficient-QA data in that region and/or after new glasses are made and tested. Revising the limiting constraints will enhance or increase the compositional processing envelope, leading to many benefits such as higher waste loading and more flexibility for flowsheet options (e.g., DFHLW).

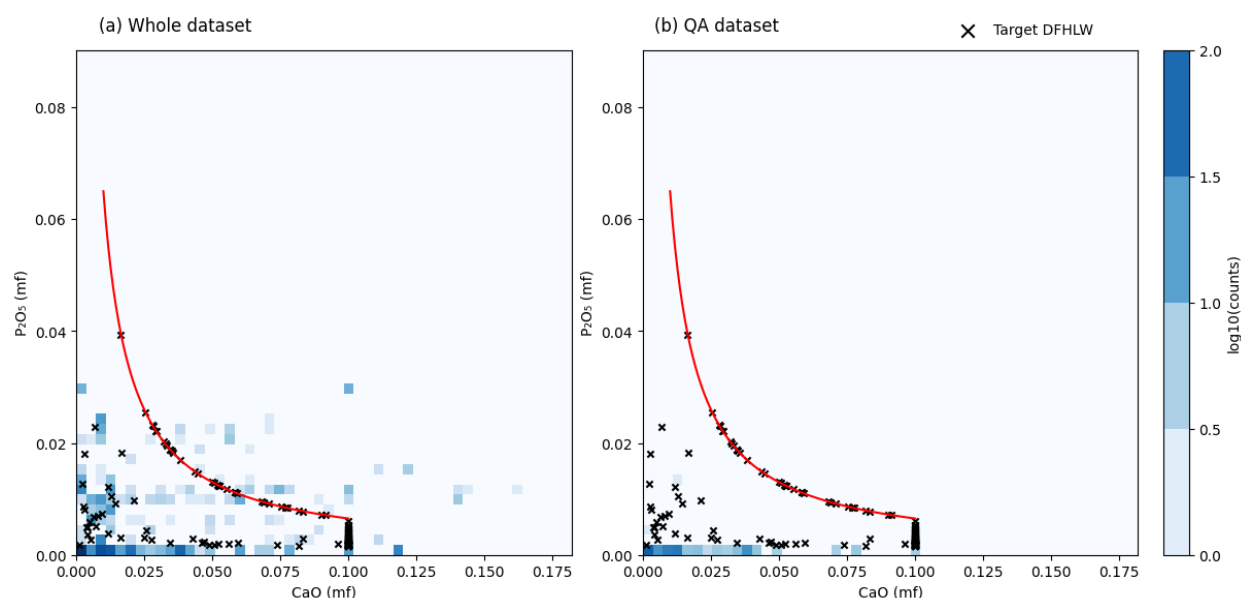


Figure 3.9. P_2O_5 versus CaO of nepheline data in the database (heatmap) with target DFHLW glasses (scatter black crosses). Red line represents the $P_2O_5 \times CaO$ constraint.

3.5 Summary of Composition Gaps

Table 3.3 summarizes the compositional gaps identified through examining the violin plots and heatmap with scatter plots for each of the 11 properties. The table includes information on whether new glasses/measurements are necessary, whether the qualification of insufficient-QA data is needed, and the compositional ranges of the gaps, as well as the basis for obtaining the compositional ranges for each glass component. For example, the first row in Table 3.3 indicates a need for new data in low B_2O_3 (<0.05 mass fraction) with varying MgO, V_2O_5 , and/or ZnO for density. The third row of Table 3.3 indicates a need for both new data and qualifying insufficient-QA data for density in low SiO_2 (<0.3 mass fraction) region.

In general, these compositional gaps exist across multiple properties:

- High Na_2O , CaO, MgO, and V_2O_5
- Low SiO_2 and B_2O_3

Table 3.3. Summary table of the composition gaps (range shown in mass fraction) by property.

Property	Composition Gaps	Need New Glass?	Covered by Insufficient-QA Data?	Comp. 1	Range 1	Comp. 2	Range 2	Note: Basis
Density	Low B ₂ O ₃ , varying MgO, V ₂ O ₅ and/or ZnO	Yes	No	B ₂ O ₃	< 0.05			Heatmap Figure B.1 to Figure B.3
Density	High Na ₂ O, CaO and V ₂ O ₅	Yes	No	Na ₂ O	> 0.24	CaO	> 0.1	Heatmap Figure B.4 and Figure B.5
Density	Low SiO ₂	Yes	Yes	SiO ₂	< 0.3			QA data min
Density	High CaO	Yes	Yes	CaO	> 0.07			EWG ave
Density	High Al ₂ O ₃	No	Yes	Al ₂ O ₃	> 0.2			QA data max
Density	High B ₂ O ₃	No	Yes	B ₂ O ₃	> 0.2			QA data max
Density	High F	No	Yes	F	> 0.01			QA data max
EC	High Cr ₂ O ₃	Yes	No	Cr ₂ O ₃	> 0.006			QA data max
EC	High F	Yes	No	F	> 0.013			QA data max
EC	High MgO	Yes	No	MgO	> 0.05			QA data max
EC	High ZrO ₂	Yes	No	ZrO ₂	> 0.09			QA data max
EC	Low B ₂ O ₃ and high V ₂ O ₅	Yes	No	B ₂ O ₃	< 0.04	V ₂ O ₅	> 0.04	Heatmap Figure B.6
EC	High MgO and high Na ₂ O	Yes	No	Na ₂ O	> 0.24	MgO	> 0.06	Heatmap Figure B.7
EC	High B ₂ O ₃	Yes	Yes	B ₂ O ₃	> 0.15			QA data max
EC	High Al ₂ O ₃	No	Yes	Al ₂ O ₃	> 0.08			EWG median
EC	Low SiO ₂	No	Yes	SiO ₂	< 0.335			QA data min
K-3	High Al ₂ O ₃	Yes	No	Al ₂ O ₃	> 0.14			QA data max
K-3	Low and high B ₂ O ₃	Yes	No	B ₂ O ₃	< 0.06 or > 0.137			QA data min/max
K-3	High Cr ₂ O ₃	Yes	No	Cr ₂ O ₃	> 0.006			QA data max
K-3	High F	Yes	No	F	> 0.005			QA data max
K-3	High MgO	Yes	No	MgO	> 0.05			QA data max
K-3	High P ₂ O ₅	Yes	No	P ₂ O ₅	> 0.034			QA data max

Property	Composition Gaps	Need New Glass?	Covered by Insufficient-QA Data?	Comp. 1	Range 1	Comp. 2	Range 2	Note: Basis
K-3	Low SiO ₂	Yes	No	SiO ₂	< 0.34			QA data min
K-3	High SO ₃	Yes	No	SO ₃	> 0.0132			QA data max
K-3	Low ZnO	Yes	No	ZnO	< 0.01			QA data min
K-3	High ZrO ₂	Yes	No	ZrO ₂	> 0.067			QA data max
K-3	Low B ₂ O ₃ varying V ₂ O ₅ or High V ₂ O ₅ varying B ₂ O ₃	Yes	No	B ₂ O ₃	< 0.05	V ₂ O ₅	> 0.04	Heatmap Figure B.8
Nepheline	High B ₂ O ₃	No	Yes	B ₂ O ₃	> 0.2			QA data max
Nepheline	High CaO	Yes	Yes	CaO	> 0.1			QA data max
Nepheline	High Cl	Yes	Yes	Cl	> 0			QA data max
Nepheline	High F	Yes	Yes	F	> 0.008			QA data max
Nepheline	Low Li ₂ O	No	Yes	Li ₂ O	< 0.005			QA data min
Nepheline	High Na ₂ O	Yes	Yes	Na ₂ O	> 0.2			QA data max
Nepheline	High P ₂ O ₅	No	Yes	P ₂ O ₅	> 0.033			QA data max
Nepheline	High SO ₃	No	Yes	SO ₃	> 0.007			QA data max
Nepheline	Low SiO ₂	No	Yes	SiO ₂	< 0.323			QA data min
Nepheline	High V ₂ O ₅	Yes	Yes	V ₂ O ₅	> 0.0			QA data max
Nepheline	High ZnO	Yes	Yes	ZnO	> 0.02			QA data max
PCT	Low B ₂ O ₃ varying MgO or high MgO varying B ₂ O ₃	Yes	No	B ₂ O ₃	< 0.05	MgO	> 0.06	Heatmap Figure B.9
PCT	Low B ₂ O ₃ varying V ₂ O ₅ or high V ₂ O ₅ varying B ₂ O ₃	Yes	No	B ₂ O ₃	< 0.05	V ₂ O ₅	> 0.04	Heatmap Figure B.10
PCT	Low B ₂ O ₃ varying ZnO	Yes	No	B ₂ O ₃	< 0.05			Heatmap Figure B.11
PCT	High B ₂ O ₃	No	Yes	B ₂ O ₃	> 0.2			QA data max
PCT	High V ₂ O ₅	No	Yes	V ₂ O ₅	> 0.0			QA data ave
PCT	High Na ₂ O	No	Yes	Na ₂ O	> 0.24			EWG ave
PCT	High CaO	No	Yes	CaO	> 0.1			EWG max
PCT	High ZnO	No	Yes	ZnO	> 0.04			EWG max
SO ₃	High Al ₂ O ₃	Yes	No	Al ₂ O ₃	> 0.24			QA data max
SO ₃	High B ₂ O ₃	Yes	No	B ₂ O ₃	> 0.2			QA data max

Property	Composition Gaps	Need New Glass?	Covered by Insufficient-QA Data?	Comp. 1	Range 1	Comp. 2	Range 2	Note: Basis
SO ₃	High Cr ₂ O ₃	Yes	No	Cr ₂ O ₃	> 0.01			QA data max
SO ₃	High F	Yes	No	F	> 0.03			QA data max
SO ₃	Low SiO ₂	Yes	No	SiO ₂	< 0.27			QA data min
Sp950	High F	Yes	No	F	> 0.02			QA data max
Sp950	High SO ₃	Yes	No	SO ₃	> 0.0045			EWG median
Sp950	High V ₂ O ₅	Yes	No	V ₂ O ₅	> 0.0			QA data ave
Sp950	Low B ₂ O ₃ varying CaO or high CaO varying B ₂ O ₃	Yes	No	B ₂ O ₃	< 0.04	CaO	> 0.1	Heatmap Figure B.12
Sp950	High Na ₂ O varying CaO or high CaO varying Na ₂ O	Yes	No	Na ₂ O	> 0.24	CaO	> 0.1	Heatmap Figure B.13
Sp950	Low Fe ₂ O ₃	Yes	Yes	Fe ₂ O ₃	< 0.014			QA data min
Sp950	High MgO	Yes	Yes	MgO	> 0.024			EWG median
Sp950	High Na ₂ O	Yes	Yes	Na ₂ O	> 0.24			EWG median
TCLP Cd	High CaO	Yes	No	CaO	> 0.1			EWG median
TCLP Cd	Low Fe ₂ O ₃	Yes	No	Fe ₂ O ₃	< 0.02			QA data min
TCLP Cd	High Al ₂ O ₃	Yes	Yes	Al ₂ O ₃	> 0.08			EWG median
TCLP Cd	High Cr ₂ O ₃	Yes	Yes	Cr ₂ O ₃	> 0.005			QA data max
TCLP Cd	High MgO	Yes	Yes	MgO	> 0.02			QA data max
TCLP Cd	High Na ₂ O	Yes	Yes	Na ₂ O	> 0.2			QA data max
TCLP Cd	High P ₂ O ₅	Yes	Yes	P ₂ O ₅	> 0.03			QA data max
TCLP Cd	High SO ₃	Yes	Yes	SO ₃	> 0.0045			EWG median
TCLP Cd	High V ₂ O ₅	Yes	Yes	V ₂ O ₅	> 0.0			QA data ave
TCLP Cd	Low SiO ₂	No	Yes	SiO ₂	< 0.3			QA data min
TCLP Cd	High ZnO	No	Yes	ZnO	> 0.216			QA data max
TCLP Cd	High ZrO ₂	No	Yes	ZrO ₂	> 0.1			QA data max
T _g	High F	Yes	No	F	> 0.003			QA data max
T _g	High Na ₂ O	Yes	No	Na ₂ O	> 0.2			QA data max
T _g	High ZnO	Yes	No	ZnO	> 0.022			QA data max
T _g	High Na ₂ O varying MgO or high MgO varying Na ₂ O	Yes	No	Na ₂ O	> 0.24			Heatmap Figure B.14

Property	Composition Gaps	Need New Glass?	Covered by Insufficient-QA Data?	Comp. 1	Range 1	Comp. 2	Range 2	Note: Basis
T _g	High P ₂ O ₅	Yes	Yes	P ₂ O ₅	> 0.0032			EWG median
T _g	High SO ₃	Yes	Yes	SO ₃	> 0.0045			EWG median
T _g	Low SiO ₂	Yes	Yes	SiO ₂	< 0.42			EWG median
T _g	High V ₂ O ₅	Yes	Yes	V ₂ O ₅	> 0.0			QA data ave
T _g	High B ₂ O ₃	No	Yes	B ₂ O ₃	> 0.2			QA data max
TL	High Al ₂ O ₃	Yes	No	Al ₂ O ₃	> 0.185			QA data max
T _L	High B ₂ O ₃	Yes	No	B ₂ O ₃	> 0.2			QA data max
T _L	High MgO	Yes	Yes	MgO	> 0.025			QA data max
T _L	High Na ₂ O	Yes	Yes	Na ₂ O	> 0.24			QA data max
T _L	High V ₂ O ₅	Yes	Yes	V ₂ O ₅	> 0.0			QA data ave
T _L	High ZnO	Yes	Yes	ZnO	> 0.013			EWG median
T _L	High Cr ₂ O ₃	No	Yes	Cr ₂ O ₃	> 0.0058			QA data max
T _L	High P ₂ O ₅	No	Yes	P ₂ O ₅	> 0.03			QA data max
T _L	High SO ₃	No	Yes	SO ₃	> 0.012			QA data max
Viscosity	High MgO and high Na ₂ O	Yes	No	Na ₂ O	> 0.24	MgO	> 0.06	Heatmap Figure B.15
Viscosity	High V ₂ O ₅	Yes	Yes	V ₂ O ₅	> 0.0			QA data ave
Viscosity	Low B ₂ O ₃	No	Yes	B ₂ O ₃	< 0.04			QA data min
Viscosity	High B ₂ O ₃	No	Yes	B ₂ O ₃	> 0.2			QA data max
Viscosity	High F	No	Yes	F	> 0.002			EWG ave

3.6 Qualification Plan

This section describes the plan to qualify the insufficient-QA data to meet the QA requirement (listed in Table 2.2) for the Hanford DFHLW process. Table 3.4 lists the number of studies (data source papers/reports) that need to be QA qualified for each property with a priority order. The priority order is based on the waste loading limiting constraints from the formulation results (shown in Figure 3.4). If the data sets cannot be qualified for use, then additional data covering those composition regions are needed. K-3 corrosion and SO₃ solubility are not listed in this table because all the available data for these two properties satisfy the QA requirement (Table 2.3).

Table 3.4. Number of studies that need to be QA qualified for each property, with a priority number. Arrows indicate the interest in the high- or low-concentration range of the components.

Property	Component of Interest	Number of Studies ^(a)	Priority
Nepheline	↑B ₂ O ₃ , ↑CaO, ↑Cl, ↑F, ↓Li ₂ O, ↑Na ₂ O, ↑P ₂ O ₅ , ↑SO ₃ , ↓SiO ₂ , ↑V ₂ O ₅ , ↑ZnO	29	1
Viscosity	↑V ₂ O ₅ , ↓B ₂ O ₃ , ↑F	9	1
PCT	↑B ₂ O ₃ , ↑V ₂ O ₅ , ↑Na ₂ O, ↑CaO, ↑ZnO	40	2
Sp950	↓Fe ₂ O ₃ , ↑MgO, ↑Na ₂ O	5	2
TCLP Cd	↑Al ₂ O ₃ , ↑Cr ₂ O ₃ , ↑MgO, ↑Na ₂ O, ↑P ₂ O ₅ , ↑SO ₃ , ↑V ₂ O ₅ , ↓SiO ₂ , ↑ZrO ₂	9	2
T _L	↑MgO, ↑Na ₂ O, ↑V ₂ O ₅ , ↑ZnO, ↑Cr ₂ O ₃ , ↑P ₂ O ₅ , ↑SO ₃	10	2
EC	↑B ₂ O ₃ , ↑Al ₂ O ₃ , ↓SiO ₂	2	2
T _g	↓SiO ₂ , ↑B ₂ O ₃	1	3
Density	↓SiO ₂ , ↑CaO, ↑Al ₂ O ₃ , ↑B ₂ O ₃ , ↑F	4	3

(a) A few studies overlap between these properties. Table 3.5 lists all the studies with citations.

Figure 3.10 is a flowchart of QA method selection, demonstrating the logic of assigning a data qualification approach for different studies. Each qualification method will follow the instructions in *QA Implementing Procedure: Qualification of Existing Data* [108]. The following paragraphs provide a brief introduction of each QA method.

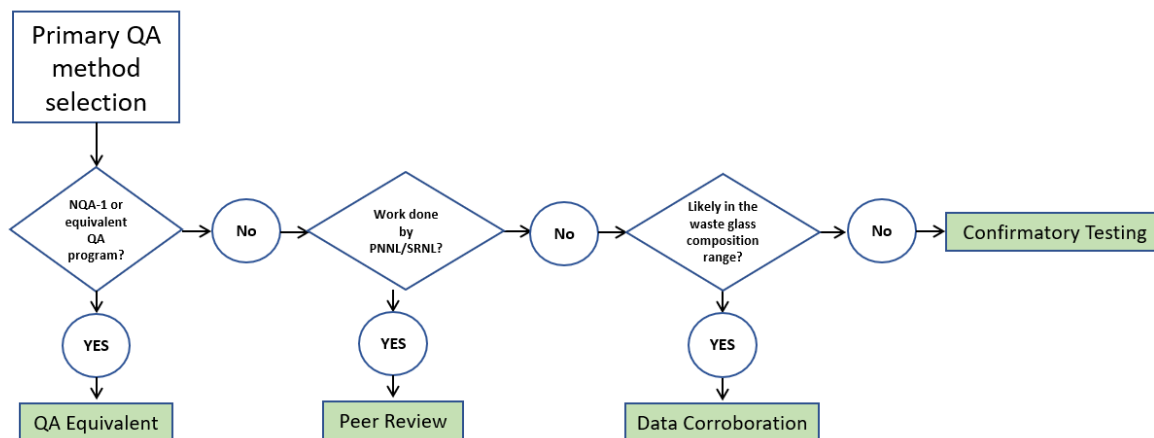


Figure 3.10. Flowchart of QA method selection.

- **QA equivalence** is determining whether the acquisition, development, or processing of subject data was performed in accordance with sound technical and administrative practices or procedures that can be demonstrated to generally meet the applicable requirements and guidance contained in the required QA program. The scope is to evaluate the extent to which conditions under which the data were generated meet the requirements and guidance contained in the required QA program and industry-acceptable practices or processes. The employed practices or procedures must demonstrate industry-acceptable scientific, engineering, or administrative practices or processes with appropriate compliance documentation.
- **Peer review** is determining whether sufficient information exists to provide reasonable assurance that the subject data is acceptable for use. The scope is to evaluate the extent to which conditions under which the data were generated meet the requirements and guidance contained in the required QA program and followed industry-acceptable scientific, engineering, or administrative practices or processes with appropriate compliance documentation.
- **Data corroboration** is determining whether existing data can be shown to substantiate or confirm parameter values of the subject data. The scope is to compare the subject data to existing qualified data and models to check if the difference is within uncertainties (e.g., experimental uncertainty and/or model prediction uncertainty).
- **Confirmatory testing** is determining whether confirmatory tests, designed and performed to generate directly comparable data, establish the quality of the subject data. The scope is to design and perform the test under the required QA program to generate comparable results for a significant subset of the study data.

Table 3.5 provides the qualification plan.

Table 3.5. Studies that need QA qualification and the assigned QA method.

Property	Component	Range	QA Level	Reference	QA Method
PCT	B ₂ O ₃	> 0.2	NQA-1	VSL-12R2470-1 [2]	QA equivalent
PCT	B ₂ O ₃	> 0.2	GLP	Marra et al., WM2010 [109]	Peer Review
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-01R3560-2 [17]	QA equivalent
PCT	V ₂ O ₅	> 0.0	GLP	PNNL-14050 [80]	Peer Review
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-04R4770-1 [110]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-07R1240-04 [14]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-04R4960-1 [111]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-06R6900-1 [45]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-05R5900-1 [112]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-07R1130-1 [24]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-10R1790-1 [64]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-09R1510-2 [113]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-13R2940-1 [102]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-14R3050-1 [95]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-15R3290-1 [93]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-16R4000-1 [65]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-17R4140-1 [114]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-17R4230-1 [115]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	VSL-17R4250-1 [116]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	PNNL-26630 [25]	QA equivalent
PCT	V ₂ O ₅	> 0.0	NQA-1	PNNL-28838 [21]	QA equivalent
PCT	Na ₂ O	> 0.24	Unknown	WSRC-TR-93-672 [13]	Peer Review
PCT	Na ₂ O	> 0.24	GLP	WSRC-MS-93-222 [69]	Peer Review
PCT	Na ₂ O	> 0.24	NQA-1	PNNL-13744 [23]	QA equivalent
PCT	Na ₂ O	> 0.24	GLP	PNNL-10918 [22]	Peer Review
PCT	Na ₂ O	> 0.24	Unknown	WSRC-STI-2007-00659 [52]	Peer Review
PCT	Na ₂ O	> 0.24	GLP	PNNL-20774 [117]	Peer Review

Property	Component	Range	QA Level	Reference	QA Method
PCT	CaO	> 0.1	GLP	INEEL-EXT-99-01322 [29]	Peer Review
PCT	CaO	> 0.1	GLP	PNNL-13483 [89]	Peer Review
PCT	CaO	> 0.1	GLP	PNNL-17950 [28]	Peer Review
PCT	CaO	> 0.1	NQA-1	VSL-04R4480-1 [16]	QA equivalent
PCT	CaO	> 0.1	NQA-1	VSL-03R3460-1 [27]	QA equivalent
PCT	CaO	> 0.1	GLP	PNNL-11865 [118]	Peer Review
PCT	ZnO	> 0.04	GLP	Johnston et al. 1990, Letter Report [67]	Peer Review
PCT	ZnO	> 0.04	GLP	Matlack et al. 1997 [119]	Peer Review
PCT	ZnO	> 0.04	NQA-1	PNWD-3045 [120]	QA equivalent
PCT	ZnO	> 0.04	NQA-1	VSL-05R5460-1 [59]	QA equivalent
PCT	ZnO	> 0.04	NQA-1	VSL-03R3470-1 [121]	QA equivalent
PCT	ZnO	> 0.04	NQA-1	VSL-03R3460-2 [92]	QA equivalent
PCT	ZnO	> 0.04	NQA-1	VSL-06R6480-3 [122]	QA equivalent
TCLP	Al ₂ O ₃	> 0.08	Unknown	Matlack et al. 1997 [119]	Peer Review
TCLP	Al ₂ O ₃	> 0.08	Unknown	WSRC-MS-98-00494 [123]	Peer Review
TCLP	Al ₂ O ₃	> 0.08	Unknown	WSRC-MS-98-00447 [124]	Peer Review
TCLP	Al ₂ O ₃	> 0.08	GLP	Matlack et al. 1997 [119]	Peer Review
TCLP	Al ₂ O ₃	> 0.08	NQA-1	VSL-07R1240-04 [14]	QA equivalent
TCLP	Al ₂ O ₃	> 0.08	NQA-1	VSL-03L3780-1 [125]	QA equivalent
TCLP	Cr ₂ O ₃	> 0.005	Unknown	Fu and Pegg 1998 [34]	Peer Review
TCLP	MgO	> 0.02	NQA-1	VSL-01R3560-2 [17]	QA equivalent
TCLP	MgO	> 0.02	NQA-1	VSL-05R5460-1 [59]	QA equivalent
Nepheline	B ₂ O ₃	> 0.22	GLP	McCloy et al. 2011 [126]	Peer Review
Nepheline	B ₂ O ₃	> 0.22	NQA-1	PNNL-20184 [19]	QA equivalent
Nepheline	B ₂ O ₃	> 0.22	NQA-1	VSL-10R1690-1 [127]	QA equivalent
Nepheline	B ₂ O ₃	> 0.22	GLP	SRNL-L3100-2011-00169 [56]	Peer Review
Nepheline	B ₂ O ₃	> 0.22	NQA-1	PNNL-27555 [84]	QA equivalent
Nepheline	B ₂ O ₃	> 0.22	NQA-1	PNNL-29023 [99]	QA equivalent
Nepheline	B ₂ O ₃	> 0.22	GLP	Kroll et al. 2019. [128]	Peer Review
Nepheline	CaO	> 0.1	Unknown	WSRC-STI-2007-00659 [52]	Peer Review

Property	Component	Range	QA Level	Reference	QA Method
Nepheline	CaO	> 0.1	GLP	PNNL-17950 [28]	Peer Review
Nepheline	CaO	> 0.1	NQA-1	Li et al.1996 [97], [129]	QA equivalent
Nepheline	CaO	> 0.1	NQA-1	PNNL-26630 [25]	QA equivalent
Nepheline	CaO	> 0.1	NQA-1	PNNL-28838 [21]	QA equivalent
Nepheline	CaO	> 0.1	NQA-1	PNNL-29847 [46]	QA equivalent
Nepheline	Cl	> 0	NQA-1	VSL-01R3560-2 [17]	QA equivalent
Nepheline	Cl	> 0	NQA-1	VSL-05R5460-1 [59]	QA equivalent
Nepheline	Cl	> 0	NQA-1	VSL-03R3460-2 [92]	QA equivalent
Nepheline	Cl	> 0	NQA-1	VSL-03R3470-1 [121]	QA equivalent
Nepheline	Cl	> 0	NQA-1	VSL-06R6480-2 [130] and VSL-08R1410-1 [86]	QA equivalent
Nepheline	Cl	> 0	NQA-1	VSL-04R4480-1 [16] and VSL-08R1410-1 [86]	QA equivalent
Nepheline	F	> 0.008	GLP	PNNL-20774 [117]	Peer Review
Nepheline	F	> 0.008	GLP	PNNL-17950 [28]	Peer Review
Nepheline	Li ₂ O	< 0.005	NQA-1	PNNL-26057 [41]	QA equivalent
Nepheline	Li ₂ O	< 0.005	NQA-1	PNNL-27138 [6]	QA equivalent
Nepheline	Li ₂ O	< 0.005	NQA-1	PNNL-20184 [19]	QA equivalent
Nepheline	Li ₂ O	< 0.005	GLP	PNNL-10987 [49]	Peer Review
Nepheline	Li ₂ O	< 0.005	GLP	WSRC-TR-2002-00426 [131]	Peer Review
Nepheline	SiO ₂	< 0.323	NQA-1	VSL-08R1360-1 [101]	QA equivalent
Nepheline	SiO ₂	< 0.323	GLP	Marra et al., WM2010 [109]	Peer Review
Nepheline	SiO ₂	< 0.323	GLP	McClane et al. 2020. [132]	QA equivalent
Viscosity	V ₂ O ₅	> 0.0	GLP	PNNL-14050 [80]	Peer Review
Viscosity	V ₂ O ₅	> 0.0	Unknown	40110-WP-0002 [133]	Data corroboration
Viscosity	B ₂ O ₃	< 0.04	GLP	PNNL-10987 [49]	Peer Review
Viscosity	B ₂ O ₃	< 0.04	GLP	Bulkley and Vienna 1997 [40]	Peer Review
Viscosity	B ₂ O ₃	< 0.04	Unknown	Matlack et al. 1997 [119], Muller and Pegg 1998 [134]	Peer Review
Viscosity	B ₂ O ₃	< 0.04	GLP	PNNL-10918 [22]	Peer Review
Viscosity	F	> 0.002	GLP	INEEL-EXT-99-01322 [29]	Peer Review
Viscosity	F	> 0.002	Unknown	Bates 1985 [135]	Peer Review

Property	Component	Range	QA Level	Reference	QA Method
Viscosity	F	> 0.002	GLP	PNNL-17950 [28]	Peer Review
EC	B ₂ O ₃	> 0.15	GLP	Marra et al., WM2010 [109]	Peer Review
EC	Al ₂ O ₃	> 0.08	GLP	PNNL-20774 [117]	Peer Review
Sp950	MgO	> 0.024	GLP	Mika et al. 1997 [48]	Peer Review
Sp950	MgO	> 0.024	GLP	Marra et al., WM2010 [109]	Peer Review
Sp950	MgO	> 0.024	GLP	Vienna 2002 dissertation [37]	Peer Review
Sp950	Na ₂ O	> 0.24	GLP	Jiricka et al. 2003 [68]	Peer Review
Sp950	Na ₂ O	> 0.24	GLP	PNNL-22631 [136]	Peer Review
T _g	SiO ₂	< 0.42	GLP	Deshkar et al. 2022 [137]	Data corroboration
Density	SiO ₂	< 0.3	GLP	Deshkar et al. 2022 [137]	Data corroboration
Density	CaO	> 0.07	GLP	INEEL-EXT-99-01322 [29]	Peer Review
Density	CaO	> 0.07	Unknown	Matlack et al. 1997 [119], Muller and Pegg 1998 [134]	Peer Review
Density	CaO	> 0.07	Unknown	40110-WP-0002 [133]	Confirmatory test
T _L	MgO	> 0.025	GLP	PNNL-12208 [76]	Peer Review
T _L	MgO	> 0.025	GLP	Vienna 2002 dissertation [37]	Peer Review
T _L	MgO	> 0.025	Unknown	Matlack et al. 1997 [119], Muller and Pegg 1998 [134]	Peer Review
T _L	MgO	> 0.025	Unknown	40110-WP-0002 [133]	Peer Review
T _L	Cr ₂ O ₃	> 0.0058	GLP	PNNL-10987 [49]	Peer Review
T _L	Cr ₂ O ₃	> 0.0058	Unknown	Fu and Pegg 1998 [34]	Peer Review
T _L	Cr ₂ O ₃	> 0.0058	GLP	PNNL-17950 [28]	Peer Review
T _L	P ₂ O ₅	> 0.03	GLP	INEEL-EXT-98-00970 [44]	Peer Review
T _L	P ₂ O ₅	> 0.03	Unknown	Crum et al. 1997 [43]	Peer Review
T _L	P ₂ O ₅	> 0.03	GLP	Reynolds and Hrma 1996 [138]	Peer Review

4.0 Summary

In this work, glass property-composition data from various institutions (e.g., PNNL, SRNL, INL, and CUA) were compiled into a single database in an Excel spreadsheet format with 17,006 rows and 305 columns. In several cases, the same glasses with different compositional views (e.g., target or analyzed) and heat treatments (e.g., quenched or CCC) were compiled separately as different rows. In the database, there are 6718 quenched glasses with the target composition. The original reports and papers cited in the database were sourced if available, and QA information from each study was consolidated into the database. This project aimed to compile these data in a single database useful to the waste vitrification facilities: Hanford WTP, the Defense Waste Processing Facility at the Savannah River Site, and potentially the Idaho Calcine Disposition Project. This compiled database will improve the accuracy/precision of glass property models predictions, which will likely lead to higher waste loading, higher waste throughput, more flexibility in flowsheet options, and reduced mission life.

A plan was developed to support future qualification of data important to Hanford DFHLW processing. To determine the potentially important compositional regions, glasses were formulated for estimates of Hanford DFHLW streams. Violin and heatmap plots were used to identify compositional gaps by comparing the database to the target DFHLW glasses for each property. Eleven properties were examined: density, electrical conductivity, viscosity, PCT, TCLP, K-3 refractory corrosion, nepheline formation, spinel formation, SO₃ solubility, liquidus temperature, and glass transition temperature. Composition gaps were used to identify studies with insufficient-QA data that would significantly improve Hanford DFHLW processing if they were qualified for use.

There are ~80 studies among the 11 properties that contain insufficient-QA data of interest for Hanford DFHLW treatment, where a data qualification plan was drafted with a priority order of properties. In the plan, methods including QA equivalence, peer review, data corroboration, and confirmatory testing have been assigned to each study for future qualification efforts.

Future proposed work includes the following:

- Making minor improvements (e.g., reference format, combining the quenched and CCC data, checking duplicated glasses/measurements) and performing maintenance on the database. SRNL is developing a web tool to host and maintain this database. The online database will provide a universal resource to labs, sites, industry, and academia.
- Performing QA to check and qualify the database (i.e., independent technical review of the compiled single database).
- Qualifying the insufficient-QA data where possible for Hanford DFHLW treatment.
- Re-evaluating target composition region for the coverage for Hanford DFHLW treatment.
- Generating new data where gaps exist for Hanford DFHLW treatment.
- Improving glass property models using the database and revised glass formulation algorithm for Hanford DFHLW treatment.

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Appendix A – Violin Plots

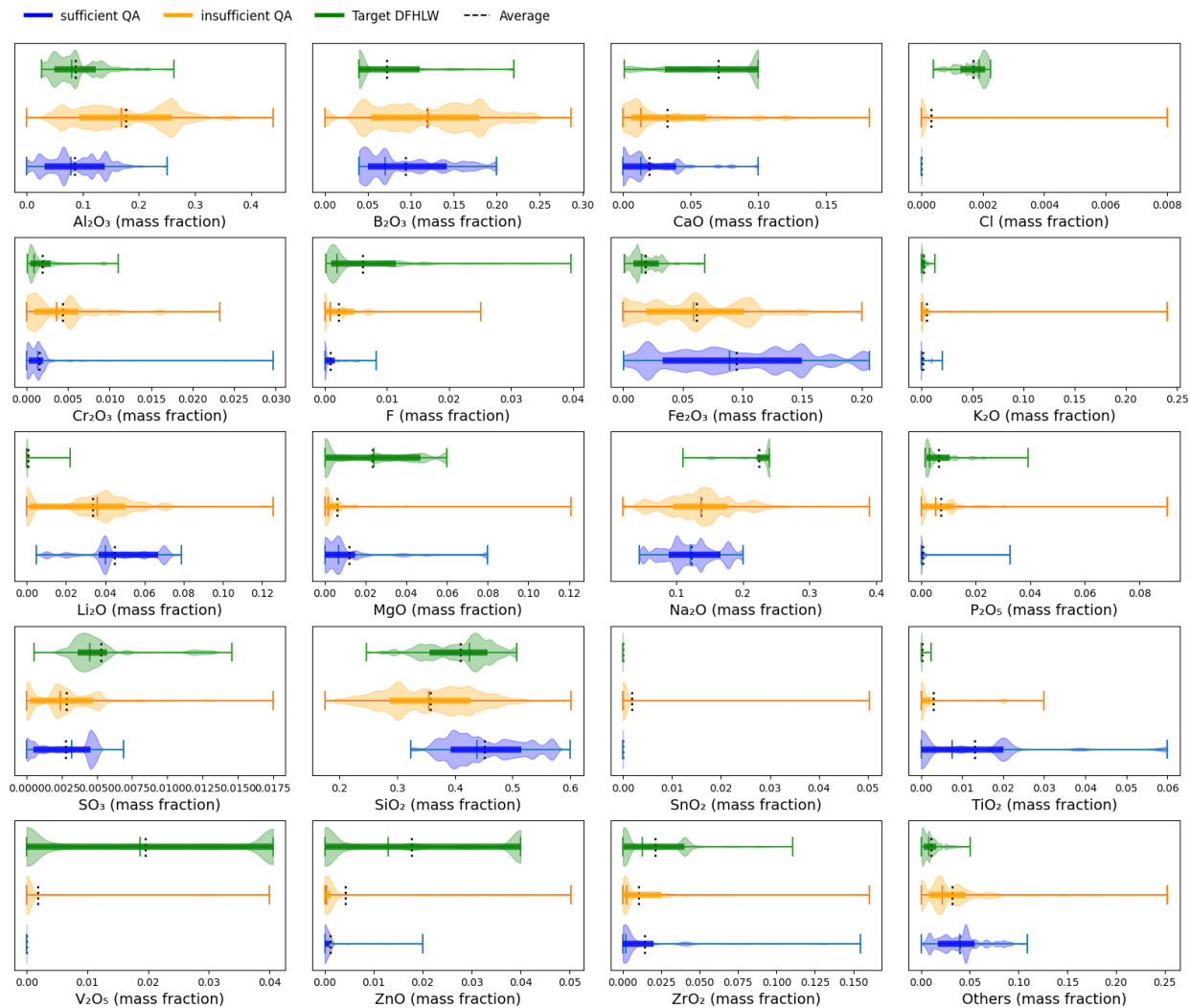


Figure A.1. Violin plots of nepheline data in the database (sufficient-QA and insufficient-QA data), along with target DFHLW glasses.

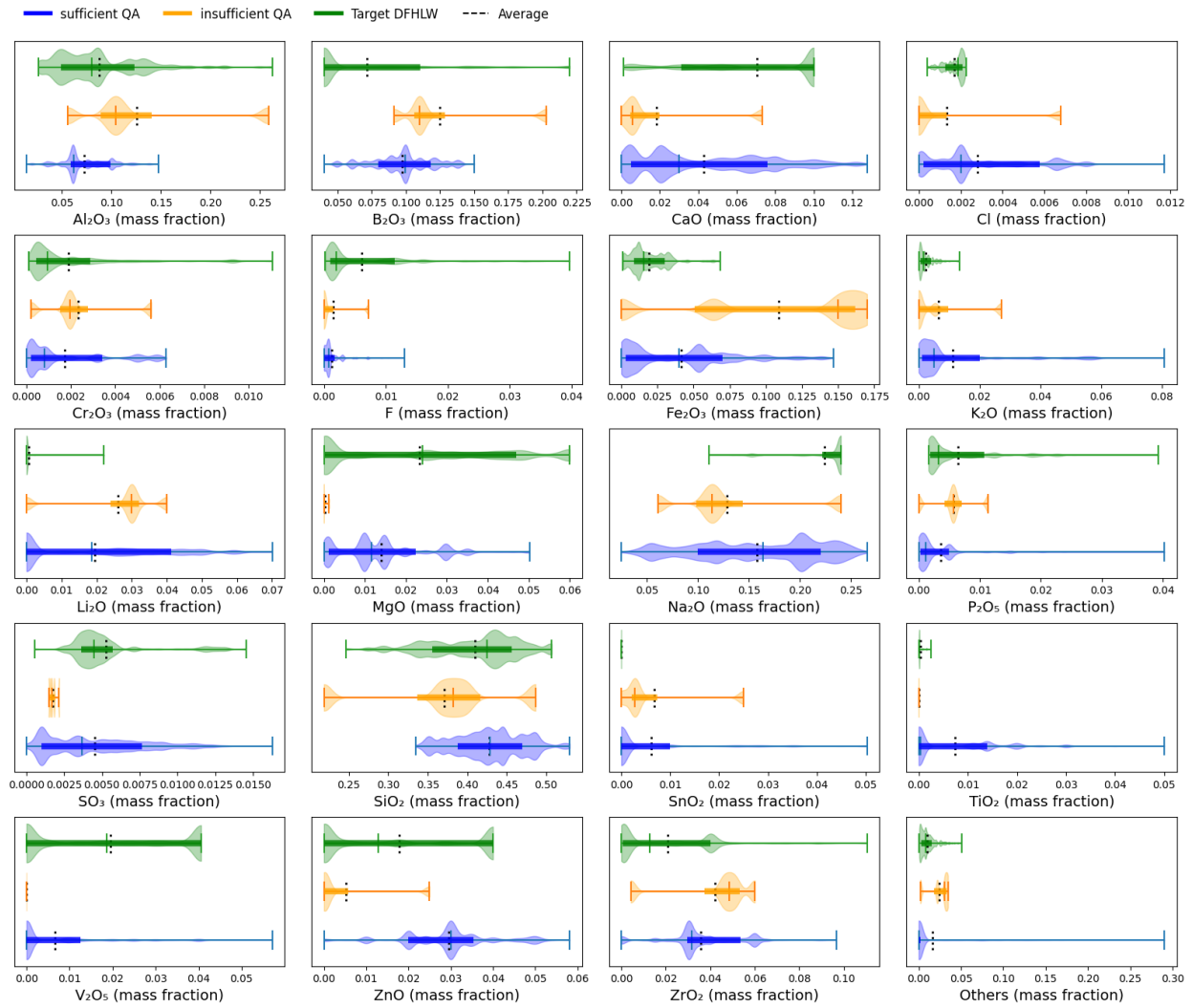


Figure A.2. Violin plots of EC data in the database (sufficient-QA and insufficient-QA data), along with target DFHLW glasses.

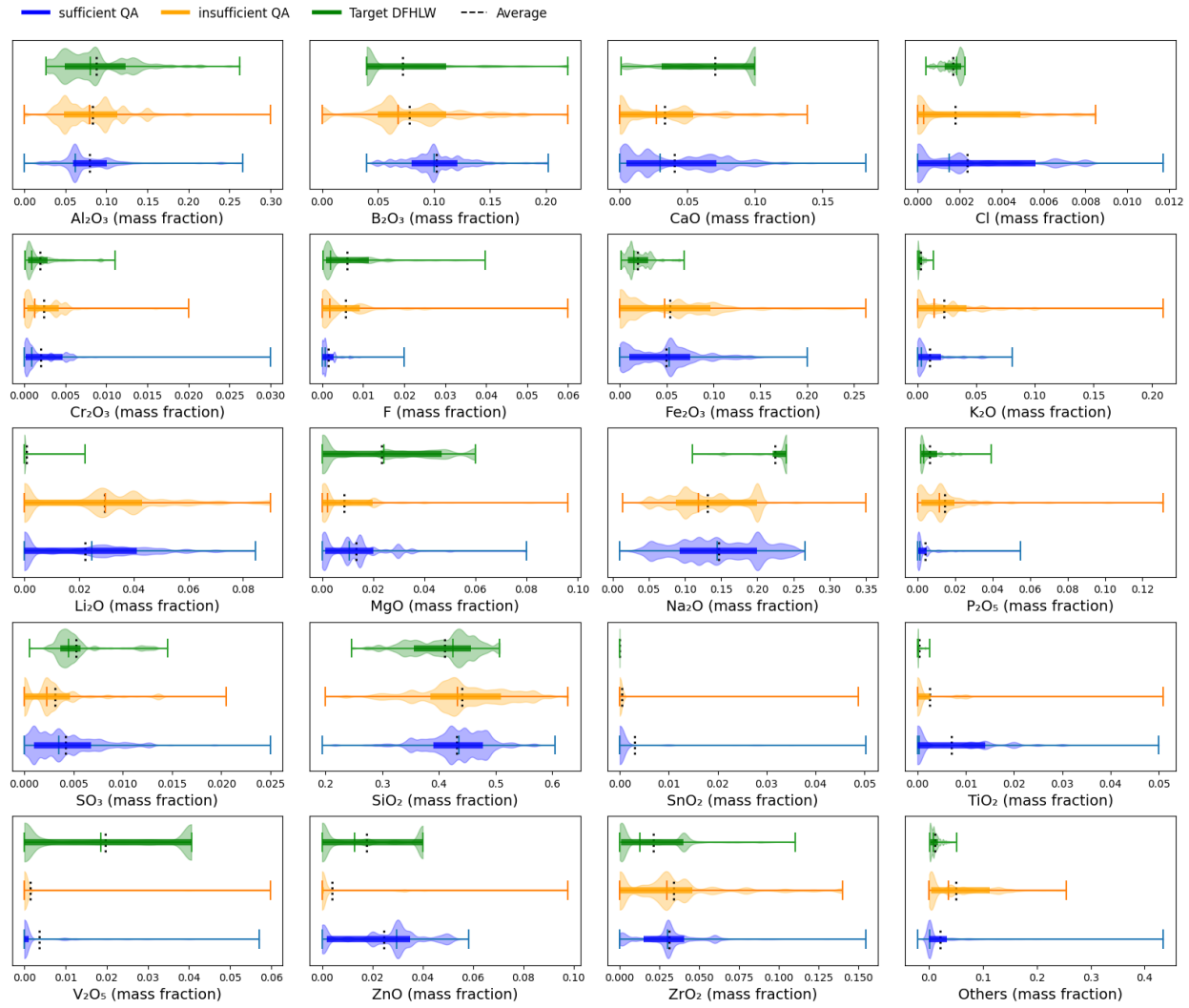


Figure A.3. Violin plots of viscosity data in the database (sufficient-QA and insufficient-QA data), along with target DFHLW glasses.

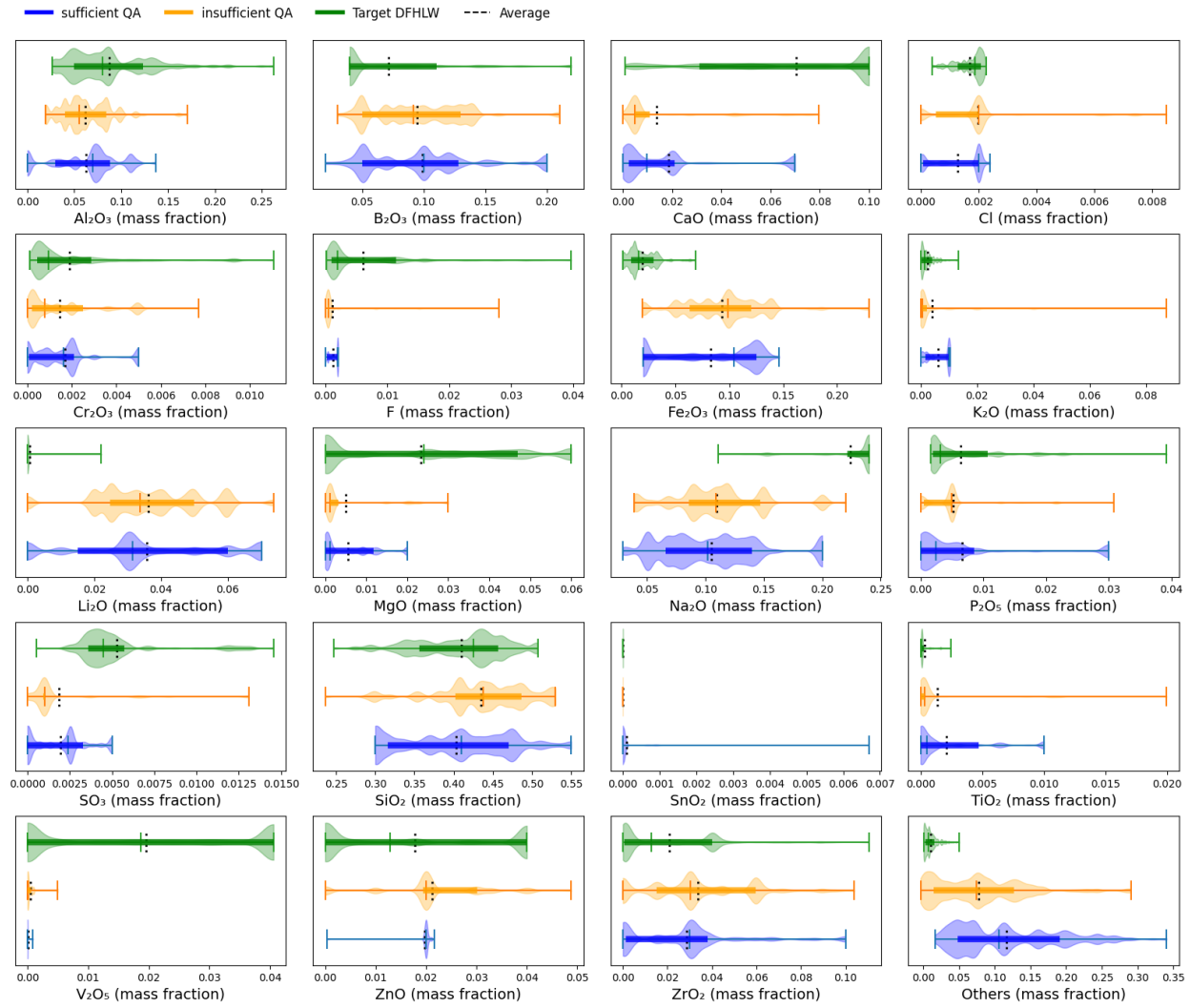


Figure A.4. Violin plots of TCLP Cd data in the database (sufficient-QA and insufficient-QA data), along with target DFHLW glasses.

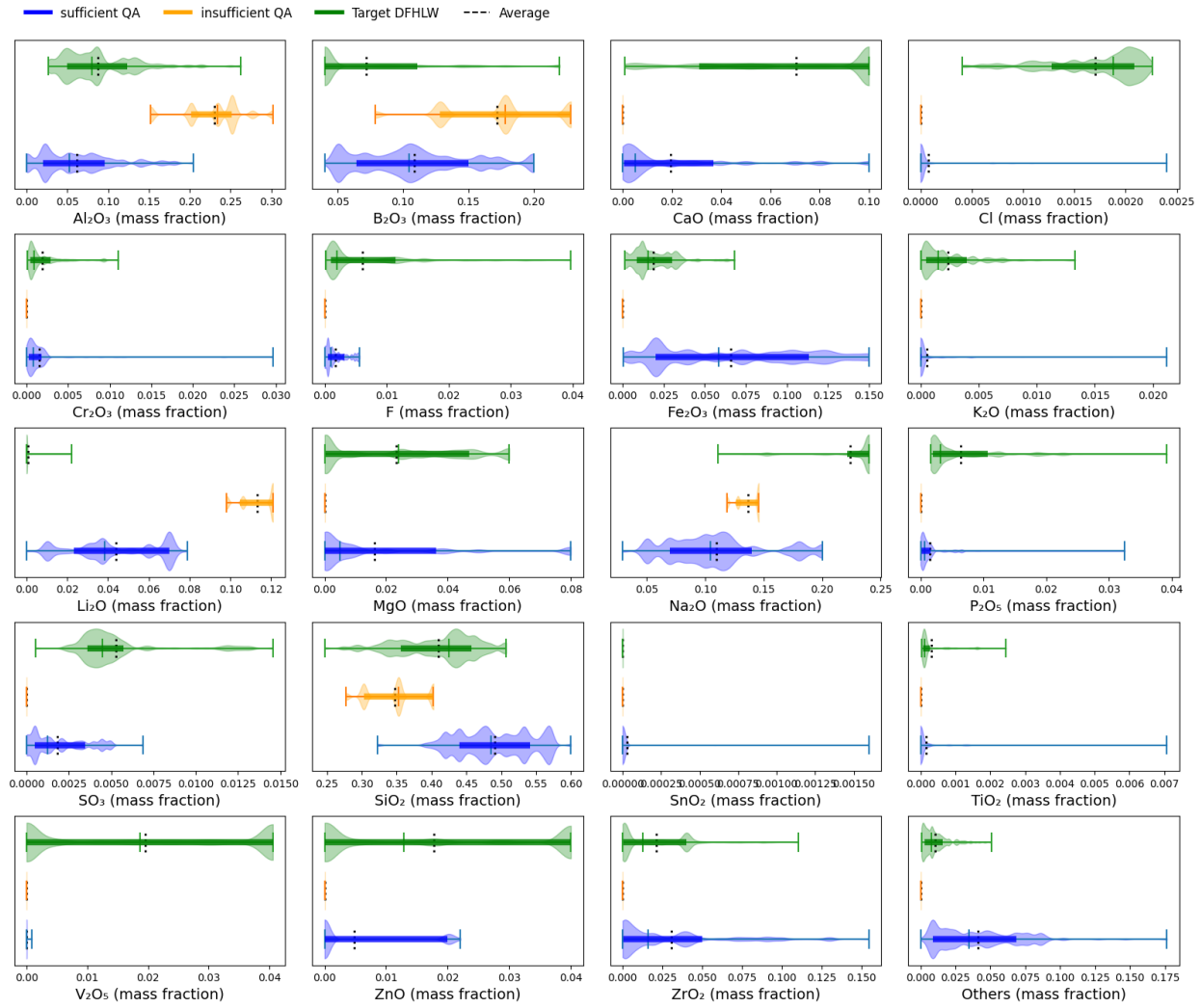


Figure A.5. Violin plots of T_g data in the database (sufficient-QA and insufficient-QA data), along with target DFHLW glasses.

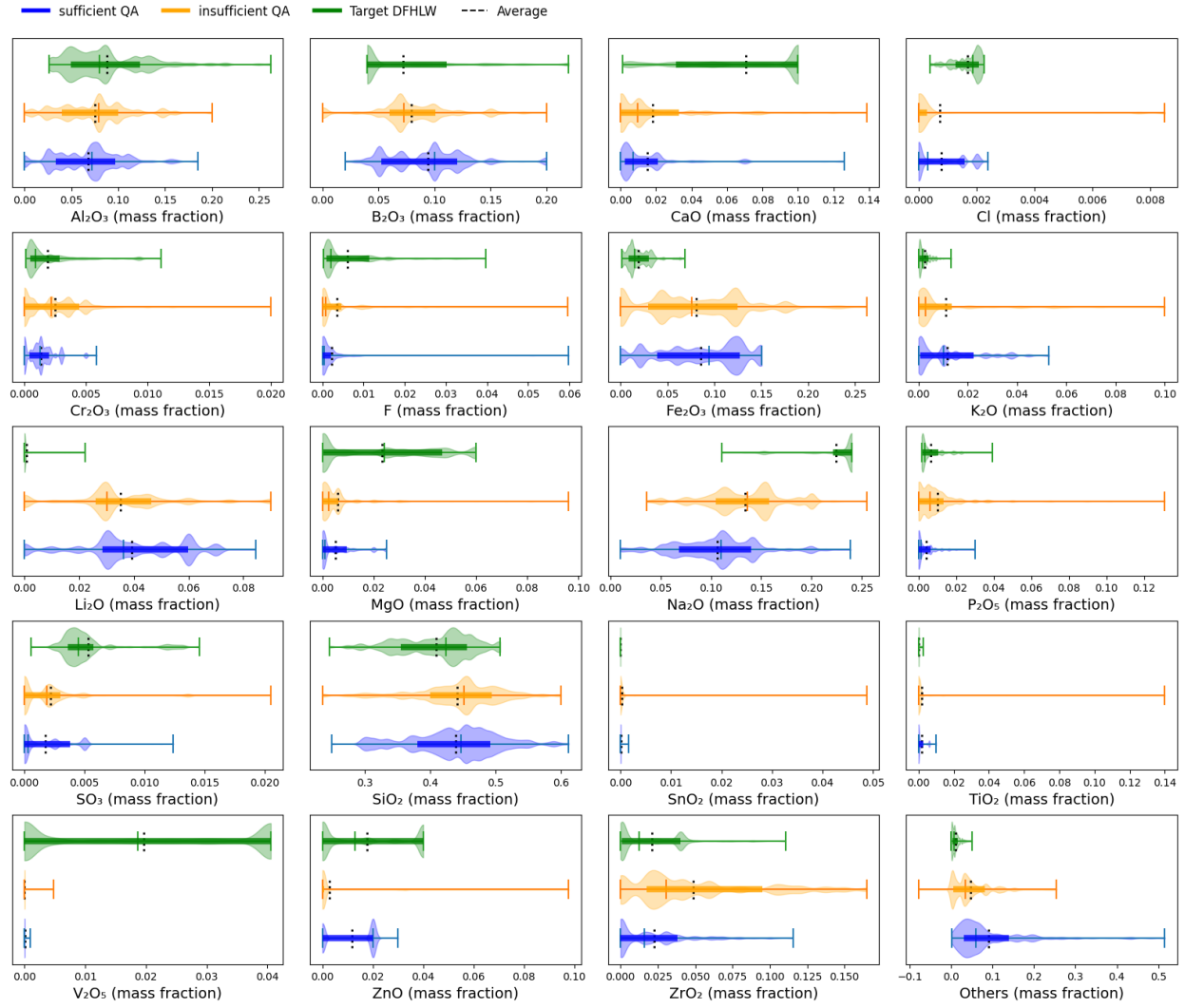


Figure A.6. Violin plots of T_L data in the database (sufficient-QA and insufficient-QA data), along with target DFHLW glasses.

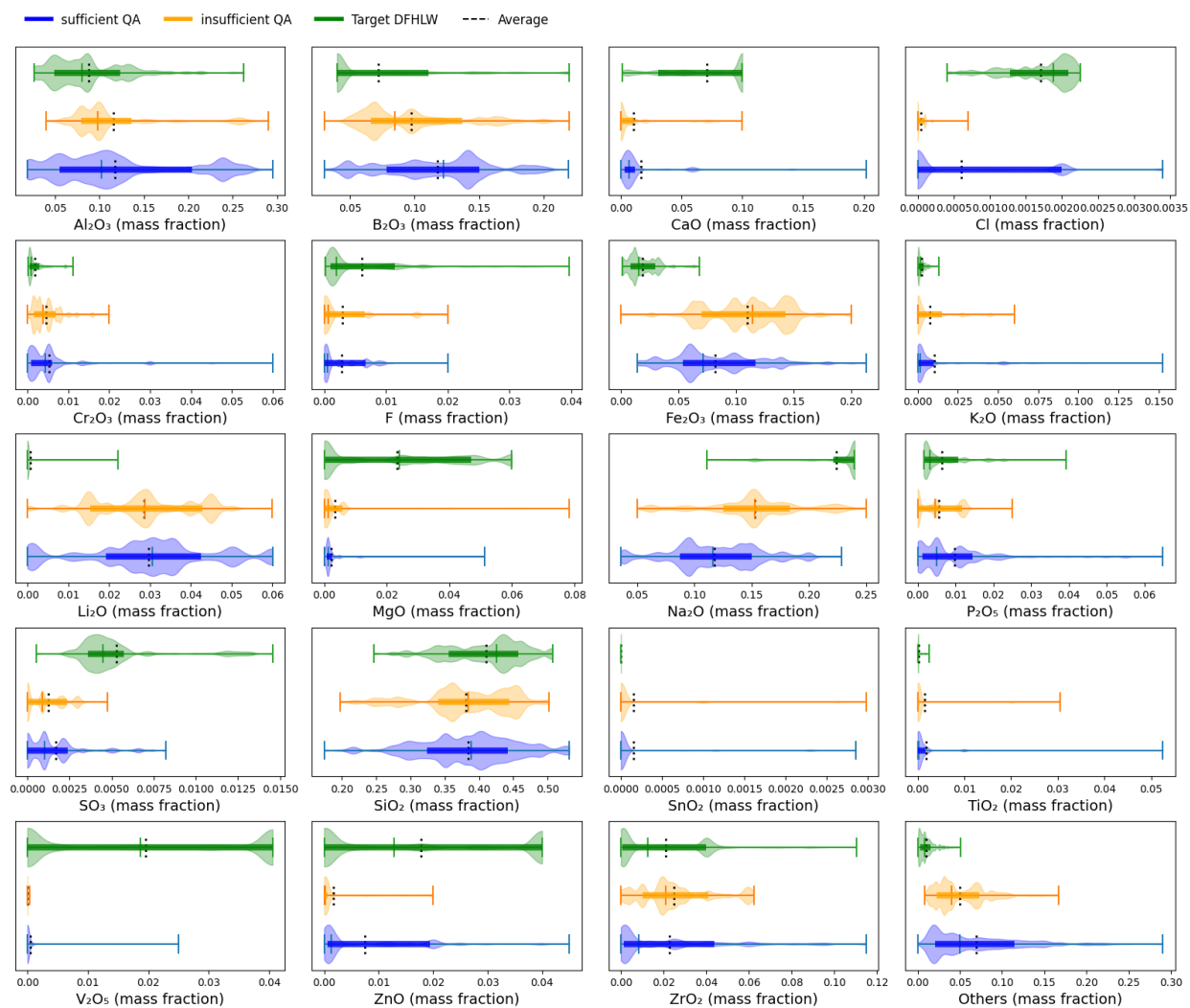


Figure A.7. Violin plots of Sp950 data in the database (sufficient-QA and insufficient-QA data), along with target DFHLW glasses.

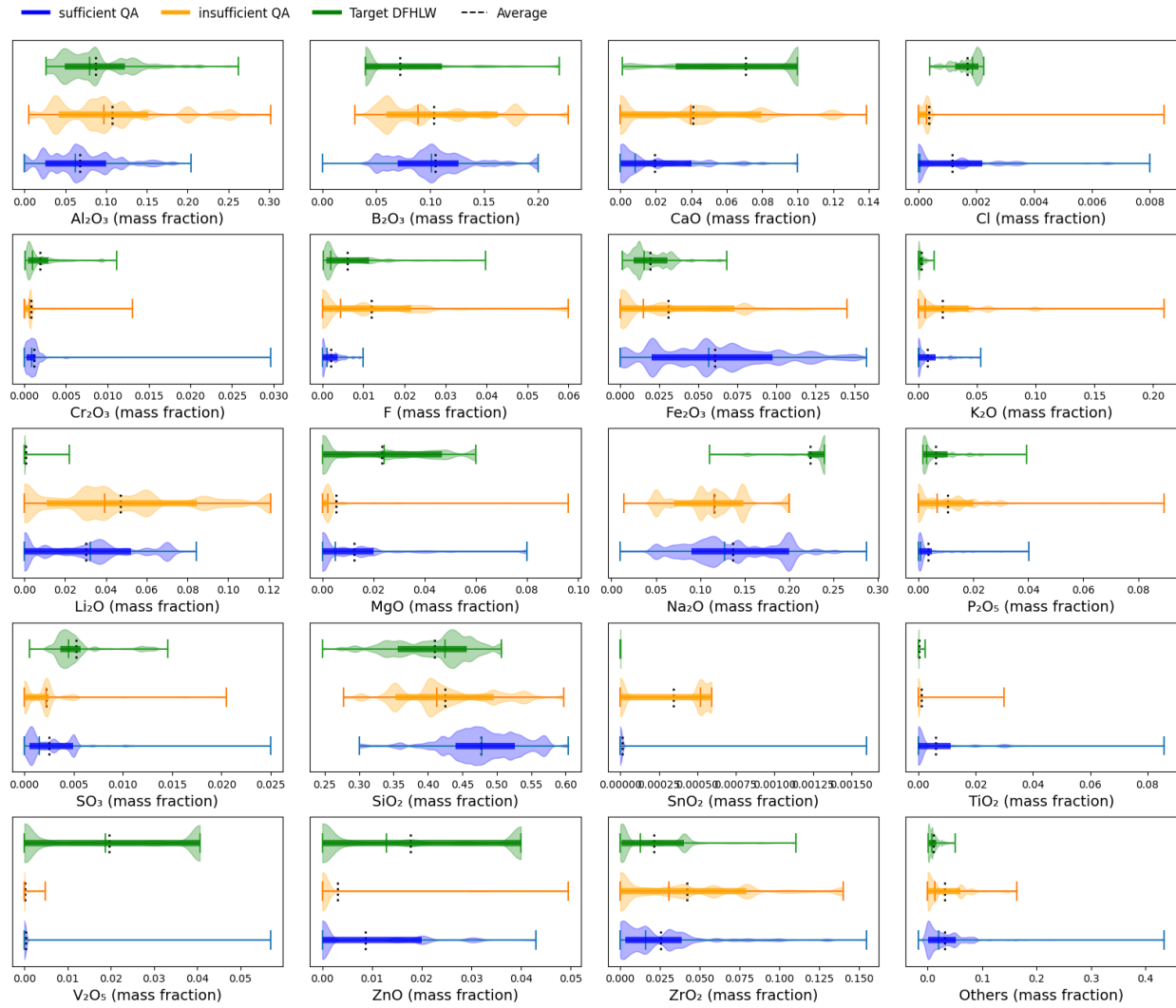


Figure A.8. Violin plots of density data in the database (sufficient-QA and insufficient-QA data), along with target DFHLW glasses.

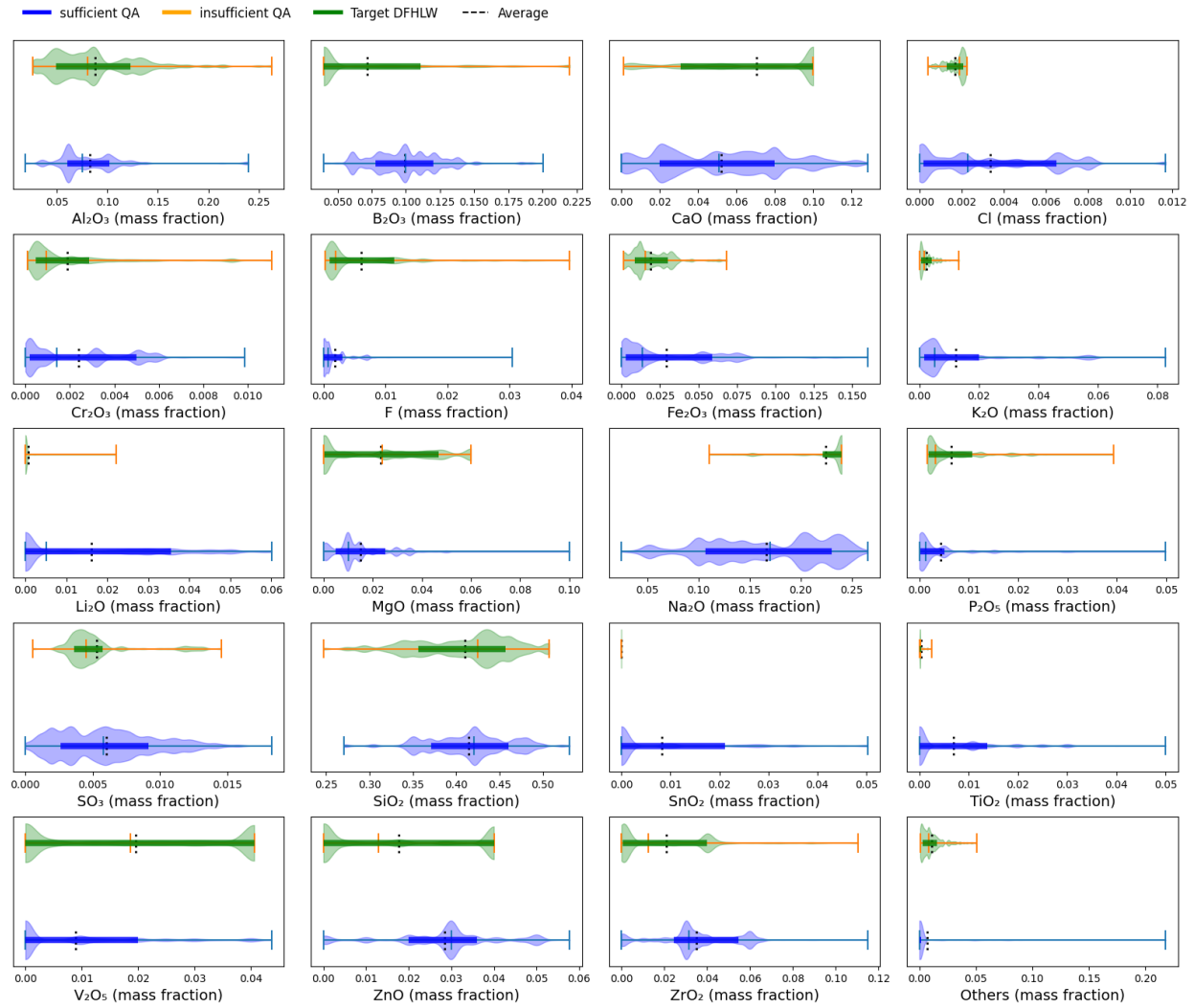


Figure A.9. Violin plots of SO_3 solubility data in the database (all data pass QA requirement), along with target DFHLW glasses. Since all the available data satisfy the QA requirement, there is no data in “insufficient QA”.

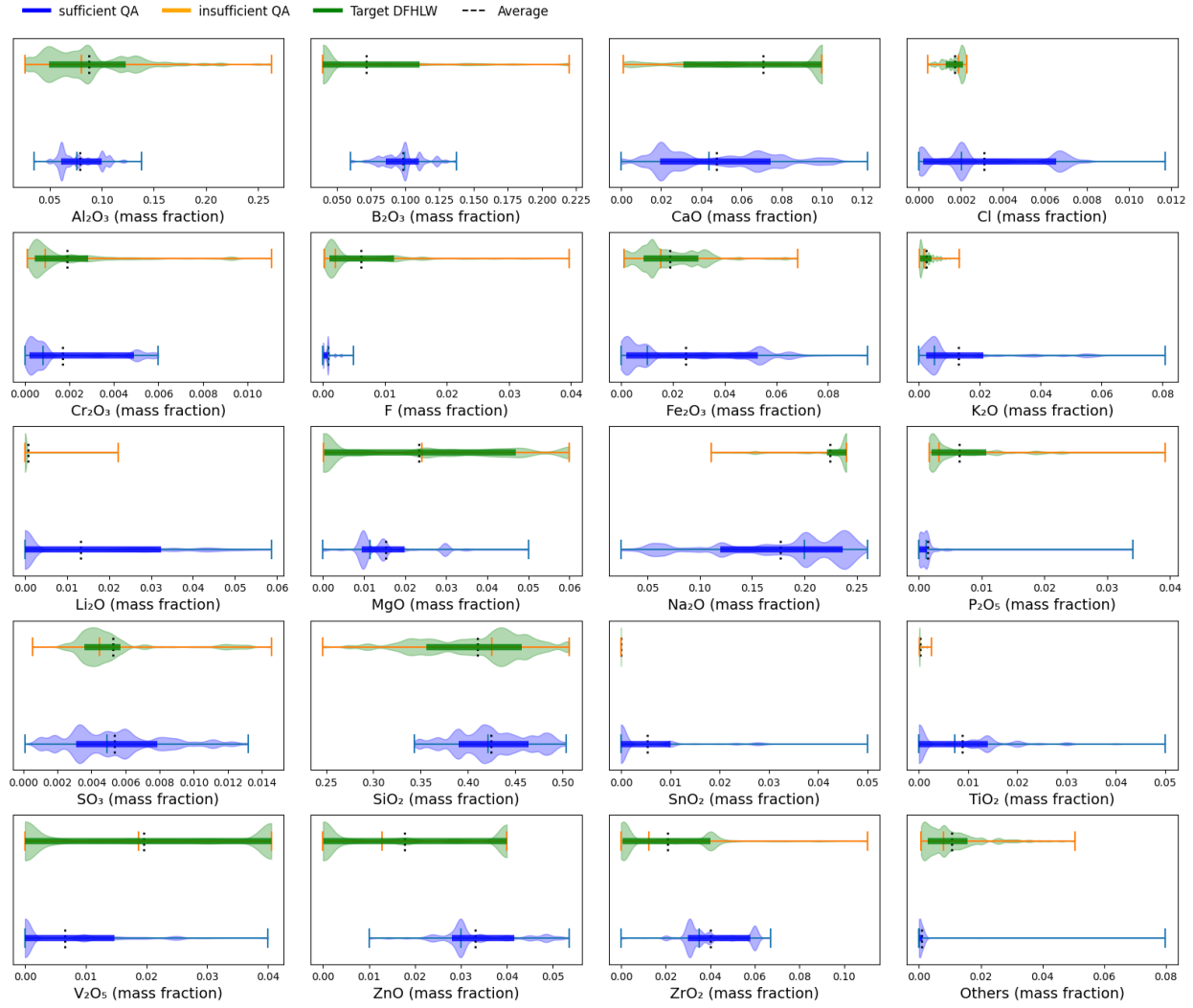


Figure A.10. Violin plots of K-3 refractory corrosion data in the database (all data pass QA requirement), along with target DFHLW glasses. Since all the available data satisfy the QA requirement, there is no data in “insufficient QA”.

Appendix B – Selected Heatmap with Scatter Plots

These figures were used to determine the gaps shown in Table 3.3.

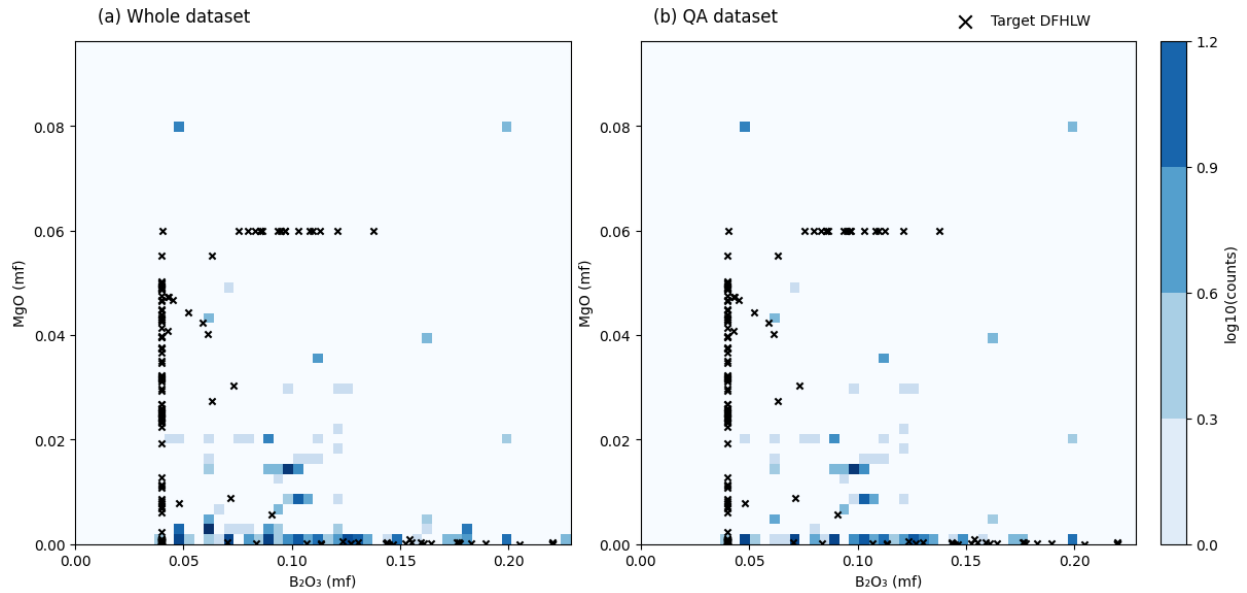


Figure B.1. MgO versus B_2O_3 of density data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

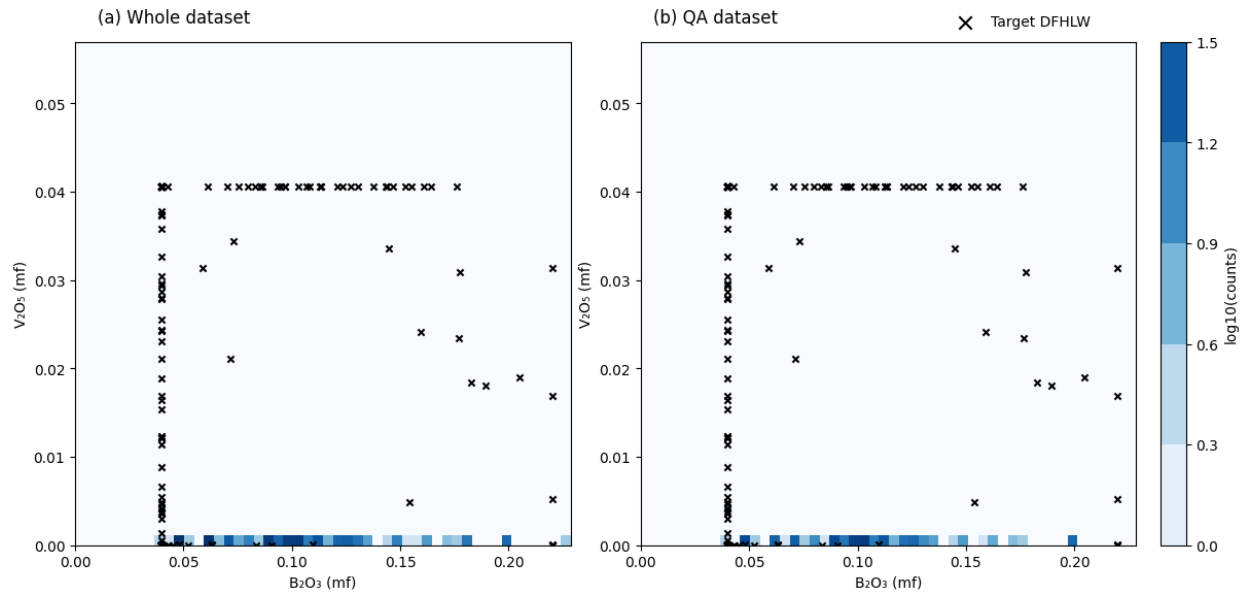


Figure B.2. V_2O_5 versus B_2O_3 of density data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

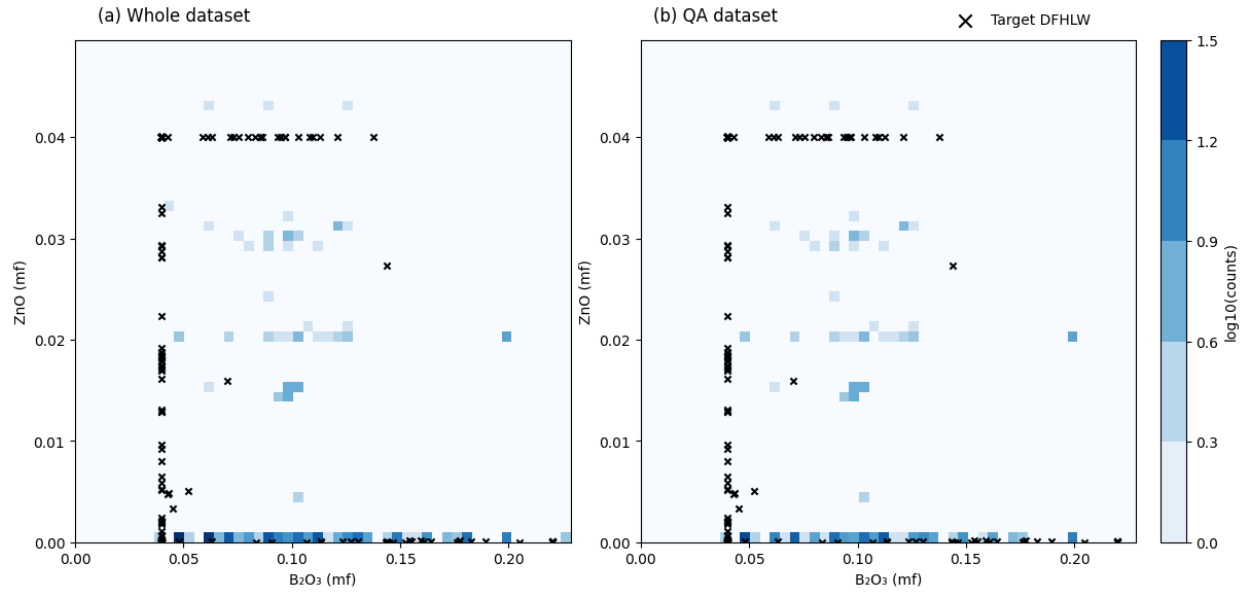


Figure B.3. ZnO versus B₂O₃ of density data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

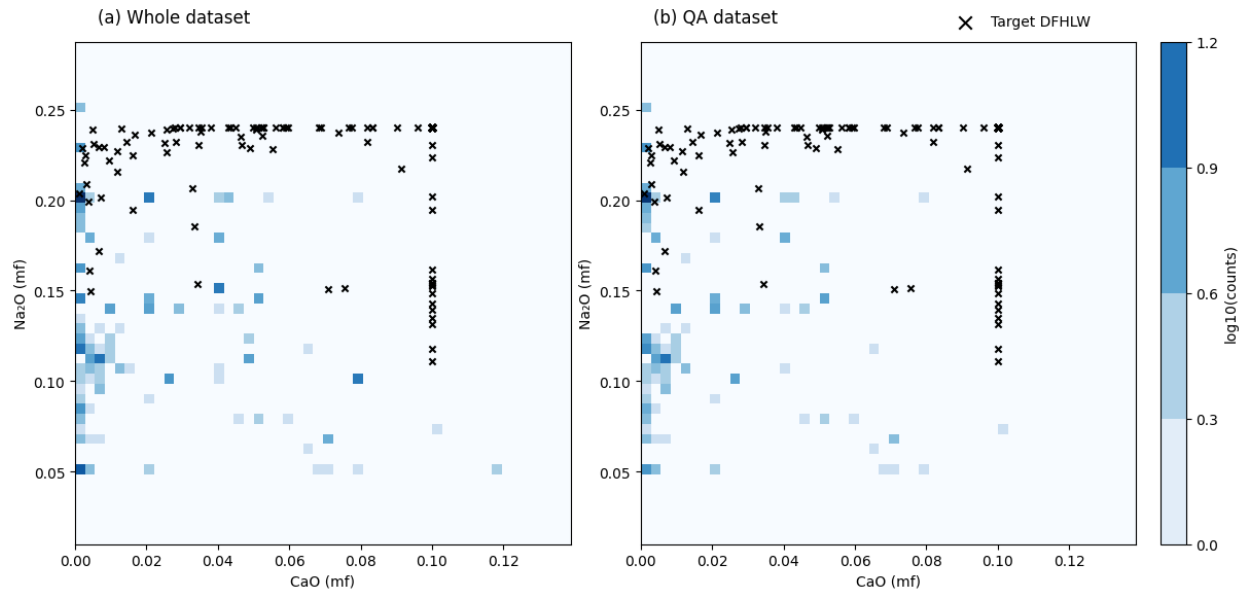


Figure B.4. Na₂O versus CaO of density data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

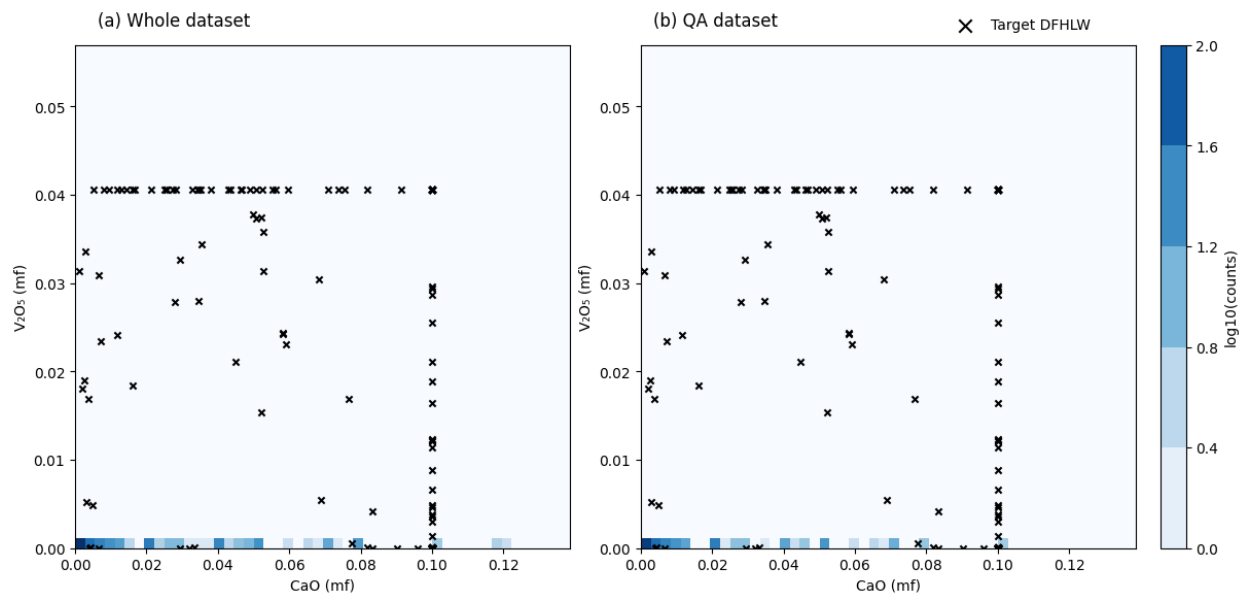


Figure B.5. V_2O_5 versus CaO of density data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

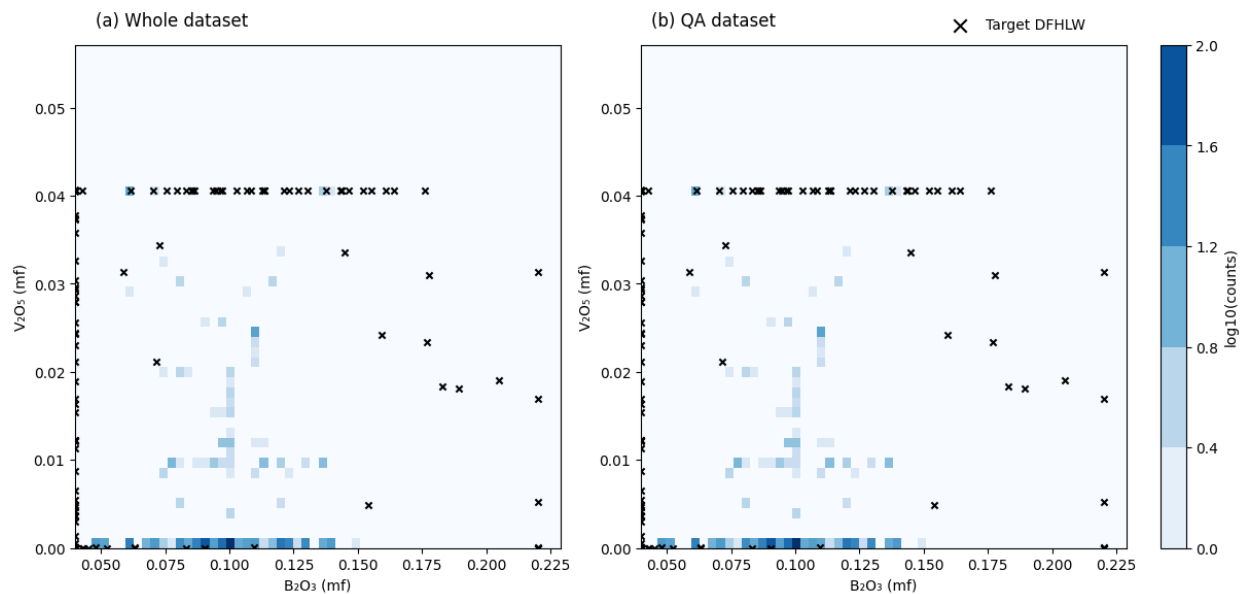


Figure B.6. V_2O_5 versus B_2O_3 of EC data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

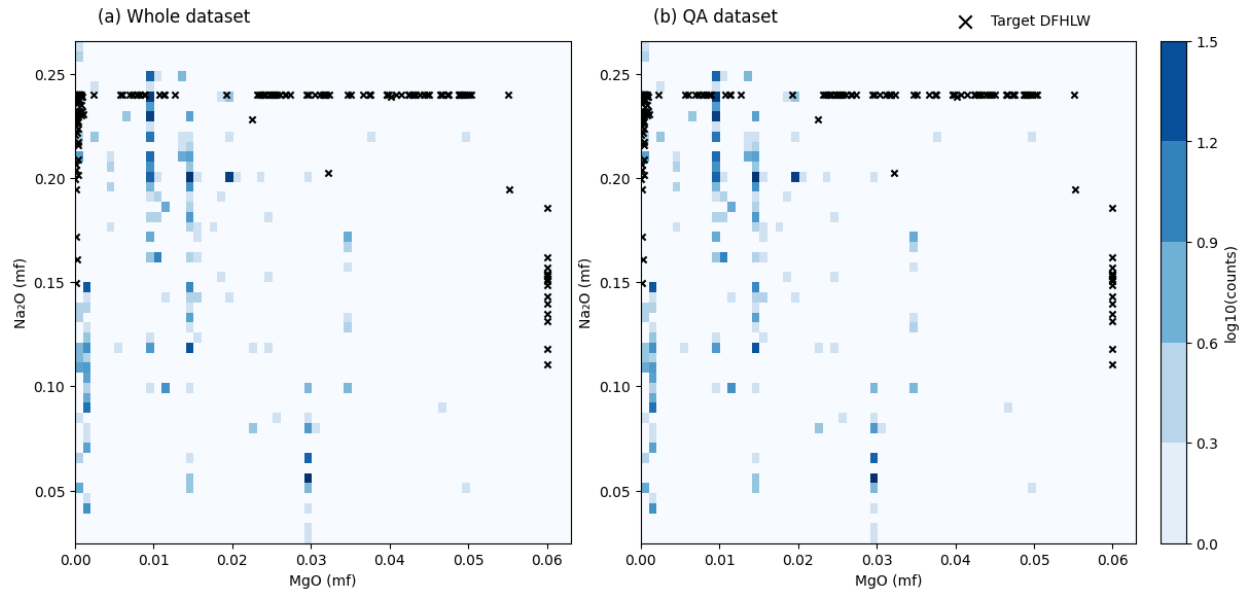


Figure B.7. Na_2O versus MgO of EC data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

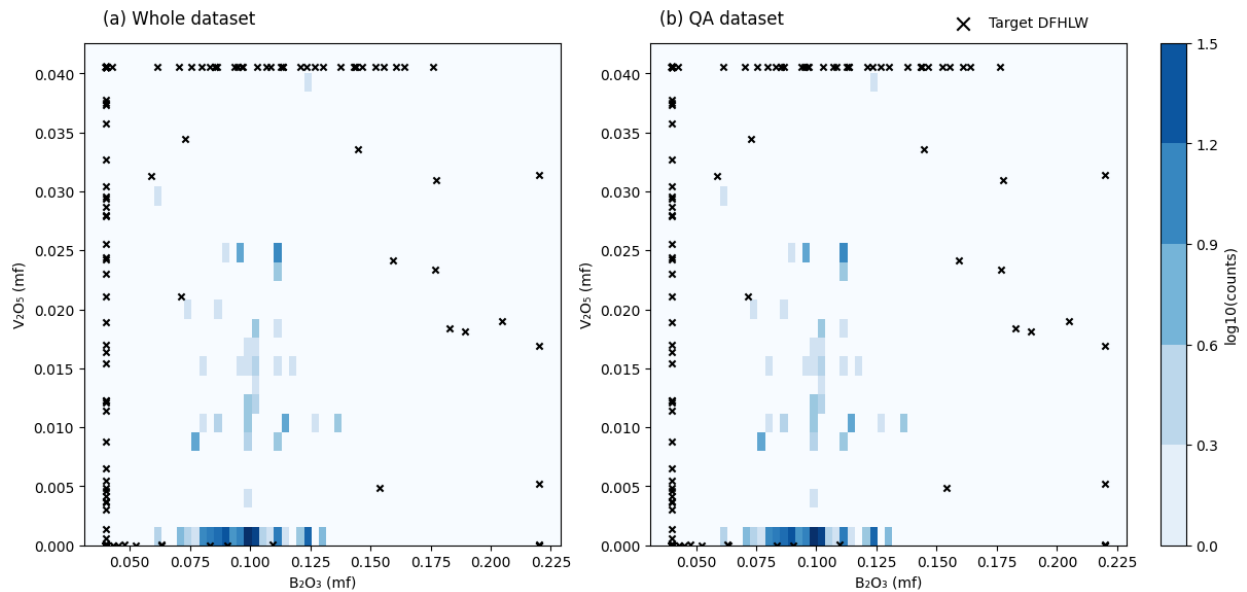


Figure B.8. V_2O_5 versus B_2O_3 of K-3 data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

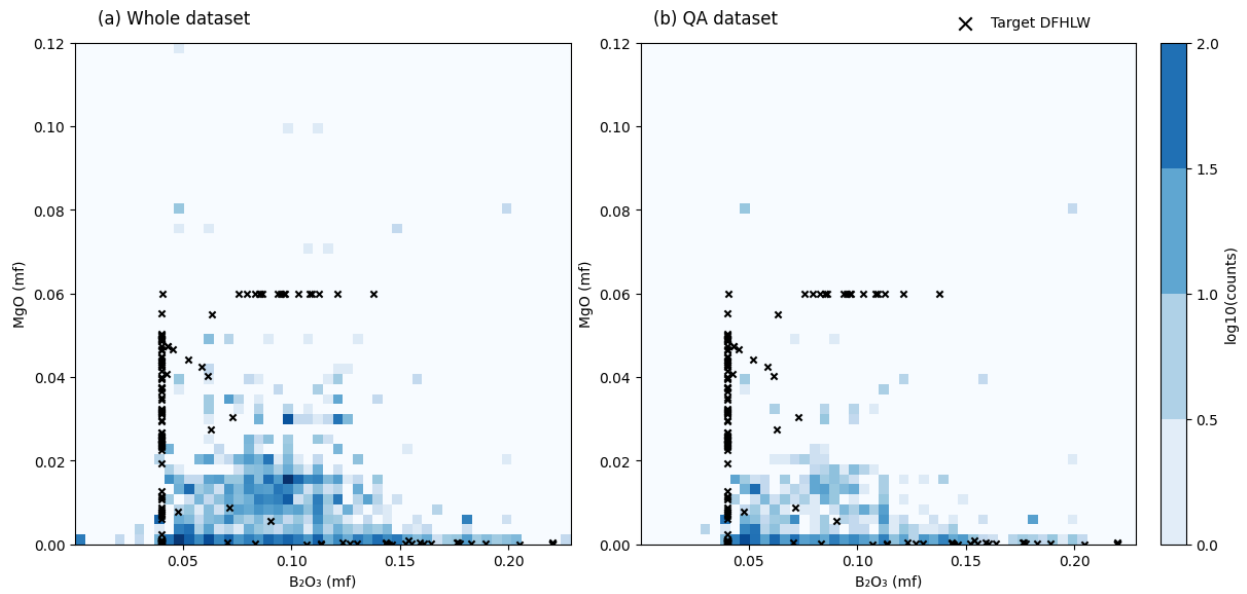


Figure B.9. MgO versus B_2O_3 of PCT data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

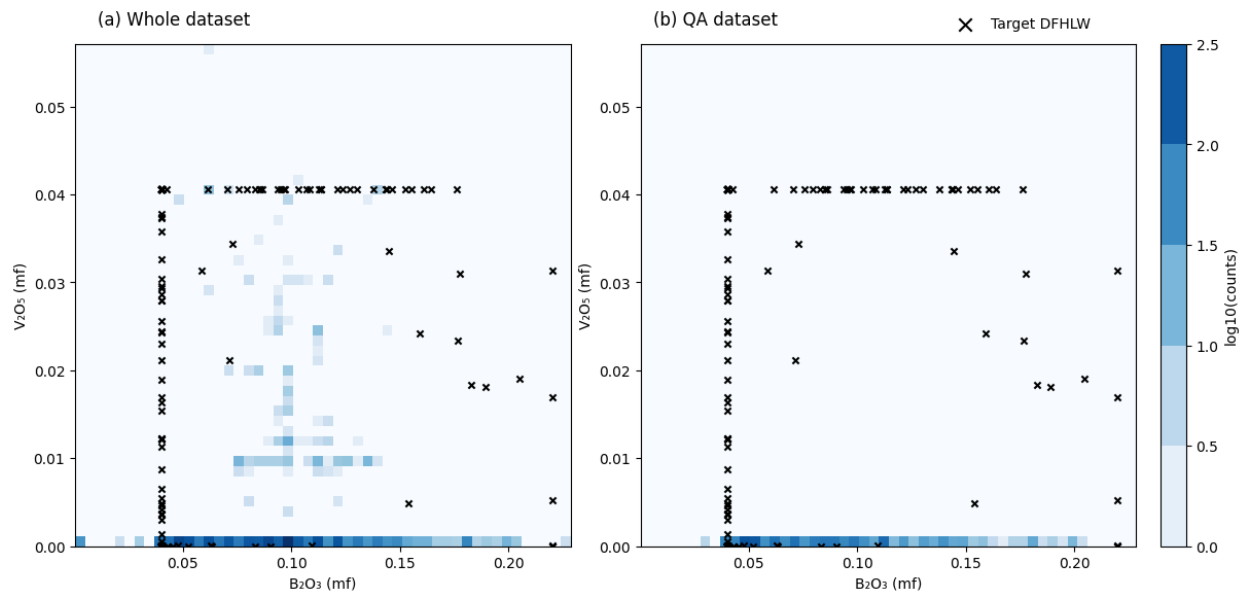


Figure B.10. V_2O_5 versus B_2O_3 of PCT data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

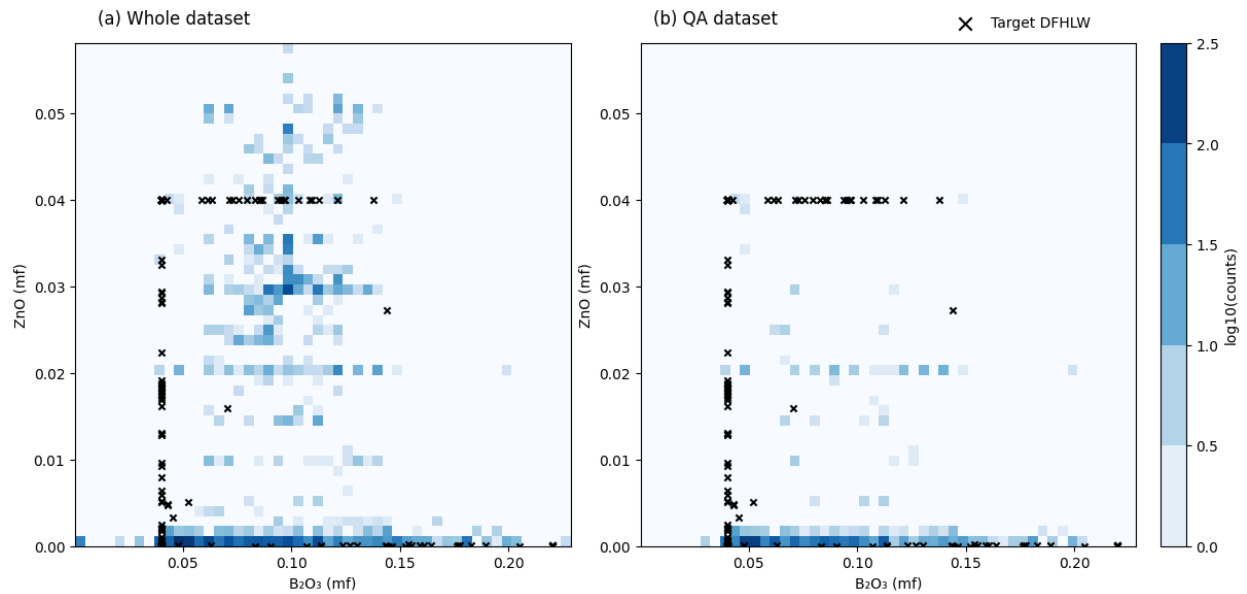


Figure B.11. ZnO versus B₂O₃ of PCT data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

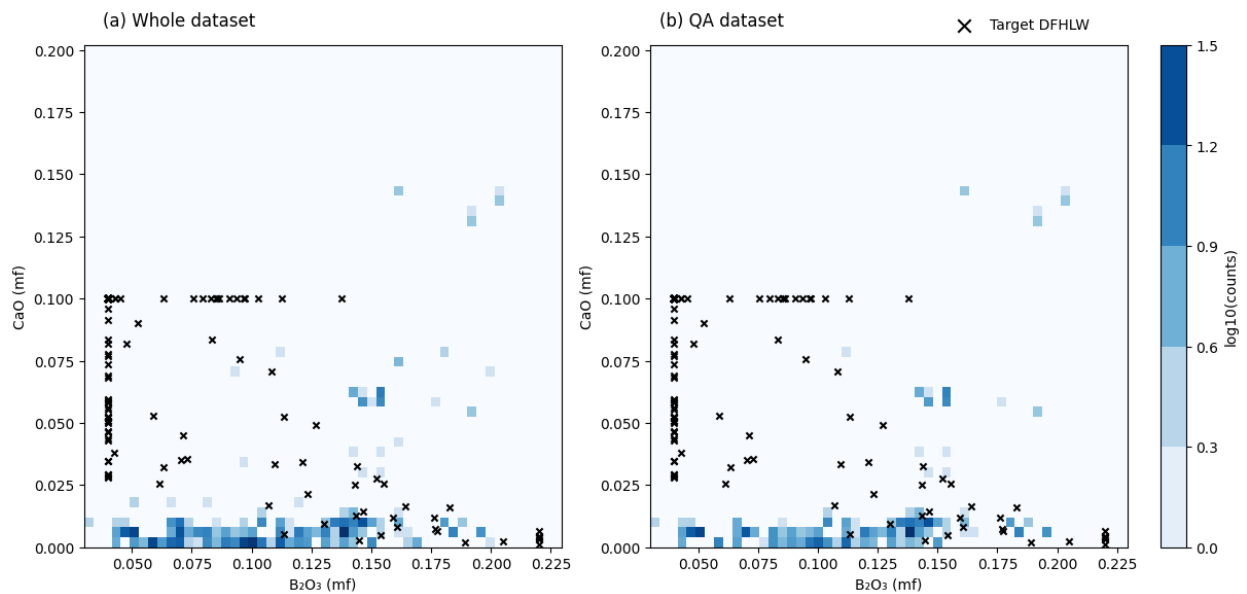


Figure B.12. CaO versus B₂O₃ of spinel formation data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

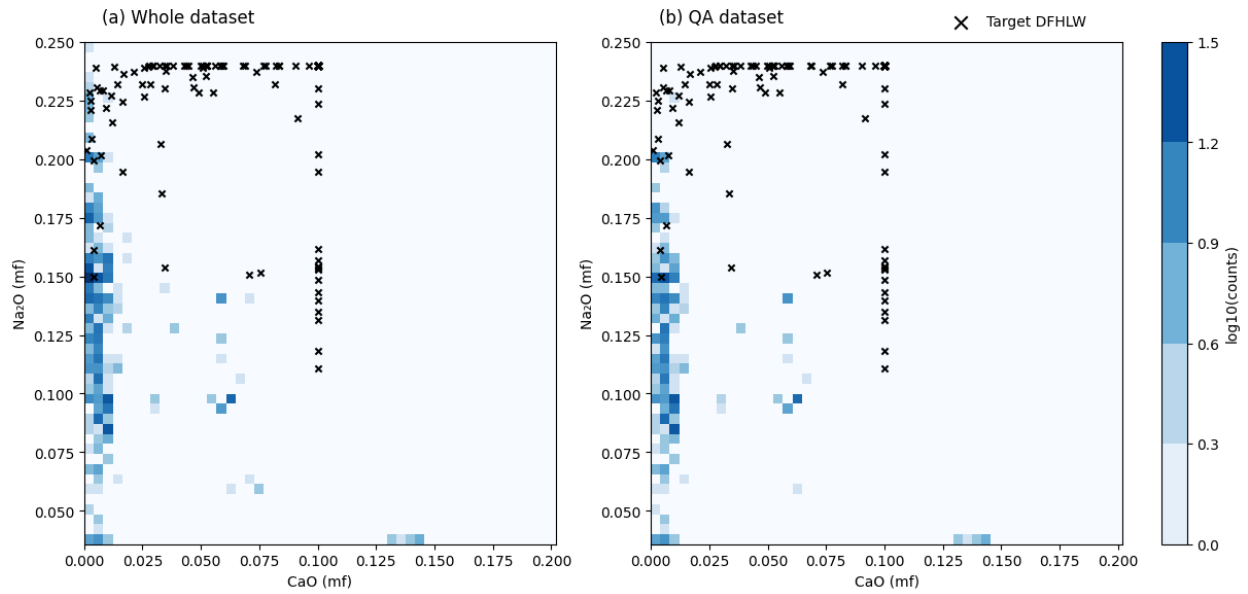


Figure B.13. Na_2O versus CaO of spinel formation data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

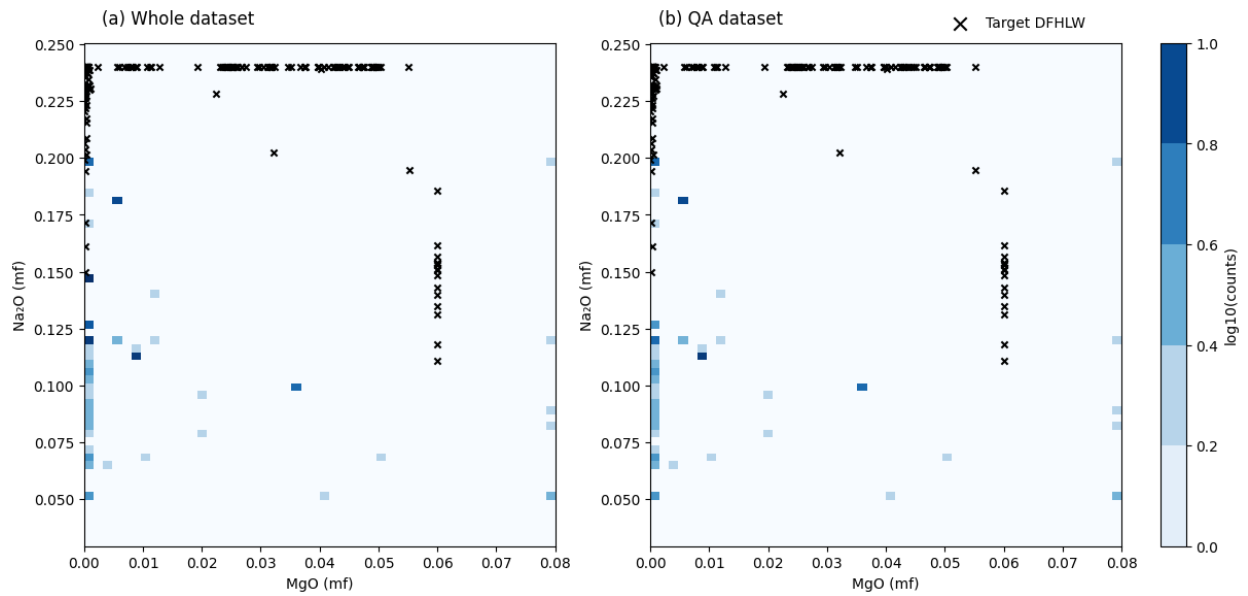


Figure B.14. Na_2O versus MgO of T_g data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

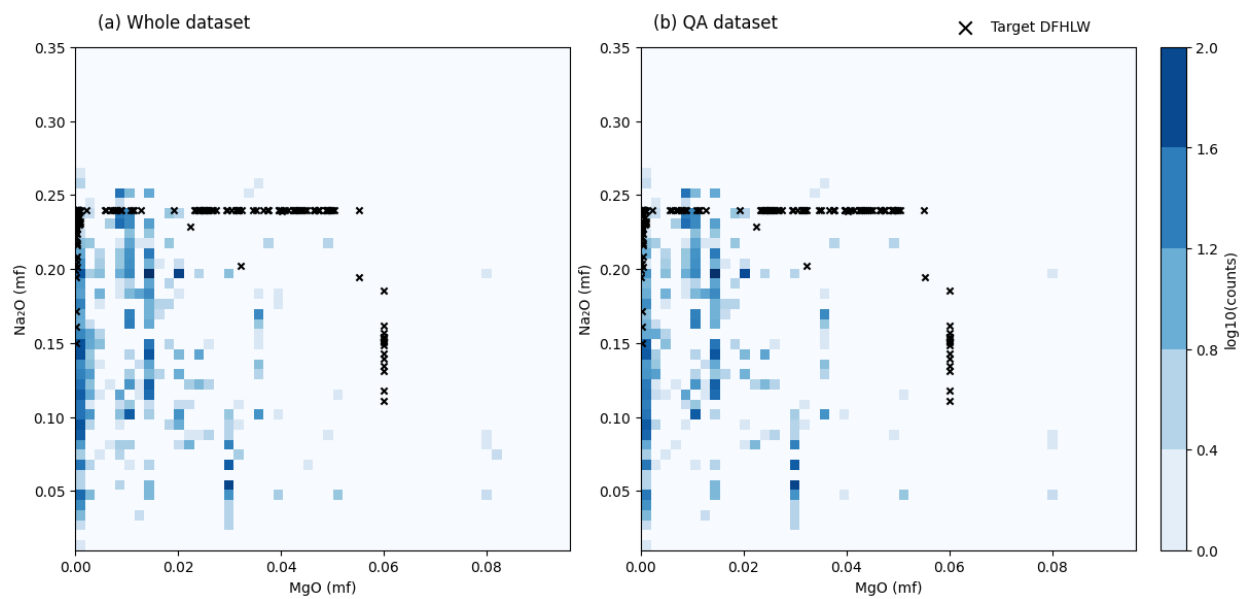


Figure B.15. Na_2O versus MgO of viscosity data in the database (heatmap) with target DFHLW glasses (scatter black crosses).

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