INTEROFFICE MEMORANDUM

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Normative Mineral Compositions of Saltstone Disposal Facility Cementitious Materials

The Saltstone Disposal Facility (SDF) relies on cementitious materials to limit radionuclide and chemical waste releases to the environment to levels that will meet federal and state regulatory requirements. The “saltstone” waste form is a grout composed of hydrated slag, fly ash, and optionally cement, and Saltstone Disposal Unit (SDU) containment barriers are composed mainly of concrete. The hydrated mineral composition of these materials affects various attributes of facility performance including:

- The bulk chemistry of pore solutions through time, which affects radionuclide and chemical species solubility and liquid-solid partitioning (K_d)
- Chemical degradation of concrete through sulfate attack and carbonation-controlled corrosion of embedded steel, leading to higher hydraulic conductivity and effective diffusion coefficient
- Decalcification of saltstone, also leading to higher hydraulic conductivity and effective diffusion coefficient
- Criticality assumptions on the concentration of radionuclides as saltstone hydrates (cures)
- Theoretical estimates of material properties, such as dry bulk density, that are used in SDF Performance Assessment flow and transport modeling.

Because cement chemistry is complex, difficult to simulate in detail, and expensive to characterize with experimental techniques, an approximate “normative” mineral composition of a hydrated cementitious material is commonly calculated. A normative analysis uses general knowledge of cement hydration reactions, equilibrium constants and kinetics, and degrees of reaction to virtually react the mix dry ingredients with water to form a plausible set of hydrated minerals constituting the cured material. Stoichiometry is used to preserve the collective mass of each chemical element through the process, based on the specified proportions and metal oxide analyses of the dry ingredients and the water-to-cementitious materials (w/c) ratio.

Normative mineral analyses of saltstone and SDF concrete were performed for the 1992 (WSRC-RP-92-1360), 2009 (SRR-CWDA-2009-00017), and 2019 (SRR-CWDA-2019-00001) SDF Performance Assessments (PAs) as documented in WSRC-RP-92-1360 Appendix D, SRNL-TR-2008-00283, and SRNL-STI-2018-00586, respectively. This report extends these previous works by analyzing both existing and proposed materials:

- Existing production saltstone composed of 45 weight percent (wt%) slag, 45 wt% fly ash, and 10 wt% cement (45/45/10 saltstone)
- Proposed cement-free saltstone composed of 60 wt% slag and 40 wt% fly ash as described in SRR-CWDA-2019-00003 (60/40/0 saltstone)

- Existing concrete used in Saltstone Disposal Unit (SDU) design types 2, 6, and 7 (SDU 2/6/7 concrete)
- Proposed concrete for future SDUs (e.g., SDU 8) identified as “Mix 3B” in mix design and testing report SRR-SDU-2019-00026 (SDU 8/Mix 3B)

and by deploying new methods:

- A slightly modified implementation of the Savannah River National Laboratory (SRNL) approach described in SRNL-STI-2018-00586
- An approach published by Herfort and Lothenbach (2016) and implemented in a spreadsheet template available from URL https://www.empa.ch/web/s308/ternary-diagram, *Calculation of ternary diagrams by mass balance calculations in MS Excel.*

The sections that follow define key inputs and assumptions, describe the normative mineral analyses, and present key results addressing the above-identified needs.

Inputs and assumptions

Table 1 specifies the ingredients and proportions associated with the saltstone and concrete mixes analyzed in this study.

Table 1. Cementitious material mix specifications.

Ingredient (lbs/yd^3)	**45/45/10 Saltstone**	**60/40/0 Saltstone**	**SDU 2/6/7 Concrete‡**	**SDU 8 / Mix 3B Concrete**
Cement (Type I/II)	157.6	—	213 (Type V)	337
Slag	709.4	947.6	284	284
Fly ash	709.4	631.8	163	—
Silica fume	—	—	50	—
Metakaolin	—	—	—	89
Sand	—	—	1046	1020
Coarse aggregate	—	—	1795	1850
Dissolved salts§	350.6	351.3	—	—
Water*	967.3	969.1	264	269
w/c†	0.614	0.614	0.372	0.379
g ingredients / kg dry mix**	1502.0	1501.9	1074.3	1075.1
Reference	SRR-CWDA-2020-00040 Table 15	SRR-CWDA-2020-00040 Table 19	C-SPP-Z-00015 Rev. 3 Attachment 03300-C	SRR-SDU-2019-00026 Table 4-7

† Water to cementitious materials (binders) mass ratio.
‡ Specifically SDU 7 floor concrete. The other SDU 2/6/7 concretes (C-SPP-Z-00015 Rev. 3 Attachment 03300-B) have the same binders, binder proportions, w/c, and sand + coarse aggregate proportion. They differ in their proportions of sand versus coarse aggregate. Aggregates are considered inert. Thus, SDU 2/6/7 concretes are the same material with respect to normative mineral composition.
§ Dissolved salts present in Decontaminated Salt Solution (DSS) to be disposed of.
* For saltstone: Water = DSS water + flush water. For Concrete: Water = Mixing water from concrete batch plant.
** For convenience, dissolved salts are considered part of the saltstone dry mix.

Table 2 summarizes major oxide analyses for the dry cements and pozzolans. The SRNL (2006) and SREL (2019) values are averages of multiple characterization results. The SRNL (2006), SIMCO (2010) and SIMCO (2012) columns are based on as-measured values and "Other" is computed as 100% minus the sum of the CaO through MgO oxides. In contrast, all SIMCO (2020) and SREL (2019) compounds including "Other" minor oxides and volatiles are normalized to 100% by rescaling all compounds. Figure 1 compares results from the various characterization efforts. Despite changes in ingredient sources over the years, the oxide proportions are generally consistent, an exception being much higher CaO measured by SREL (2019) for fly ash.

Table 2. Characterization of major oxides in dry cements and pozzolans.

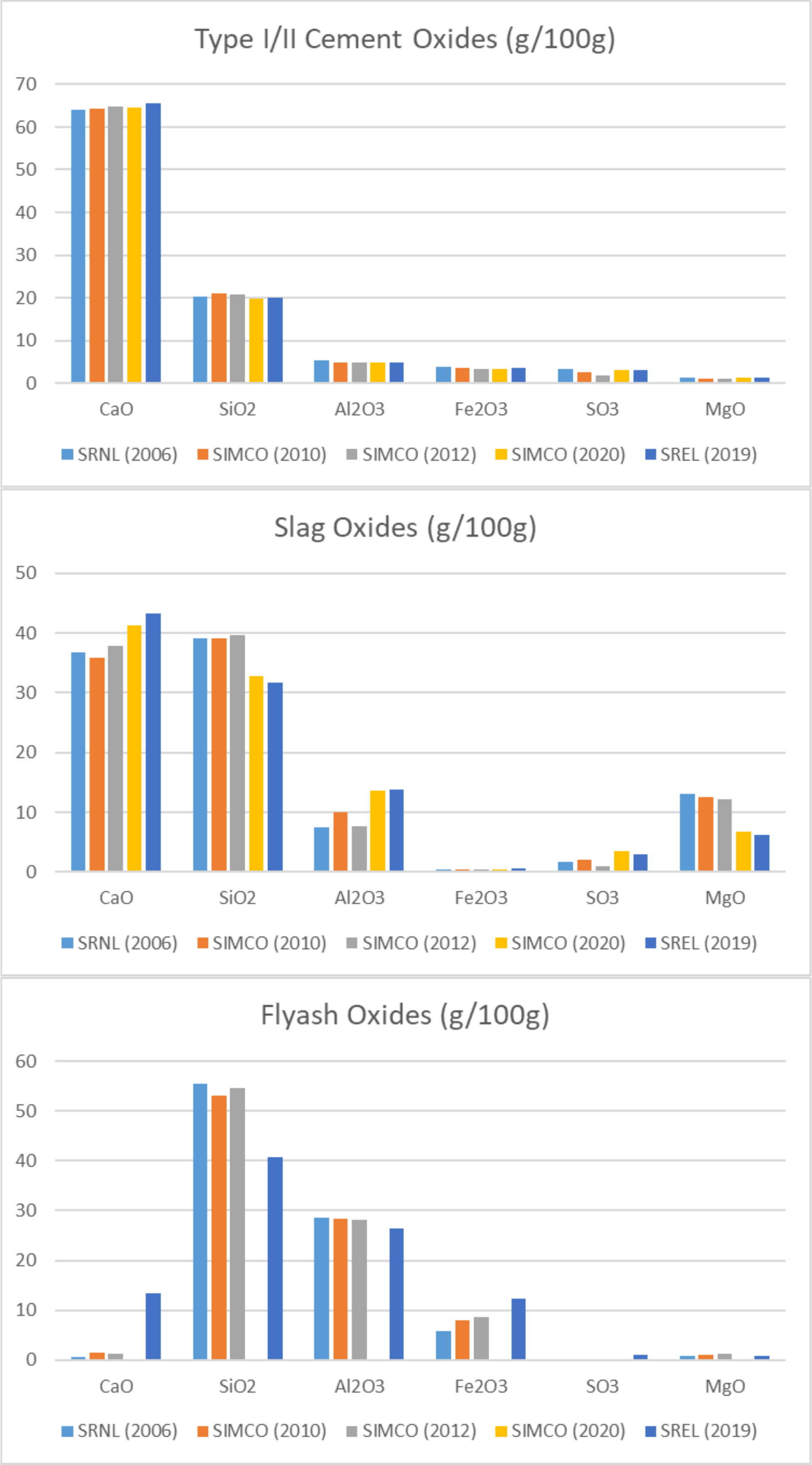
Oxide (g/100 g)	**SRNL (2006)†**	**SIMCO (2010) ‡**	**SIMCO (2012) ‡**	**SIMCO (2020) ‡**	**SREL (2019)§**
Cement					
CaO	64.0	64.3	64.8	64.4	65.4
SiO2	20.4	21.0	20.9	19.7	20.1
Al2O3	5.30	4.91	4.80	4.87	4.94
Fe2O3	3.75	3.50	3.43	3.35	3.58
SO3	3.25	2.64	1.75	2.97	3.19
MgO	1.20	0.95	1.05	1.28	1.34
Other	2.15	2.70	3.27	3.35	1.38
Slag					
CaO	36.8	35.8	37.8	41.2	43.3
SiO2	39.2	39.1	39.6	32.8	31.7
Al2O3	7.50	10.1	7.61	13.6	13.7
Fe2O3	0.35	0.36	0.47	0.41	0.57
SO3	1.75	1.99	1.05	3.46	3.04
MgO	13.0	12.6	12.2	6.75	6.15
Other	1.50	0.05	1.27	1.79	1.52
Fly ash					
CaO	0.65	1.41	1.32		13.4
SiO2	55.5	53.1	54.5		40.6
Al2O3	28.6	28.4	28.1		26.5
Fe2O3	5.80	7.99	8.65		12.4
SO3	0.20	0.00	0.00		1.03
MgO	0.85	1.00	1.19		0.75
Other	8.40	8.10	6.24		5.34
Silica fume					
CaO			0.60		
SiO2			95.0		
Al2O3			0.18		
Fe2O3			0.07		
SO3			0.18		
MgO			0.22		
Other			3.75		
Metakaolin					
CaO				0.16	
SiO2				53.1	
Al2O3				43.6	
Fe2O3				1.56	
SO3				0.02	
MgO				0.07	
Other				1.53	

† WSRC-TR-2006-00067.

‡ SIMCO data taken from reports listed in Reference section of this memo with matching publication year.

§ SREL Doc. No. R-19-0001.

Figure 1. Comparison of major oxides analyses for cement, slag, and fly ash.



Representation of cement hydration reactions and associated stoichiometry can be simplified using the cement chemist notation (CCN) presented in Table 3. For example, calcium oxide reacts with water to form calcium hydroxide:

$$CaO + H_2O \rightarrow Ca(OH)_2 \qquad (1)$$

Using CCN shorthand the same reaction can be abbreviated as

$$C + H \rightarrow CH \qquad (2)$$

because $Ca(OH)_2$ is stoichiometrically equivalent to $CaO \cdot H_2O$. CCN is not intended to capture the molecular structure of hydrated compounds but does preserve the mass of the reactants. CCN is generally used throughout the normative mineral composition analyses that follow, except for those compounds not included in Table 3 such as pyrrhotite (FeS).

Hydration reactions are not instantaneous due to finite chemical kinetics and mass transfer rates and may be precluded altogether for larger particle sizes where the core material is indefinitely isolated from the pore solution (source of H reactant). Thus, the degree of hydration is generally less than 100% at any point after the dry ingredients are mixed with water. Supplementary cementitious materials (SCM), that is the non-cement reactive ingredients, are notably less reactive than ordinary Portland cement, as evidenced by experimental measurements using techniques such as those reviewed by Scrivener et al. (2015). The degree of reaction/hydration of each binder is affected by several factors including mix composition such as SCM replacement level, solution pH, temperature, particle size distribution, and curing time (e.g. Lothenbach et al. 2011). Portland cement is more reactive than slag which is more reactive than fly ash (Lothenbach et al. 2011). The degree of reaction/hydration can be characterized for an overall mix (e.g. Xu et al. 2017) or for individual binders. The latter characterization better supports a normative mineral composition analysis and is the focus of Table 4, which summarizes several literature results for the binders present in the saltstones and concretes of this study.

Most of the results in Table 4 characterize degrees of reaction for curing periods under one year, whereas the SDF Performance Assessment considers material behavior past 1000 years. Two degree of hydration cases are considered in this study to support SDF PA timeframes: 100% and partial hydration. The first case assumes all binders will have ample time to completely react with water. The second case assumes 100% of cement, 70% of slag, and 40% of the other binders react, which represents the high end of the short-term results summarized in Table 4.

Table 3. Cement chemist notation (CCN).

Cement Chemist Notation	Molecular Formula	Name
C	CaO	Calcium oxide, or lime
S	SiO_2	Silicon dioxide, or silica
A	Al_2O_3	Aluminum oxide, or alumina
F	Fe_2O_3	Iron oxide, or rust
$\bar{S}$ or s	SO_3	Sulfur trioxide
M	MgO	Magnesium oxide, or periclase
T	TiO_2	Titanium dioxide, or titania
K	K_2O	Potassium oxide
N	Na_2O	Sodium oxide
$\bar{C}$ or c	CO_2	Carbon dioxide
P	P_2O_5	Phosphorus hemi-pentoxide
H	H_2O	Water

Table 4. Summary of literature survey on degree of hydration of cements and pozzolans.

Cement	Slag	Fly Ash	Silica Fume	Metakaolin	Condition	Reference
		15-30%			90d	Lam et al. (2000)
		30%	30-55%	40-55%	90d	Poon et al. (2001)
65-80%	40%	20%			~90d	Feng et al. (2004)
90% pred.					100d	Feng et al. (2014)
~100%	60-75%				20 y	Taylor et al. (2010)
		35%			140d	Haha et al. (2010)
	50%				180d	Haha et al. (2011)
	55-60%				100d	Le Saout et al. (2011)
85%		35%			150d	De Weerdt et al. (2011)
55-65%					~90d	Zhang and Scherer (2011)
	65-70%				2y	Kocaba et al. (2012)
	35%			20-25%	28d	Snellings et al. (2014)
75-95% 28d		15-40% 90d				Berodier, Scrivener (2015)
		60-75%			1y	Durdzinski et al. (2015)
	40-85%	12-50%			1y	Han et al. (2016)
		25-35%			2y	Bui et al. (2018)
		25%				Giergiczny (2019)
			35-65%			Liao et al. (2019)
		15-30%			180d	Wang and Ishida (2019)
	55%	22%	10%	40%	w/c 0.35, 23C	Ramanathan et al. (2019)
	55%	39%	20%	20%	w/c 0.35, 50C	
	40%	10%	40%	40%	w/c 0.50, 23C	
	70%	35%	30%	60%	w/c 0.50, 50C	
100%	**70%**	**40%**	**40%**	**40%**	**Long-term**	**Selections for this study**

Normative mineral analysis based on SRNL method

The first set of normative mineral analyses uses the method of SRNL-STI-2018-00586 with a few modifications:

- Portlandite and gypsum were added to the assumed set of potential minerals present in hydrated product to consume excess calcium and sulfur, respectively, if needed
- To ensure strict mass conservation, a) trace oxygen not accounted for when converting iron oxide and sulfur trioxide to pyrrhotite (FeS), b) minor oxides, and c) volatiles are tracked through the virtual hydration process
- The amount of unreacted water residing in the pore space of cured product is explicitly tracked.

Table 5 lists the resulting minerals and other compounds assumed to be potentially present in a hydrated paste. The assumed mineral set is a simplification of reality. For example, in the hydration model the crystalline mineral CSH represents all hydrated calcium and silica compounds. In practice however, a range of stoichiometries is observed and the structure is largely amorphous. This variable composition of hydrated calcium-silica compounds is commonly referred to as $C\text{-}S\text{-}H$ gel to distinguish it from the crystalline mineral CSH. As a second example, calcium-aluminum-silicate hydrates of variable composition, denoted $C\text{-}A\text{-}S\text{-}H$, have been observed but are not included in Table 5.

Table 5. Assumed mineral set for modified SRNL normative mineral composition analysis.

Potential Minerals	Cement Chemist Notation	Molecular Formula
Portlandite	CH	$Ca(OH)_2$
Calcium Silicate Hydrate	CSH	$CaSiO_3 \cdot H_2O$
Hydrotalcite	$4M \cdot A \cdot 10H$	$Mg_4Al_2O_7 \cdot 10H_2O$
Kaolinite	$A \cdot 2S \cdot 2H$	$Al_2Si_2O_5(OH)_4$
Gibbsite	$0.5A \cdot 1.5H$	$Al(OH)_3$
Gypsum	$Cs2H$	$CaSO_4 \cdot 2H_2O$
Pyrrhotite	—	FeS
Unreacted Quartz	S	SiO_2
Unreacted iron oxide	F	Fe_2O_3
Unreacted (pore) water	H	H_2O
Trace oxygen	—	O

Expanding upon SRNL-TR-2008-00283 and SRNL-STI-2018-00586, the general steps for reacting dry ingredients with water are:

- All sulfur available for reaction is combined with stoichiometric iron to form pyrrhotite (FeS), which represents reducing capacity. In some cases, pyrrhotite is limited by iron availability. Oxygen in the reactants becomes ‘trace oxygen’ in the products list.
- All available magnesium and stoichiometric aluminum are reacted with water to form hydrotalcite.
- Calcium-silicate-hydrate is formed based on the more limiting availability of calcium and silica.
- Portlandite, kaolinite, gibbsite, and gypsum consume any chemically available calcium, aluminum, and/or sulfur remaining after the above reactions.
- Any remaining silica and iron are captured as unreacted quartz and iron oxide, respectively.
- Unreacted mix water is assumed to occupy the pore space of the cured product.
- “Inert A” includes all minor oxides and volatiles in the dry binders, plus any fractions of the major oxides unavailable for reaction because the assumed degree of hydration is less than 100%.
- “Inert B” represents sand and coarse aggregate in concretes, and salt waste in saltstone mixes.

In SRNL-STI-2018-00586, the hydration products resulting from this process were then adjusted to match measured dry bulk density and chemical reduction capacity values to support equilibrium geochemistry modeling using *The Geochemist’s Workbench* software. This follow-on step is also performed in this study and discussed below in the section entitled *Equilibrium chemistry modeling inputs*.

Tables 6 and 7 summarize the results of the virtual hydration process assuming complete and partial hydration, respectively. Recall that the partial hydration scenario assumes the following degrees of reaction: cement 100%, slag 70%, other binders 40%. Note that the sums of all constituents (total products) match the total ingredient masses listed in Table 1. The same information is displayed graphically by Figures 2 and 3.

Besides reducing the cumulative mass of hydrated products, partial reaction also changes constituent proportions. Notably, Portlandite is generally absent in the 100% hydrated binders but generally present under partial hydration. Portlandite buffers the pore solution at pH = 12.4, whereas pH is lower when only calcium-silicate-hydrate gel (C-S-H) is present.

Table 6. Estimated mineral compositions for complete hydration using the modified SRNL method, unadjusted for measured bulk density and reduction capacity.

Constituent (g/kg dry mix)†	45/45/10 Saltstone	60/40/0 Saltstone	SDU 2/6/7 Concrete	SDU 8 / Mix 3B Concrete
Portlandite, CH	0.0	0.0	0.0	50.6
Calcium Silicate Hydrate, CSH	460.3	613.1	164.8	129.0
Hydrotalcite, $4M \cdot A \cdot 10H$	142.6	90.2	31.8	18.2
Kaolinite, $4M \cdot A \cdot 10H$	271.2	0.0	0.0	0.0
Gibbsite, $0.5A \cdot 1.5H$	0.0	203.7	23.5	33.6
Quartz, S	33.4	14.2	9.0	0.0
Pyrrhotite, FeS	9.2	20.1	3.1	4.3
Gypsum, $Cs2H$	0.0	0.0	0.0	3.6
Unreacted F	22.5	25.0	3.1	0.0
Unreacted (pore) water, H	344.2	312.4	31.1	25.7
Oxygen, O	7.6	16.4	2.6	3.5
Inert A	29.1	24.9	5.3	5.0
Inert B	181.9	182.0	800.1	801.7
Total product	1502.0	1501.9	1074.3	1075.1

† Dry mix includes dissolved salts.

Table 7. Estimated mineral compositions for partial hydration using the modified SRNL method, unadjusted for measured bulk density and reduction capacity.

Constituent (g/kg dry mix)†	45/45/10 Saltstone	60/40/0 Saltstone	SDU 2/6/7 Concrete	SDU 8 / Mix 3B Concrete
Portlandite, CH	0.0	19.6	17.1	57.5
Calcium Silicate Hydrate, CSH	357.6	362.2	111.8	93.9
Hydrotalcite, $4M \cdot A \cdot 10H$	97.8	61.1	22.6	13.7
Kaolinite, $4M \cdot A \cdot 10H$	85.7	0.0	0.0	0.0
Gibbsite, $0.5A \cdot 1.5H$	16.9	103.6	11.6	20.3
Quartz, S	0.0	0.0	0.0	0.0
Pyrrhotite, FeS	7.0	13.0	2.6	3.9
Gypsum, $Cs2H$	0.0	0.0	0.0	2.6
Unreacted F	8.3	6.4	1.5	0.0
Unreacted (pore) water, H	396.3	387.8	41.9	35.4
Oxygen, O	5.7	10.6	2.2	3.2
Inert A	344.8	355.8	62.9	43.0
Inert B	181.9	182.0	800.1	801.7
Total product	1502.0	1501.9	1074.3	1075.1

† Dry mix includes dissolved salts.

Figure 2. Normative mineral compositions assuming <u>complete</u> hydration using the modified SRNL method, unadjusted for measured bulk density and reduction capacity.

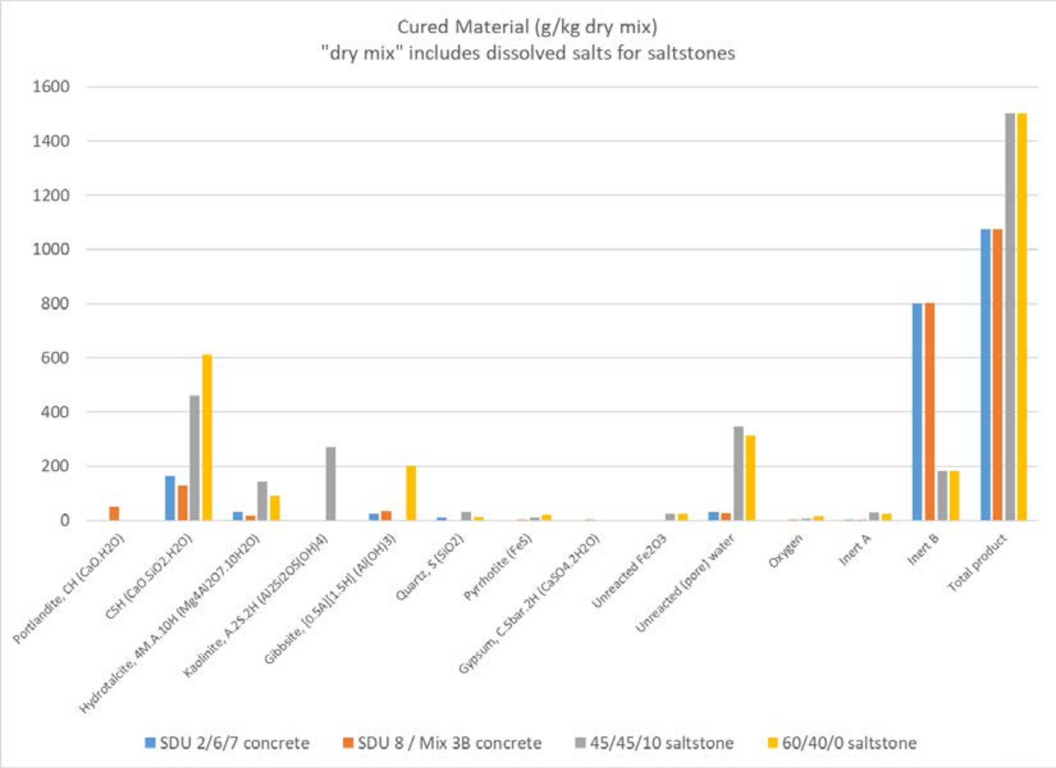
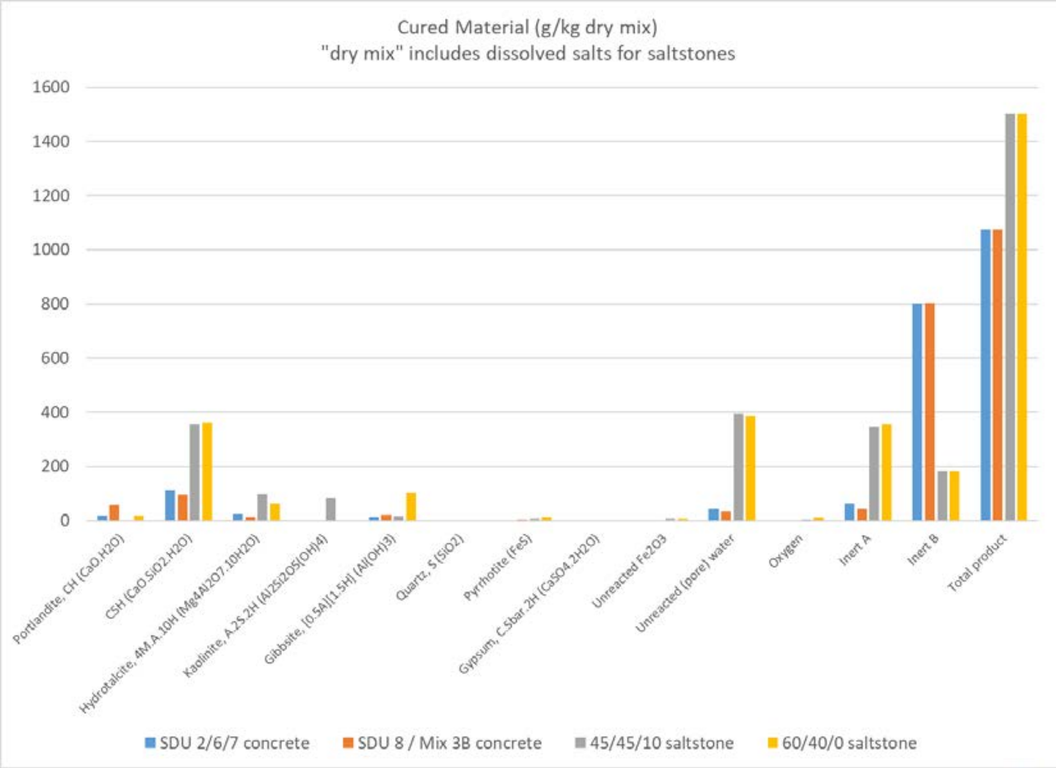


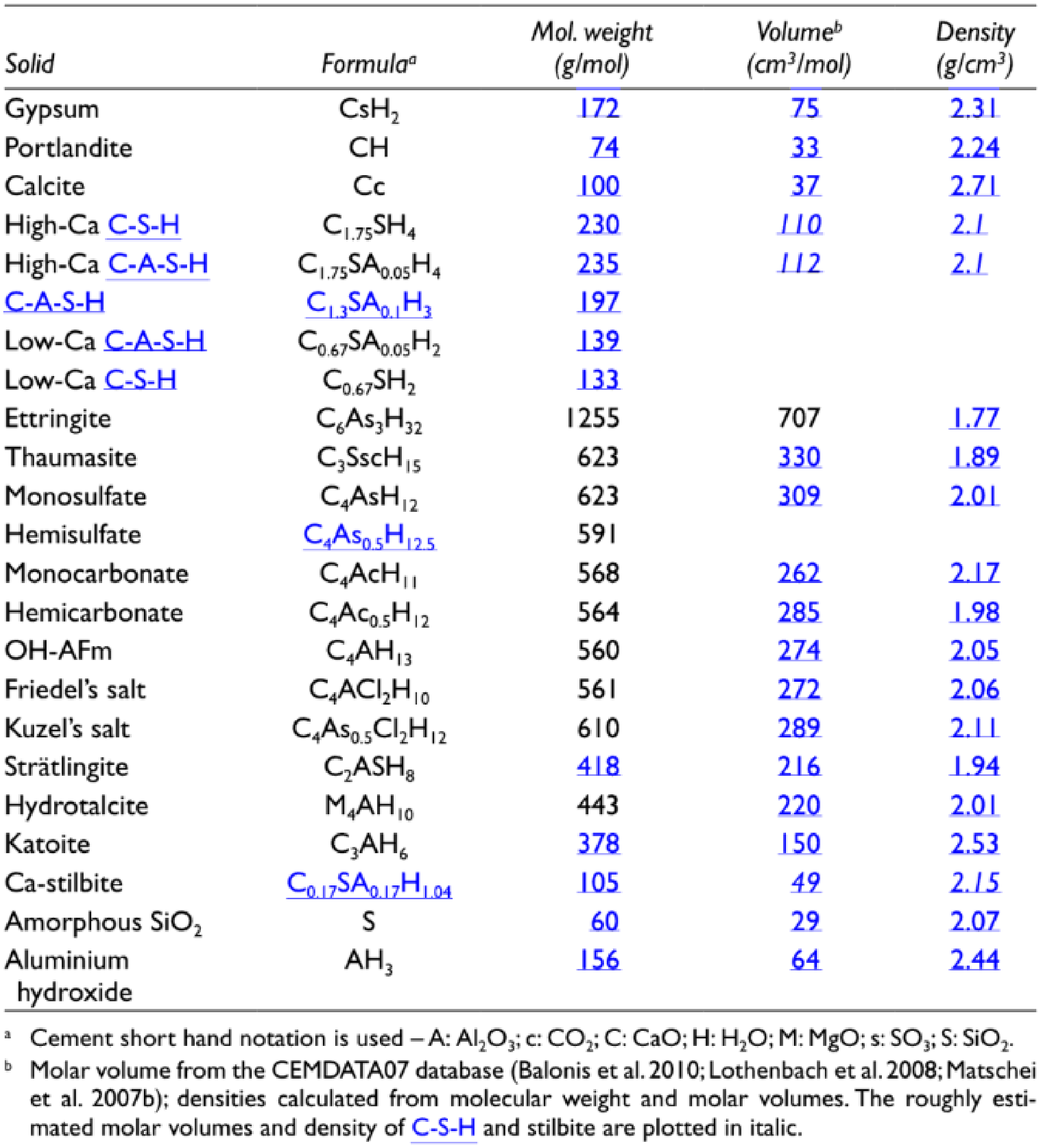
Figure 3. Normative mineral compositions assuming <u>partial</u> hydration using the modified SRNL method, unadjusted for measured bulk density and reduction capacity.



Normative mineral analysis based on Herfort and Lothenbach (2016)

The second set of normative mineral analyses considered in this study is based on a method published by Herfort and Lothenbach (2016) and implemented in a spreadsheet template available from URL https://www.empa.ch/web/s308/ternary-diagram, *Calculation of ternary diagrams by mass balance calculations in MS Excel*. The Herfort and Lothenbach method considers the much larger set of potential hydrated minerals shown in Figure 4, an excerpt from their publication. The spreadsheet template requires w/c ≥ 0.50 compared to w/c = 0.38± for SDU concretes. Because spreadsheet formulas are protected, and thus inaccessible to the user, w/c = 0.50 is applied to SDU concrete hydration. The impacts of this deviation are additional unreacted water and probably minimal influence on the normative mineral composition. Figures 5 and 6 illustrate mineral compositions under the complete and partial hydration scenarios, where principal species are displayed on ternary diagrams based on CaO, SiO_2, and Al_2O_3 oxide content. Note that units differ between the modified-SRNL and Herfort-Lothenbach normative analyses: g/kg dry mix versus g/100g hydrated binder. The hydration products from both analyses are presented in units of g/m^3 in the next section entitled *Equilibrium chemistry modeling inputs* and can be more directly compared there.

Figure 4. Hydrated minerals considered by Herfort and Lothenbach (2016).



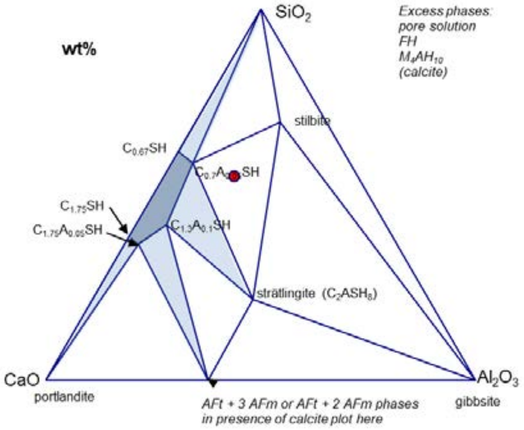
Solid	Formula[a]	Mol. weight (g/mol)	Volume[b] (cm^3/mol)	Density (g/cm^3)
Gypsum	CsH_2	172	75	2.31
Portlandite	CH	74	33	2.24
Calcite	Cc	100	37	2.71
High-Ca C-S-H	$C_{1.75}SH_4$	230	*110*	*2.1*
High-Ca C-A-S-H	$C_{1.75}SA_{0.05}H_4$	235	*112*	*2.1*
C-A-S-H	$C_{1.3}SA_{0.1}H_3$	197		
Low-Ca C-A-S-H	$C_{0.67}SA_{0.05}H_2$	139		
Low-Ca C-S-H	$C_{0.67}SH_2$	133		
Ettringite	$C_6As_3H_{32}$	1255	707	1.77
Thaumasite	C_3SscH_{15}	623	330	1.89
Monosulfate	C_4AsH_{12}	623	309	2.01
Hemisulfate	$C_4As_{0.5}H_{12.5}$	591		
Monocarbonate	C_4AcH_{11}	568	262	2.17
Hemicarbonate	$C_4Ac_{0.5}H_{12}$	564	285	1.98
OH-AFm	C_4AH_{13}	560	274	2.05
Friedel's salt	$C_4ACl_2H_{10}$	561	272	2.06
Kuzel's salt	$C_4As_{0.5}Cl_2H_{12}$	610	289	2.11
Strätlingite	C_2ASH_8	418	216	1.94
Hydrotalcite	M_4AH_{10}	443	220	2.01
Katoite	C_3AH_6	378	150	2.53
Ca-stilbite	$C_{0.17}SA_{0.17}H_{1.04}$	105	*49*	*2.15*
Amorphous SiO_2	S	60	29	2.07
Aluminium hydroxide	AH_3	156	64	2.44

[a] Cement short hand notation is used – A: Al_2O_3; c: CO_2; C: CaO; H: H_2O; M: MgO; s: SO_3; S: SiO_2.

[b] Molar volume from the CEMDATA07 database (Balonis et al. 2010; Lothenbach et al. 2008; Matschei et al. 2007b); densities calculated from molecular weight and molar volumes. The roughly estimated molar volumes and density of C-S-H and stilbite are plotted in italic.

Figure 5. Normative mineral compositions for complete hydration using the Herfort and Lothenbach (2016) method.

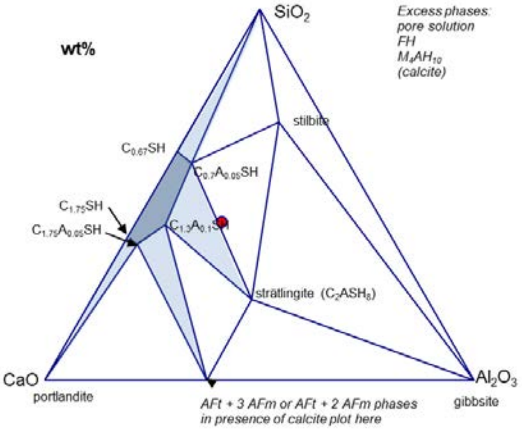
45/45/10 Saltstone:



output: hydrates

	g/100 g hydrated binder
water	16.68
strätlingite	15.40
Ca-stilbite	21.04
C0.67A0.05SH1.8	32.77
gypsum	0.40
calcite	0.00
ferrihydrite	2.68
hydrotalcite	11.04
C-A-S-H total	*32.8*
Ca/Si	0.67
Al/Si	0.05

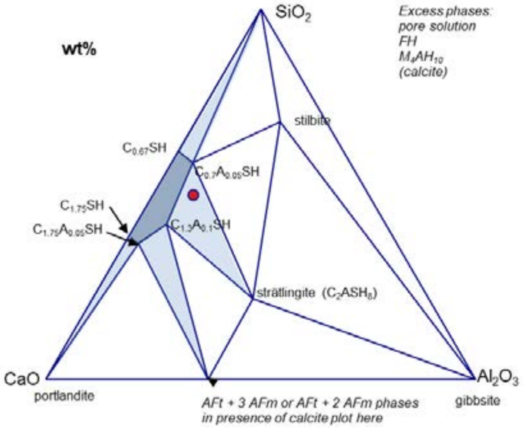
60/40/0 Saltstone:



output: hydrates

	g/100 g hydrated binder
water	13.50
strätlingite	35.66
Ca-stilbite	2.16
C0.67A0.05SH1.8	37.34
gypsum	0.56
calcite	0.00
ferrihydrite	3.76
hydrotalcite	7.01
C-A-S-H total	*37.3*
Ca/Si	0.67
Al/Si	0.05

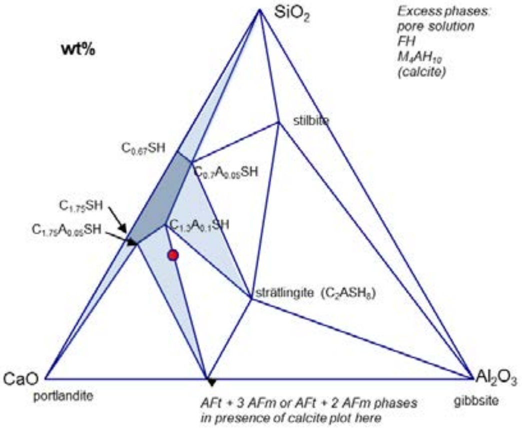
SDU 2/6/7 Concrete:



output: hydrates

	g/100 g hydrated binder
water	7.12
C1.3A0.1SH3	19.70
strätlingite	9.24
C0.67A0.05SH1.8	48.19
ettringite	2.73
calcite	0.00
ferrihydrite	2.25
hydrotalcite	10.79
C-A-S-H total	*67.9*
Ca/Si	0.85
Al/Si	0.06

Mix 3B Concrete:

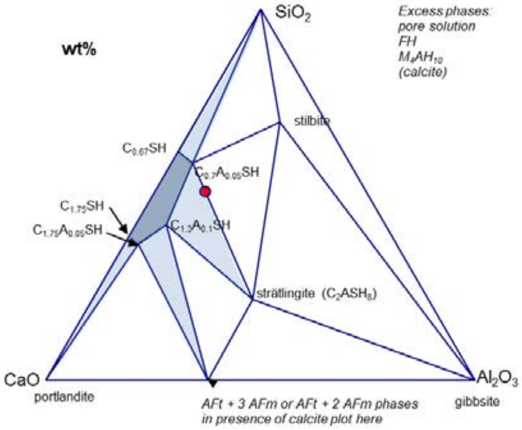


output: hydrates

	g/100 g hydrated binder
water	6.31
C1.3A0.1SH3	64.96
strätlingite	1.04
Hydrogarnet	5.67
Hemicarbonate	0.00
Hemisulphate	14.29
ferrihydrite	1.49
hydrotalcite	6.24
C-A-S-H total	*65.0*
Ca/Si	1.30
Al/Si	0.10

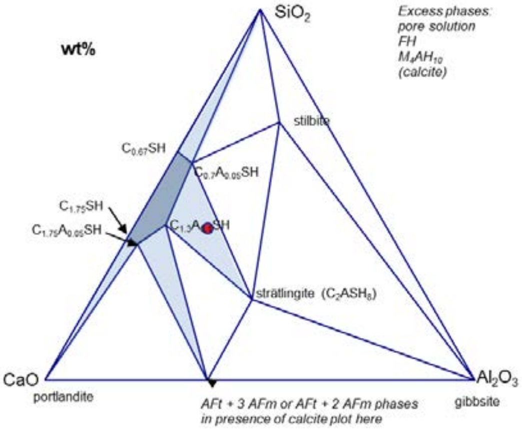
Figure 6. Normative mineral compositions for partial hydration using the Herfort and Lothenbach (2016) method.

45/45/10 Saltstone:



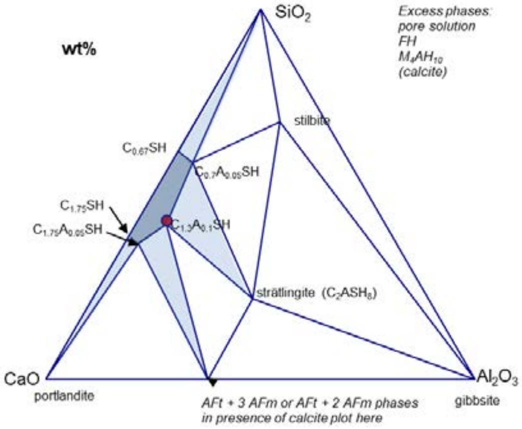
output: hydrates	g/100 g hydrated binder
water	31.43
C1.3A0.1SH3	0.66
strätlingite	12.03
C0.67A0.05SH1.8	43.04
ettringite	1.15
calcite	0.00
ferrihydrite	1.68
hydrotalcite	10.02
C-A-S-H total	*43.7*
Ca/Si	0.68
Al/Si	0.05

60/40/0 Saltstone:



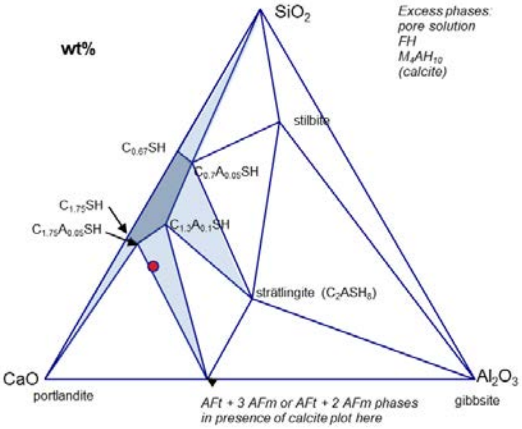
output: hydrates	g/100 g hydrated binder
water	32.15
C1.3A0.1SH3	14.49
strätlingite	22.60
C0.67A0.05SH1.8	21.54
ettringite	0.74
calcite	0.00
ferrihydrite	2.12
hydrotalcite	6.37
C-A-S-H total	*36.0*
Ca/Si	0.92
Al/Si	0.07

SDU 2/6/7 Concrete:



output: hydrates	g/100 g hydrated binder
water	18.31
C1.3A0.1SH3	57.44
C0.67SH1.8	6.34
C1.75SH4.4	3.29
ettringite	3.29
calcite	0.00
ferrihydrite	1.82
hydrotalcite	9.51
C-A-S-H total	*67.1*
Ca/Si	1.26
Al/Si	0.09

Mix 3B Concrete:



output: hydrates	g/100 g hydrated binder
water	11.02
C1.3A0.1SH3	11.95
C1.75A0.05SH4.3	50.99
Hydrogarnet	2.70
Hemicarbonate	0.00
Hemisulphate	16.38
ferrihydrite	1.56
hydrotalcite	5.40
C-A-S-H total	*62.9*
Ca/Si	1.66
Al/Si	0.06

<u>Equilibrium chemistry modeling inputs</u>

Previous simulations of saltstone and SDF concrete equilibrium chemistry using *The Geochemist's Workbench* (SRNL-TR-2008-00283, SRNL-STI-2018-00586) were initialized with normative mineral compositions expressed in units of g/m^3. To support potential geochemical modeling, Tables 8 through 11 present normative mineral compositions in these dimensional units for the modified-SRNL and Herfort-Lothenbach methods and complete and partial hydration scenarios. The common units also facilitate direct comparison of results from the two analysis methods.

Table 8. Normative mineral composition for <u>complete</u> hydration using the modified-SRNL method expressed as g/m^3.

Constituent	45/45/10 Saltstone	60/40/0 Saltstone	SDU2/6/7 Concrete	Mix 3B Concrete
Portlandite, CH	-	-	-	107,495
Calcium Silicate Hydrate, CSH	526,267	702,214	347,243	274,062
Hydrotalcite, $4M \cdot A \cdot 10H$	163,028	103,286	66,967	38,609
Kaolinite, $4M \cdot A \cdot 10H$	309,992	-	-	-
Gibbsite, $0.5A \cdot 1.5H$	-	233,354	49,547	71,450
Quartz, S	38,157	16,246	18,869	-
Pyrrhotite, FeS	10,553	23,027	6,582	9,036
Gypsum, $Cs2H$	-	-	-	7,686
Unreacted F	25,729	28,674	6,491	-
Unreacted (pore) water, H	393,537	357,778	65,558	54,565
Oxygen, O	8,634	18,841	5,386	7,393
Inert A	33,218	28,544	11,208	10,520
Inert B	208,003	208,418	1,685,497	1,702,702
Total product	1,717,118	1,720,381	2,263,347	2,283,519
Total - pore water	1,323,581	1,362,603	**2,197,789**	**2,228,954**
Total - pore water - Inert B	**1,115,579**	**1,154,185**	512,293	526,252

Table 9. Normative mineral composition for <u>partial</u> hydration using the modified-SRNL method expressed as g/m^3.

Constituent	45/45/10 Saltstone	60/40/0 Saltstone	SDU2/6/7 Concrete	Mix 3B Concrete
Portlandite, CH	-	22,471	36,117	122,025
Calcium Silicate Hydrate, CSH	408,793	414,868	235,609	199,523
Hydrotalcite, $4M \cdot A \cdot 10H$	111,780	69,971	47,514	29,120
Kaolinite, $4M \cdot A \cdot 10H$	97,930	-	-	-
Gibbsite, $0.5A \cdot 1.5H$	19,327	118,619	24,488	43,178
Quartz, S	-	-	-	-
Pyrrhotite, FeS	7,997	14,850	5,572	8,263
Gypsum, $Cs2H$	-	-	-	5,418
Unreacted F	9,516	7,305	3,056	-
Unreacted (pore) water, H	453,087	444,194	88,364	75,168
Oxygen, O	6,543	12,150	4,559	6,761
Inert A	394,142	407,536	132,572	91,359
Inert B	208,003	208,418	1,685,497	1,702,702
Total product	1,717,118	1,720,381	2,263,347	2,283,519
Total - pore water	1,264,031	1,276,187	**2,174,983**	**2,208,350**
Total - pore water - Inert B	**1,056,029**	**1,067,770**	489,486	505,649

Table 10. Normative mineral composition for <u>complete</u> hydration using the Herfort-Lothenbach method expressed as g/m³.

45/45/10 Saltstone		60/40/0 Saltstone		SDU2/6/7 Concrete		Mix 3B Concrete	
H (unreacted)	251,688	H (unreacted)	204,165	H (unreacted)	44,985	H (unreacted)	39,893
C_2ASH_8	232,415	C_2ASH_8	539,192	$C_{1.3}A_{0.1}SH_3$	124,455	$C_{1.3}A_{0.1}SH_3$	410,438
$C_{0.17}A_{0.17}SH_{1.04}$	317,509	$C_{0.17}A_{0.17}SH_{1.04}$	32,711	C_2ASH_8	58,356	C_2ASH_8	6,573
$C_{0.67}A_{0.05}SH_{1.8}$	494,559	$C_{0.67}A_{0.05}SH_{1.8}$	564,519	$C_{0.67}A_{0.05}SH_{1.8}$	304,474	C_3AH_6	35,817
CsH_2	5,964	CsH_2	8,524	$C_6As_3H_{32}$	17,228	$C_4Ac_{0.5}H_{12}$	-
Cc	-	Cc	-	Cc	-	$C_4As_{0.5}H_{12.5}$	90,293
F_5H_9	40,371	F_5H_9	56,904	F_5H_9	14,197	F_5H_9	9,384
M_4AH_{10}	166,671	M_4AH_{10}	105,990	M_4AH_{10}	68,169	M_4AH_{10}	39,454

Table 11. Normative mineral composition for <u>partial</u> hydration using the Herfort-Lothenbach method expressed as g/m³.

45/45/10 Saltstone		60/40/0 Saltstone		SDU2/6/7 Concrete		Mix 3B Concrete	
H (unreacted)	355,264	H (unreacted)	359,562	H (unreacted)	92,543	H (unreacted)	60,573
$C_{1.3}A_{0.1}SH_3$	7,405	$C_{1.3}A_{0.1}SH_3$	162,024	$C_{1.3}A_{0.1}SH_3$	290,361	$C_{1.3}A_{0.1}SH_3$	65,687
C_2ASH_8	135,987	C_2ASH_8	252,712	$C_{0.67}SH_{1.8}$	32,042	$C_{1.75}A_{0.05}SH_{4.3}$	280,221
$C_{0.67}A_{0.05}SH_{1.8}$	486,448	$C_{0.67}A_{0.05}SH_{1.8}$	240,937	$C_{1.75}SH_{4.4}$	16,645	C_3AH_6	14,846
$C_6As_3H_{32}$	13,003	$C_6As_3H_{32}$	8,225	$C_6As_3H_{32}$	16,641	$C_4Ac_{0.5}H_{12}$	-
Cc	-	Cc	-	Cc	-	$C_4As_{0.5}H_{12.5}$	90,027
F_5H_9	19,014	F_5H_9	23,692	F_5H_9	9,185	F_5H_9	8,565
M_4AH_{10}	113,253	M_4AH_{10}	71,282	M_4AH_{10}	48,071	M_4AH_{10}	29,702

SRNL-STI-2018-00586 also adjusted its initial normative mineral compositions to exactly match bulk density and reduction capacity recommended for SDU 2/6/7 concrete and 45/45/10 saltstone for PA modeling. Using the same process, the initial compositions presented in Tables 8 and 9 were adjusted to reproduce the values recommended for Performance Assessment modeling shown in Table 12, even though these values may be biased for compliance modeling conservatism. The adjusted normative mineral compositions are given in Tables 13 and 14.

Table 12. Recommended values for Performance Assessment compliance modeling.

Parameter	45/45/10 Saltstone	Ref.	60/40/0 Saltstone	Ref.	SDU 2/6/7 Concrete	Ref.	Mix 3B Concrete	Ref.
Dry bulk density (g/mL)	0.932	(a), (c)	0.932	(c)	2.18	(a), (b)	2.25	(b)
Reduction capacity (µeq/g)	500	(c), (d), (e), (f)	500	(c), (g)	209	(d), (f)	209	(h)
Porosity	0.656	(a), (c)	0.656	(c)	0.110	(a), (b)	0.129	(b)

(a) SRR-CWDA-2018-00004, Rev. 1, Table 1

(b) SRR-CWDA-2020-00036, Table 5

(c) SRR-CWDA-2020-00040, Table 21

(d) SRNL-STI-2018-00586, Table 2-1, "Compliance Case"

(e) SRR-CWDA-2018-00048, Table 4, "Compliance" modeling case

(f) SRR-CWDA-2019-00001, Table 4.3-9, "Compliance" value

(g) Assumed to be the same as 45/45/10 saltstone

(h) Assumed to be the same as SDU 2/6/7 concrete

Table 13. Normative mineral composition for <u>complete</u> hydration using the modified-SRNL method adjusted to recommended bulk density and reduction capacity expressed as g/m^3.

Constituent	45/45/10 Saltstone	60/40/0 Saltstone	SDU2/6/7 Concrete	Mix 3B Concrete
Portlandite, CH	-	-	-	108,702
Calcium Silicate Hydrate, CSH	440,818	574,608	344,673	277,138
Hydrotalcite, $4M \cdot A \cdot 10H$	136,557	84,517	66,471	39,042
Kaolinite, $4M \cdot A \cdot 10H$	259,659	-	-	-
Gibbsite, $0.5A \cdot 1.5H$		190,948	49,180	72,252
Quartz, S	31,962	13,294	18,730	-
Pyrrhotite, FeS	6,396	6,396	5,007	5,168
Gypsum, $Cs2H$	-	-	-	7,772
Unreacted F	21,552	23,463	6,443	-
Unreacted (pore) water, H	329,639	292,763	65,073	55,177
Oxygen, O	7,232	15,417	5,346	7,476
Inert A	27,824	23,357	11,125	10,638
Inert B	208,003	208,418	1,673,025	1,721,812
Total product	1,469,641	1,433,180	2,245,073	2,305,177
Total - pore water	1,140,002	1,140,417	**2,180,000**	**2,250,000**
Total - pore water - Inert B	**932,000**	**931,999**	506,975	528,188

Table 14. Normative mineral composition for <u>partial</u> hydration using the modified-SRNL method adjusted to recommended bulk density and reduction capacity expressed as g/m^3.

Constituent	45/45/10 Saltstone	60/40/0 Saltstone	SDU2/6/7 Concrete	Mix 3B Concrete
Portlandite, CH	-	19,753	36,210	124,507
Calcium Silicate Hydrate, CSH	361,040	364,703	236,216	203,581
Hydrotalcite, $4M \cdot A \cdot 10H$	98,723	61,510	47,637	29,713
Kaolinite, $4M \cdot A \cdot 10H$	86,490	-	-	-
Gibbsite, $0.5A \cdot 1.5H$	17,069	104,276	24,551	44,056
Quartz, S	-	-	-	-
Pyrrhotite, FeS	6,396	6,396	5,007	5,168
Gypsum, $Cs2H$	-	-	-	5,528
Unreacted F	8,405	6,422	3,063	-
Unreacted (pore) water, H	400,159	390,484	88,592	76,697
Oxygen, O	5,779	10,681	4,571	6,899
Inert A	348,100	358,258	132,913	93,217
Inert B	208,003	208,418	1,689,834	1,737,331
Total product	1,540,161	1,530,901	2,268,592	2,326,697
Total - pore water	1,140,002	1,140,417	**2,180,000**	**2,250,000**
Total - pore water - Inert B	**932,000**	**932,000**	490,166	512,669

<u>Degradation analysis inputs</u>

The cementitious materials degradation analyses described in SRNL-STI-2018-00077 Rev. 1 and SRR-CWDA-2019-00001 Section 4.4.2 rely on estimates of aluminum and calcium concentrations in mol/g-solid to model sulfate attack on concrete and calcium concentrations in mol/cm^3 to model carbonation in concrete and decalcification of saltstone. Table 15 summarizes these concentrations estimated for the cured materials.

Table 15. Degradation analysis inputs.

Parameter	45/45/10 Saltstone	60/40/0 Saltstone	SDU2/6/7 Concrete	Mix 3B Concrete
Modified SRNL method w/property adjustment, complete hydration				
Aluminum concentration, mol/g-solid	2.82E-03	3.04E-03	4.27E-04	4.90E-04
Calcium concentration, mol/g-solid	3.53E-03	4.60E-03	1.18E-03	1.59E-03
Calcium concentration, mol/cm^3	3.29E-03	4.29E-03	2.57E-03	3.58E-03
Modified SRNL method w/property adjustment, partial hydration				
Aluminum concentration, mol/g-solid	1.43E-03	1.73E-03	2.43E-04	3.11E-04
Calcium concentration, mol/g-solid	2.89E-03	3.21E-03	1.03E-03	1.44E-03
Calcium concentration, mol/cm^3	2.69E-03	2.99E-03	2.25E-03	3.23E-03
Herfort-Lothenbach method, complete hydration				
Aluminum concentration, mol/g-solid	2.59E-03	2.74E-03	4.26E-04	4.89E-04
Calcium concentration, mol/g-solid	3.27E-03	4.19E-03	1.19E-03	1.58E-03
Calcium concentration, mol/cm^3	4.11E-03	5.48E-03	2.69E-03	3.64E-03
Herfort-Lothenbach method, partial hydration				
Aluminum concentration, mol/g-solid	1.34E-03	1.64E-03	2.42E-04	3.08E-04
Calcium concentration, mol/g-solid	2.75E-03	3.05E-03	1.03E-03	1.41E-03
Calcium concentration, mol/cm^3	3.18E-03	3.51E-03	2.28E-03	3.20E-03

Criticality analysis input

N-NCS-Z-00001 Rev. 13, *Nuclear Criticality Safety Evaluation for Z-Area (U)*, Section 5.2.2 uses the ratio of salt solution mass to pore solution mass computed from the normative mineral analysis (“hydration calculation”) in the 1992 Saltstone PA (WSRC-RP-92-1360, Table D.3-2); that ratio is 88.68 / 72.89 = 1.22. Tables 16 through 19 present the comparable calculation for 45/45/10 and 60/40/0 saltstone assuming complete and partial hydration. “Salt solution” is the sum of “Salt” and “Water” among the mix ingredients. “Pore solution” is the sum of “Unreacted (pore) water” and “Inert B (salt)” among the cured products. The mass ratio results are summarized as

- 1.30 = 45/45/10 saltstone, complete hydration
- 1.38 = 60/40/0 saltstone, complete hydration
- 1.18 = 45/45/10 saltstone, partial hydration
- 1.20 = 60/40/0 saltstone, partial hydration

and indicate little difference between the two saltstone formulations.

Table 16. Salt / pore solution ratio for 45/45/10 saltstone and complete hydration.

Ingredient	g/100g dry mix
C (CaO)	19.24
S (SiO2)	36.56
A (Al2O3)	14.01
F (Fe2O3)	3.09
Sbar (SO3)	0.84
M (MgO)	5.16
Inert A	2.91
Inert B	18.19
Water	50.20

Ingredient	g/100g	g/kg
Binders	81.81	818.1
Salt	18.19	181.9
Water	50.20	502.0
Total	150.20	1502.0
Salt solution		683.9

Hydrated grout	g/kg
Portlandite, CH (CaO.H2O)	0.0
CSH (CaO.SiO2.H2O)	460.3
Hydrotalcite, 4M.A.10H (Mg4Al2O7.10H2O)	142.6
Kaolinite, A.2S.2H (Al2Si2O5(OH)4)	271.2
Gibbsite, [0.5A][1.5H] (Al(OH)3)	0.0
Quartz, S (SiO2)	33.4
Pyrrhotite (FeS)	9.2
Gypsum, C.Sbar.2H (CaSO4.2H2O)	0.0
Unreacted Fe2O3	22.5
Unreacted (pore) water	344.2
Oxygen	7.6
Inert A	29.1
Inert B (Salt)	181.9
Total	1502.0
Pore solution	526.2
Salt solution / pore solution ratio	1.30

Table 17. Salt / pore solution ratio for 45/45/10 saltstone and partial hydration.

Ingredient	g/100g dry mix
C (CaO)	14.94
S (SiO2)	19.99
A (Al2O3)	6.75
F (Fe2O3)	1.47
Sbar (SO3)	0.64
M (MgO)	3.54
Inert A	34.48
Inert B	18.19
Water	50.20

Ingredient	g/100g	g/kg
Binders	81.81	818.1
Salt	18.19	181.9
Water	50.20	502.0
Total	150.20	1502.0
Salt solution		683.9

Hydrated grout	g/kg
Portlandite, CH (CaO.H2O)	0.0
CSH (CaO.SiO2.H2O)	357.6
Hydrotalcite, 4M.A.10H (Mg4Al2O7.10H2O)	97.8
Kaolinite, A.2S.2H (Al2Si2O5(OH)4)	85.7
Gibbsite, [0.5A][1.5H] (Al(OH)3)	16.9
Quartz, S (SiO2)	0.0
Pyrrhotite (FeS)	7.0
Gypsum, C.Sbar.2H (CaSO4.2H2O)	0.0
Unreacted Fe2O3	8.3
Unreacted (pore) water	396.3
Oxygen	5.7
Inert A	344.8
Inert B (Salt)	181.9
Total	1502.0
Pore solution	578.3
Salt solution / pore solution ratio	1.18

Table 18. Salt / pore solution ratio for 60/40/0 saltstone and complete hydration.

Ingredient	g/100g dry mix
C (CaO)	25.62
S (SiO2)	28.87
A (Al2O3)	15.40
F (Fe2O3)	4.33
Sbar (SO3)	1.83
M (MgO)	3.26
Inert A	2.49
Inert B	18.20
Water	50.19

Ingredient	g/100g	g/kg
Binders	81.80	818.0
Salt	18.20	182.0
Water	50.19	501.9
Total	150.19	1501.9
Salt solution		683.9

Hydrated grout	g/kg
Portlandite, CH (CaO.H2O)	0.0
CSH (CaO.SiO2.H2O)	613.1
Hydrotalcite, 4M.A.10H (Mg4Al2O7.10H2O)	90.2
Kaolinite, A.2S.2H (Al2Si2O5(OH)4)	0.0
Gibbsite, [0.5A][1.5H] (Al(OH)3)	203.7
Quartz, S (SiO2)	14.2
Pyrrhotite (FeS)	20.1
Gypsum, C.Sbar.2H (CaSO4.2H2O)	0.0
Unreacted Fe2O3	25.0
Unreacted (pore) water	312.4
Oxygen	16.4
Inert A	24.9
Inert B (Salt)	182.0
Total	1501.9
Pore solution	494.3
Salt solution / pore solution ratio	**1.38**

Table 19. Salt / pore solution ratio for 60/40/0 saltstone and partial hydration.

Ingredient	g/100g dry mix
C (CaO)	16.62
S (SiO2)	16.22
A (Al2O3)	8.18
F (Fe2O3)	1.82
Sbar (SO3)	1.18
M (MgO)	2.21
Inert A	35.58
Inert B	18.20
Water	50.19

Ingredient	g/100g	g/kg
Binders	81.80	818.0
Salt	18.20	182.0
Water	50.19	501.9
Total	150.19	1501.9
Salt solution		683.9

Hydrated grout	g/kg
Portlandite, CH (CaO.H2O)	19.6
CSH (CaO.SiO2.H2O)	362.2
Hydrotalcite, 4M.A.10H (Mg4Al2O7.10H2O)	61.1
Kaolinite, A.2S.2H (Al2Si2O5(OH)4)	0.0
Gibbsite, [0.5A][1.5H] (Al(OH)3)	103.6
Quartz, S (SiO2)	0.0
Pyrrhotite (FeS)	13.0
Gypsum, C.Sbar.2H (CaSO4.2H2O)	0.0
Unreacted Fe2O3	6.4
Unreacted (pore) water	387.8
Oxygen	10.6
Inert A	355.8
Inert B (Salt)	182.0
Total	1501.9
Pore solution	569.7
Salt solution / pore solution ratio	**1.20**

Material property insights

Certain material properties and conditions can be estimated from the mix formulations (Table 1), normative mineral compositions (Tables 8 through 11 and Tables 13 and 14), and recommended porosity (Table 12). Values based on Tables 8 and 9 are presented in Table 20. A lower bound on dry bulk density is the density of the unreacted dry ingredients. The best estimate for dry bulk density is the density of the "Total product" (e.g., g/mL-hydrated-solid) minus the density of "Unreacted (pore) water" (e.g., g/mL-hydrated-solid), that is, the density of the dry ingredients plus the fraction of the water that reacts. Initial water saturation, not including any dissolved solids, is the volume of unreacted water divided by the pore volume.

Table 20. Calculated material properties and conditions.

Calculated Value	45/45/10 Saltstone	60/40/0 Saltstone	SDU 2/6/7 Concrete	Mix 3B Concrete
Density of unreacted dry ingredients (g/mL) †	0.935	0.937	2.11	2.12
Recommended porosity from Table 12	0.656	0.656	0.110	0.129
Complete saturation				
Dry bulk density, ρ_b (g/mL)	1.12	1.15	2.20	2.23
Volume of unreacted water (mL-water/mL)	0.394	0.358	0.066	0.055
Water saturation (mL-water/mL-void) ‡	0.60	0.55	0.60	0.43
Partial saturation				
Dry bulk density, ρ_b (g/mL)	1.06	1.07	2.17	2.21
Volume of unreacted water (mL-water/mL)	0.453	0.444	0.088	0.075
Water saturation (mL-water/mL-void) ‡	0.69	0.68	0.80	0.58

† lower bound on dry bulk density

‡ not including any dissolved solids (salt)

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