Calculated-Measured Log(η [Pa-s])

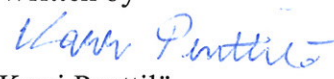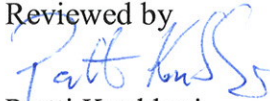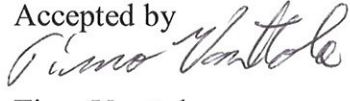# Molten corium and concrete thermodynamics and viscosity

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Confidentiality: Public

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Molten corium and concrete thermodynamics and viscosity		
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Summary

NUCLEA database (**TDNucl/2005**) for in- and ex-vessel applications containing 18 + 2 elements was acquired for FactSage dabase program. This database is same as the one used in **GEMINI2** program from **ThermoData**. It covers the entire field from metal to oxide domains. It allows the user to calculate the thermochemical equilibrium state at any step of a severe accident and to use the results of the thermodynamic approach for improving the predictions of thermo-hydraulic or other accidents codes. A thermodynamic system of selected elements and phases was created from this database for calculations with **ChemSheet** tool in Excel. First melting temperatures of different conctretes were calculated (from **HECLA** project experiments). Then melting behaviour of corium with different amount of steel and concrete were calculated at different temperatures. In the reactor pressure vessel the molten corium forms two immiscible liquid phases, the lighter oxide rich phase is formed on top and the heavier metal-rich phase on bottom. Then these hot molten phases will penetrate the steel wall of the reactor vessel and flow to concrete cavity causing conctrete ablation. **CSFoam** tool was used to calculate the viscosity of this mixture at various temperatures and compositions. **CSFoam** tool contains correlations to calculate the viscosity, the surface tension and the density of molten oxide system. The calculation of the viscosity is based on the bonding state of oxygen in molten silicate and the flow mechanism of melts with a network structure. The needed viscosity parameters for uranium oxide (UO_2) and zirconium oxide (ZrO_2) were assessed and added to **CSFoam** tool.

Confidentiality	Public

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Contents

1   Introduction

In a severe accident in a nuclear power plant, the core of the reactor melts forming corium, also called lava-like fuel containing material (LFCM). It consists of nuclear fuel, fission products, control rods, structural materials from the affected parts of the reactor, products of their chemical reaction with air, water and steam. If the molten corium cannot be cooled inside the reactor, it will penetrate the reactor pressure vessel and flow into the concrete cavity under the reactor. This causes a molten core – concrete interaction (MCCI). The concrete in the floor and sidewalls of the cavity starts to melt. The process is driven by the high initial temperature of the molten corium and the decay heat that is generated inside the melt by the radioactive decay of the fission products.

The phase composition of the corium at varying temperatures and amounts of steel and concrete can be studied with suitable thermodynamic databases and calculation tools. Since 1990 some people were interested in the assessment of thermodynamical data for a number of compounds of reactor materials and fission products based on the recommendations of a specialists meeting held at JRC-Ispra, Italy in 1990 [1]. Critical assessments have been made on a very large number of compounds and systems. **NUCLEA** is a Thermodynamic Database built for collecting all this knowledge. NUCLEA database is available from **ThermoData** in France and it is used in **GEMINI2** program. NUCLEA database is also available for **FactSag**e version 6.2 database program (**TDNucl**/2005). **FactSage** can be used to create a thermodynamic system of selected elements and phases that can be saved as a data-file, and this can then be used with **ChemSheet** tool in Excel. In this project the thermodynamic calculations of corium system were done with **ChemSheet** version 1.84.

2   Methods

2.1   Factsage

FactSage® was introduced in 2001 as the fusion of the **F*A*C*T** / **FACT-Win** and **ChemSage** thermochemical packages (formerly **SOLGASMIX**). **F*A*C*T** - Facility for the Analysis of Chemical Thermodynamics - started in 1976 as a joint research project between two universities: McGill University (W.T. Thompson) and the École Polytechnique de Montréal (C.W. Bale and A.D. Pelton). The **Windows®** version **FACT-Win** was released in 1999 and offered a fully integrated thermochemical database system that coupled proven software with self-consistent critically assessed thermodynamic data. By this time **F*A*C*T** had expanded well beyond chemical metallurgy and was being employed in other fields of chemical thermodynamics by pyrometallurgists, hydrometallurgists, chemical engineers, corrosion engineers, inorganic chemists, geochemists, ceramists, electrochemists, environmentalists, etc. See Figure 1 for **FactSage** main dialog window.
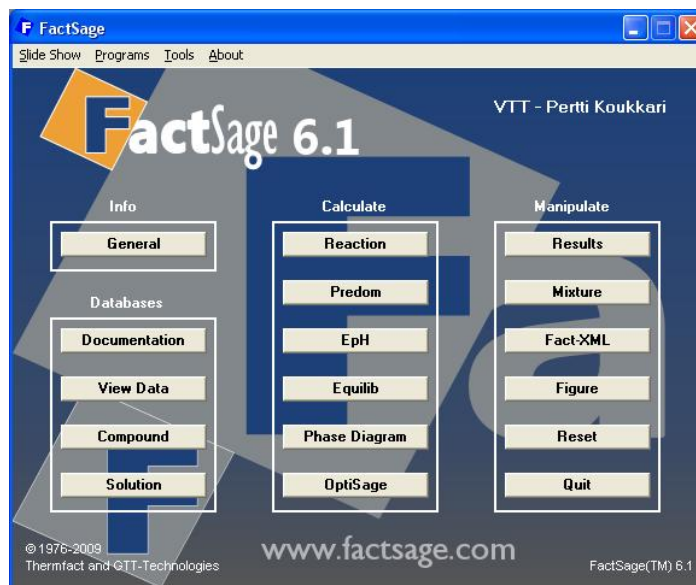
Figure 1. FactSage main dialog window.

In **FactSag**e there are two types of thermochemical databases – compound (pure substances) databases and solution databases. The **View Data**, **Compound** and **Solution** modules permit one to list and manipulate the database files.

Compound databases are for stoichiometric solid, liquid and gaseous species such as CaO(s), NaCl(l), SO_2(g). Compound data may include allotropes, for example graphite C(s1) and diamond C(s2), and isomers, for example ethylene C_2H_4O(liq1) and acetylene C_2H_4O (liq2). Depending upon the type of phase (solid, liquid, gas) and data availability, the stored properties include H^o(298.15 K), S^o(298.15 K), $C_p(T)$, magnetic data (Curie or Néel temperature and average magnetic moment per atom), molar volumes (298.15 K) coupled with expansivities, compressibilities and pressure derivatives of bulk moduli as functions of T. Infinitely dilute aqueous solution data and non-ideal gas are also stored in the compound databases, as are bibliographical references.

Solution databases are for solid and liquid alloys, carbides, nitrides and carbonitrides, concentrated aqueous solutions, ceramics, salts, mattes, slags, etc. The data are stored in the form of Gibbs energy functions for the phase constituents and temperature dependent model parameters for calculating the Gibbs energy of mixing among the phase constituents. **FactSage** supports more than 10 different solution models including simple polynomial models (Redlich-Kister and Legendre polynomials) combined with different higher order extrapolations (Muggianu, Kohler, Toop), the Unified Interaction Parameter Model, Modified Quasichemical models for short-range ordering in pair and quadruplet approximations, the Pitzer model (for concentrated aqueous solutions) and sublattice models such as the Compound Energy Formalism. Additional solution models are constantly planned.

TDNucl Thermodata nuclear database is a database for in- and ex-vessel applications containing 18 + 2 elements : O, U, Zr, Ag, In, B, C, Fe, Cr, Ni, Ba, La, Sr, Ru, Al, Ca, Mg, Si, Ar, H and including the 15 oxide system: UO_2, ZrO_2, In_2O_3, B_2O_3, FeO, Fe_2O_3, Cr_2O_3, NiO, BaO, La_2O_3, SrO, Al_2O_3, CaO, MgO, SiO_2. It covers the entire field from metal to oxide domains. It allows the user to

calculate the thermochemical equilibrium state at any step of a severe accident and to use the results of the thermodynamic approach for improving the predictions of thermo-hydraulic or other accidents codes.

Figure 2, Figure 3, Figure 4, Figure 5, Figure 6 and Figure 7 shows calculated phase diagrams of some selected binary (and ternary) subsystems of **NUCLEA** database. In general these subsystems are combined to describe a multicomponent system of phases, with interactions between the components in each mixture phase. These interactions are described with a suitable thermodynamic phase model and the model parameters are fitted from the measured data.

In binary phase diagrams the temperature is given on the Y-axis and the molar fraction of the second component on the X-axis (so called T-x phase diagram). The lines in the diagram are the phase boundaries, i.e. temperatures with given composition at which a phase is formed. In each of these binary phase diagram the phase that exists at the highest temperature region is the liquid phase.

When the temperature is decreased the liquid phase starts to solidify. Liquid mixture phase does not solidify at a fixed temperature, but there is so called mushy zone where the liquid phase and the solid phase coexist – in a binary system (two independent system components) there can co-exist max. two phases at fixed temperature and composition. This zone is quite narrow in UO_2-ZrO_2 subsystem (Figure 2) but quite wide in SiO_2-ZrO_2 subsystem (Figure 4). Also liquid phase may separate into two immiscible liquid phases (so called miscibility gap), like in SiO_2-ZrO_2 subsystem (Figure 4). The temperature where the liquid phase starts to solidify is called liquidus temperature, and the temperature where all the liquid phase has solidified is called solidus temperature. The composition which has the lowest solidus temperature is called the eutectic point. The eutectic point for FeO-ZrO_2 (Figure 7) is 1600 K and ~7 mole-% ZrO_2. Even though it appears that at this point the whole liquid phase is solidified at a fixed temperature, there is still very narrow temperature range where it takes place (but this is not visible from the diagram).
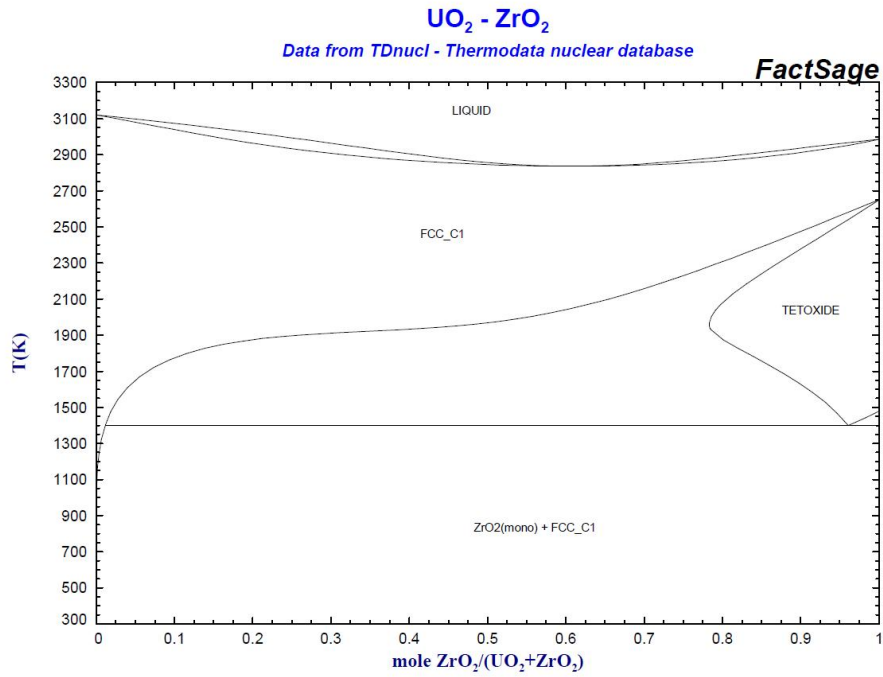
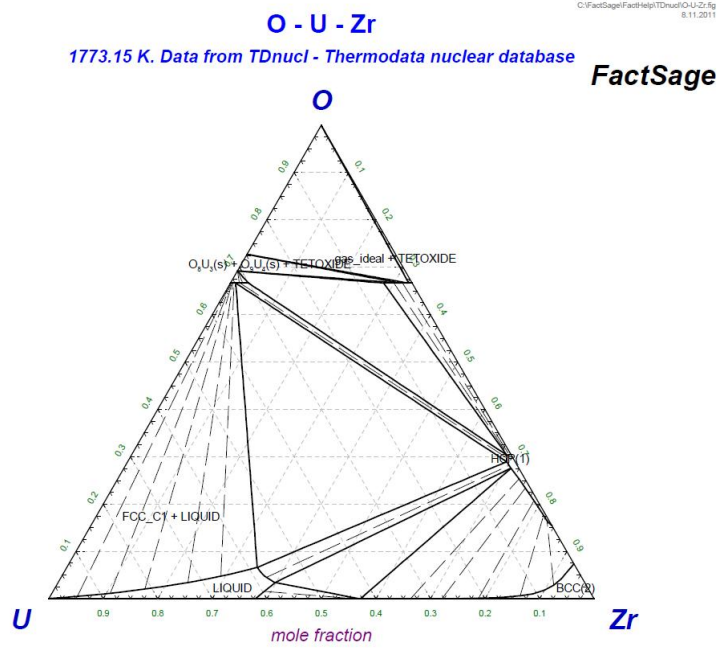*Figure 2. UO₂-ZrO₂ subsystem – binary phase diagram.*

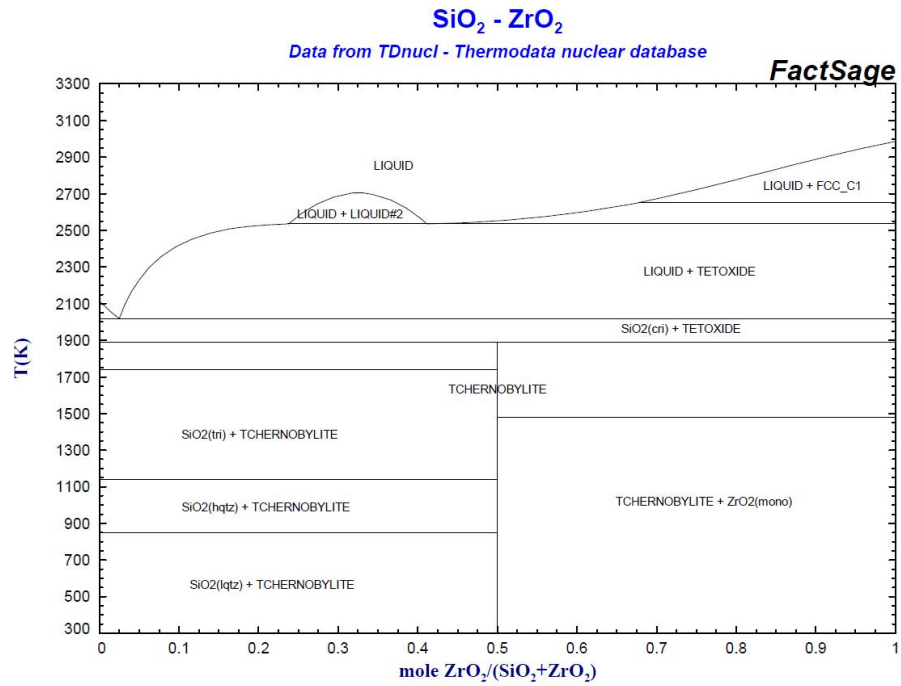*Figure 3. UO₂-ZrO₂ subsystem – ternary phase diagram.*
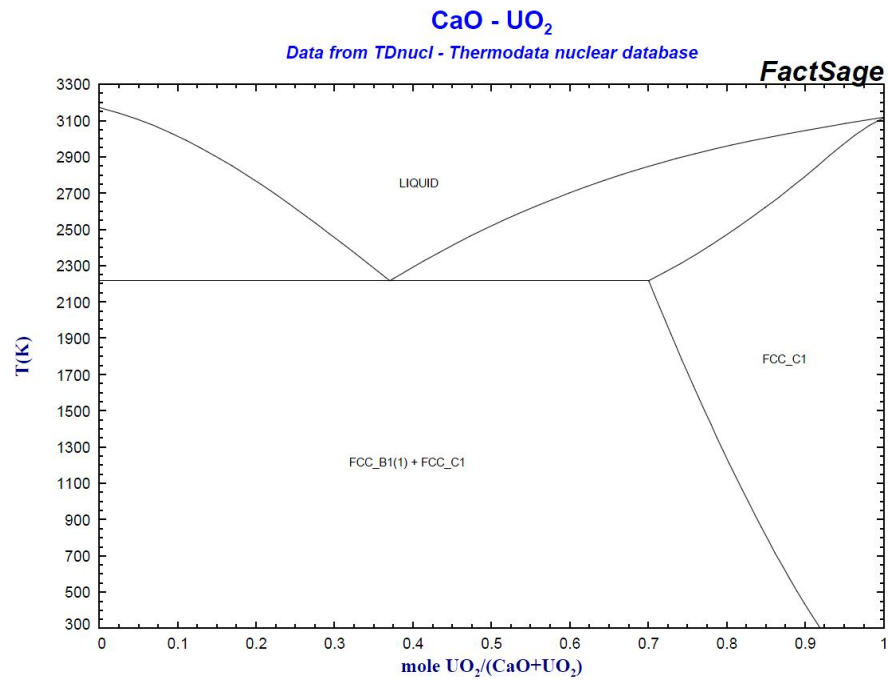
Figure 4. SiO₂-ZrO₂ subsystem – binary phase diagram.

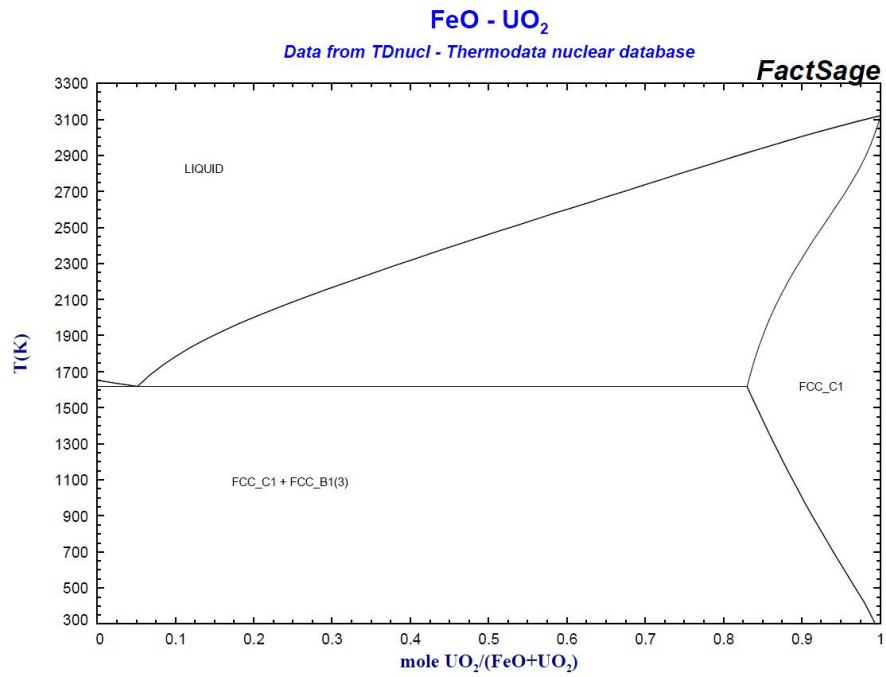*Figure 5. CaO-UO₂ subsystem – binary phase diagram.*
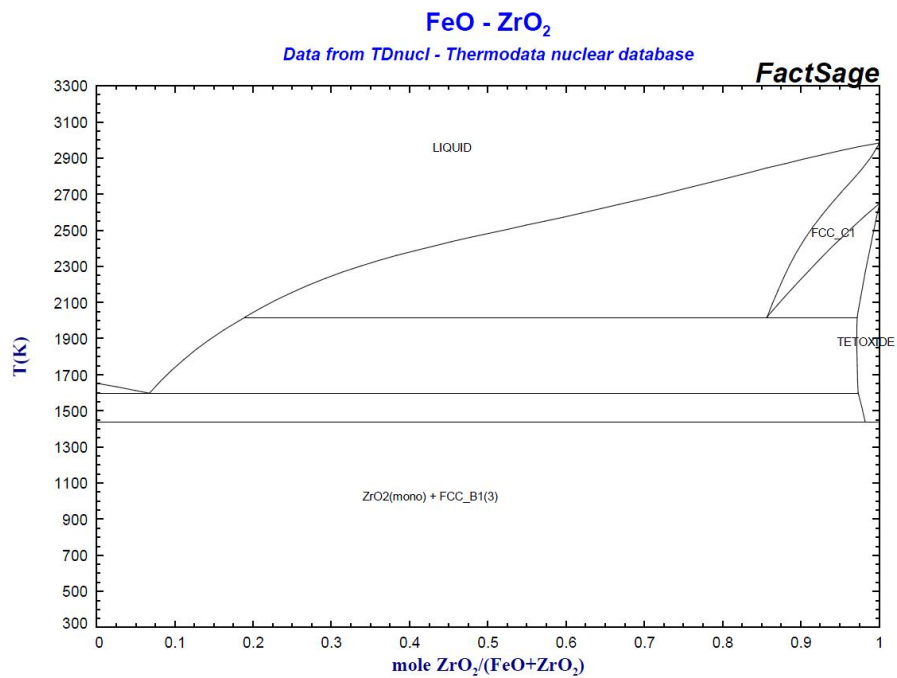
Figure 6. FeO-UO$_2$ subsystem – binary phase diagram.

Figure 7. FeO-ZrO$_2$ subsystem – binary phase diagram.

2.2   ChemSheet

Chemsheet works as an add-in program of general thermodynamics in Excel [2] (see Figure 8). The thermochemical programming library **ChemApp** is used in combination with its application-specific thermochemical data. The non-ideal solution models cover concentrated aqueous solutions, dilute and concentrated alloys, liquid slags and molten salts, solid solutions, non-ideal gases and non-stoichiometric systems. **ChemSheet** is straightforward and easy to use and requires no programming skills other than normal Excel use. To the user, the process model can be just one Excel-file.
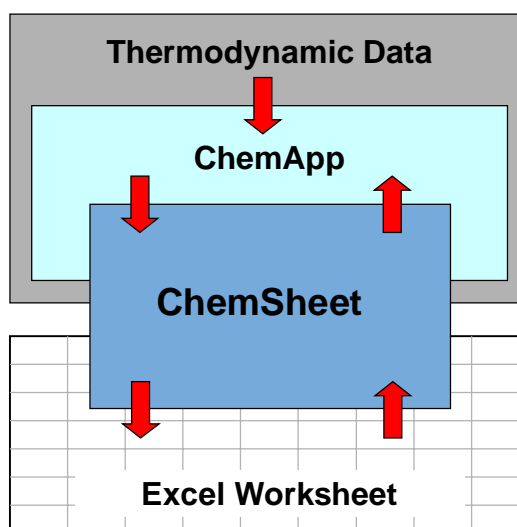

Figure 8 ChemSheet Interface.

In **ChemSheet** as in **ChemApp** there are two different calculation methods: using global conditions of the System, or using Streams. Using Global conditions, user merely needs to set single values for temperature and pressure, and enter incoming constituents to define the initial composition of the system. A stream is a means for transferring non-reacted matter to a reaction zone. It has constant temperature and pressure, and contains one or more constituents. When using Streams, you set the three variables: temperature, pressure and composition for each input stream and set single conditions for temperature and pressure of the system

ChemSheet can calculate one or more equilibrium steps. For each step user needs to define the initial conditions, either global or stream conditions (equilibrium temperature, pressure and the initial composition, and same for any incoming streams, its temperature, pressure and composition). **ChemSheet** contains one variable (worksheet name variable), *StepIndex*, that can be used in formulas so that values of these conditions can vary between the steps (for example to change the equilibrium temperature between the steps).

In this project **ChemSheet** is used for the thermodynamic calculation of the corium system. The calculated results (like composition of the molten oxide phase) are stored directly to Excel worksheet cells and then it easy to use them as input for the calculation of the viscosity model, as **CSFoam** tool used for it is also implemented as an Excel add-in.

2.3 Thermodynamic system

Thermodynamic system for **ChemSheet** can be generated in **FactSage** and then exported as a thermodynamic file (so called transparent data-file, which is in binary, encrypted formed, if it contains any solution data). Figure 9 shows a selection of the solution/mixture phases that are found from the **NUCLEA** database for the given reactants. These reactants include all the components that are needed for the calculation of the corium system.
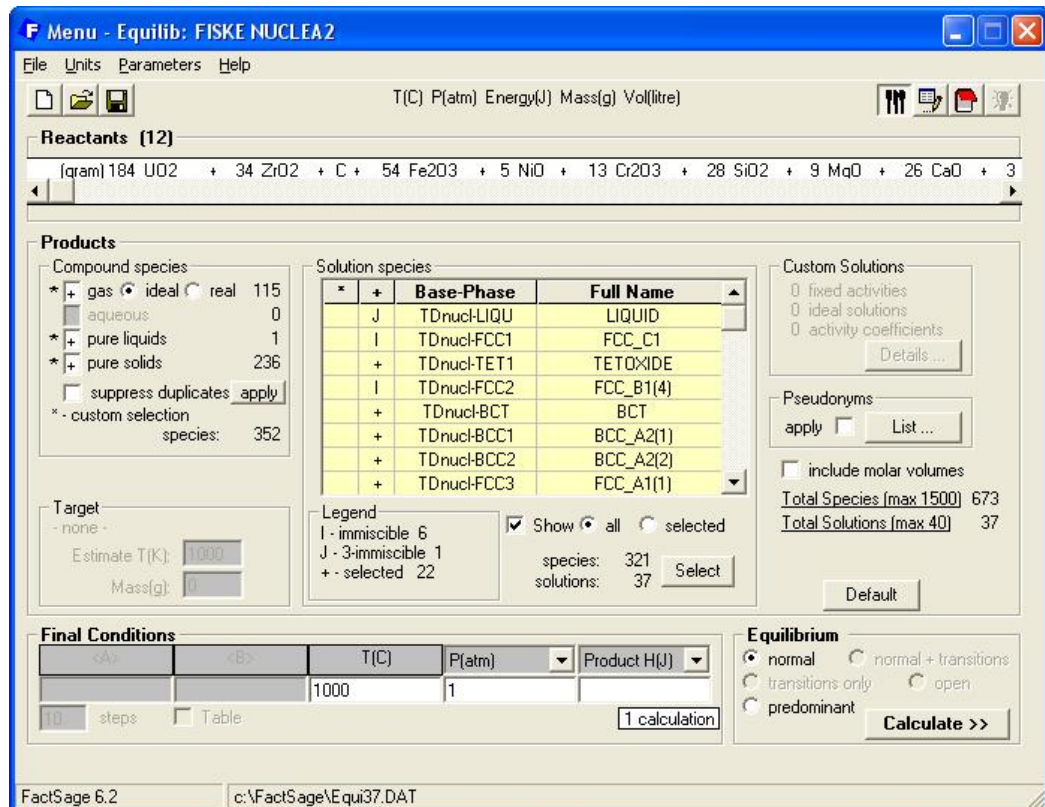

Figure 9. Selected NUCLEA system in FactSage.

The included elements are: C, H, N, O, Al, Ca, Cr, Fe, Mg, Ni, Si, U, Zr. All together there 115 gas phase species, 37 solution phases containing 321 species, one pure liquid phase (water) and 236 pure solid phases (mainly pure metals and oxides). The most important solution phases is the LIQUID phase, it contains 26 species, metals and oxides. It describes the molten corium. There are two instances of this LIQUID phase in the thermodynamic system (LIQUID#1 and LIQUID#2) as the molten metal/oxide system can separate into two immiscible phases, the metal-rich and the oxide rich phase. This means that these two phases ant their species have same thermodynamic parameters (same reference states and interaction energies) but their phase compositions are different. The species in the LIQUID phase are (as named in the NUCLEA database): Al1, (Al2O3):2.000, (Al4O12Si3):3.000, (Al2CaO8Si2):2.000, C1, Ca1, Ca1O1, Ca1O3Si1, Ca2O4Si1, Cr1, (Cr2O3):2.000, Fe1, Fe1O1, (Fe2O3):2.000, Mg1, Mg1O1, Mg2O4Si1, Ni1, Ni1O1, O1, O2Si1, O2U1, O2Zr1, Si1, U1, Zr1. For example (Al4O12Si3):3.000 species corresponds to $2/3Al_2O_3 \cdot SiO_2$ species which is an aluminium silicate (stoichiometries are scaled so that there is max. one mole of any metal in a species).

2.4 CSFoam

CSFoam tool is been developed in ELEMET/EffArc project for the calculation of physical properties of oxide slags in steel making. For example in the case of electric arc furnace it is used for the calculation of the slag foaming which is dependent on the slag viscosity, surface tension and density. For this project new components were added into it in order to calculate the viscosity and the density of the molten corium with concrete components.

CSFoam tool is an Excel add-in like **ChemSheet**. It contains internal database for selected components and three functions to calculate the viscosity, the surface tension, and the density. The interfaces of these routines are shown in Table 1.

Table 1. CSFoam routines with input parameters.

Property	Param1	Param2	Param3	Param4	Param5
VISCOSITY	TEMPERATURE	COMPOSITION	NAMES	COMP_UNIT	PROP_UNIT
TENSION	TEMPERATURE	COMPOSITION	NAMES	COMP_UNIT	PROP_UNIT
DENSITY	TEMPERATURE	COMPOSITION	NAMES	COMP_UNIT	PROP_UNIT

Figure 10 shows how a property value (in this case density) can be calculated in Excel worksheet.

Figure 10. Using a function in worksheet to calculate the density of molten oxide.

2.5   Viscosity model

Assuming that the structure of aluminosilicate melts consists of only three kinds of chemical bonds, i.e. Si–BO (bridging oxygen in Si tetrahedral unit), Al–BO (bridging oxygen in Al tetrahedral unit with a charge-compensating cation), and Si–NBO (NBO: non-bridging oxygen connected with Si) with a charge-compensating cation (see Figure 11), it is possible to calculate the bonding states of oxygen in molten aluminosilicate by the chemical composition of slag without any thermodynamic parameters. Owing to a simple method, the evaluation of the bonding states of oxygen can be conducted in various kinds of silicate systems including alkali oxides as well as alkaline earth oxides. Silicate slag has a network structure made of bonding SiO_4^{4-} units and Si tetrahedral ions. Si tetrahedral ions are combined through bridging oxygen. The non-bridging oxygen ion O^- and the free oxygen ion O^{2-} are generated by partially cutting off the network structure of Si tetrahedral when adding some basic oxides such as CaO and Na_2O in silicate slag. The non-bridging oxygen ion and the free oxygen ion have a larger mobility than the bridging oxygen in the network structure because there are "cutting-off" points. It is assumed that the movements of "cutting-off" points cause a viscous flow that lowers the activation energy of viscosity [3].
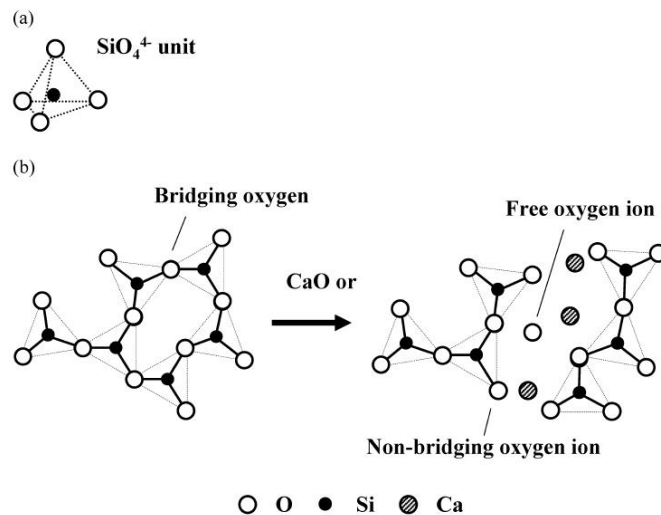
Figure 11. Structure of silicate slag.

Originally this method was applied to system of SiO_2–CaO–MgO–FeO–K_2O–Na_2O–Al_2O_3 system describing aluminosilicate melts containing alkali oxides in melting furnaces. Since then CrO, Cr_2O_3, Fe_2O_3, NiO, TiO_2, UO_2 and ZrO_2 have been added.

The viscosity on liquid oxide is calculated as:

$$\eta = A \cdot \exp\left(\frac{E_v}{R \cdot T}\right) \qquad (1)$$

where A is a constant (4.80×10^{-8}), E_v is the activation energy, R is the gas constant (8.314), and T is the temperature.

The activation energy is calculated as:

$$E_v = \frac{E}{1+ \sqrt{\sum_i \alpha_i \cdot N_{(NBO+FO)_i} + \sum_j \alpha_{j\,in\,Al} \cdot N_{(Al-BO)_j}}} \qquad (2)$$

where E is the activation energy of pure SiO_2 (5.21×10^5), α_i and α_j are model parameters in Table 2, $N_{(NBO+FO)}$ of the sum of the fractions of non-bridging oxygen ion (NBO) and free oxygen ion (FO), $N_{(Al+BO)}$ the fraction of the bridging oxygen (BO) in the Al tetrahedral unit, i is the component of melt except SiO_2, i.e. CaO, MgO FeO, K_2O, Na_2O, Al_2O_3, and j is the charge-compensating ion from component i except Al_2O_3, i.e. Ca^{2+}, Mg^{2+}, Fe^{2+}, K^+ and Na^+.

Table 2. Component parameters in the viscosity model.

M_xO_y	α_i	j	$\alpha_{j\,in\,Al}$
CaO	4	Ca2+	1.46
MgO	3.43	Mg2+	1.56
FeO	6.05	Fe2+	3.15
K2O	6.25	K+	-0.69
Na2O	7.35	Na+	0.27
CrO	3	Cr2+	0.6
Cr2O3	4.7	Cr3+	5
NiO	6.05	Ni2+	3.15
TiO2	4	Ti4+	1.46
Fe2O3	6.05	Fe3+	3.15
UO2	0.5	U4+	3
ZrO2	0.65	Zr4+	5
Al2O3	1.14		

Slag system is divided into three groups of species, a) SiO_2, b) M_xO_y (CaO, MgO, FeO, K_2O, Na_2O, ...) and c) Al_2O_3. Then relative mole numbers for these groups are calculated so that:

$$a + \sum_i b_i + c = 1 \qquad (3)$$

Sum of fractions $N_{(NBO+FO)i}$ and $N_{(Al+BO)j}$ are calculated as:

$$N_{(Al-BO)_j} = \frac{n_{(O-Al\,in\,Al)_j}}{n_{Total-O}} = 4c\left(b_i \Big/ \sum_i b_i\right) \Big/ \left(2a + \sum_i b_i + 3c\right) \qquad \sum_i b_i \geq c$$

$$N_{(Al-BO)_j} = \frac{n_{(O-Al\,in\,Al)_j}}{n_{Total-O}} = 4b_i \Big/ \left(2a + \sum_i b_i + 3c\right) \qquad \sum_i b_i < c \qquad (4)$$

$$N_{(NBO-BO)_i} = \frac{n_{(O-M)_i}}{n_{Total-O}} = \left\{b_i - c\left(b_i \Big/ \sum_i b_i\right)\right\} \Big/ \left(2a + \sum_i b_i + 3c\right) \qquad \sum_i b_i \geq c$$

$$N_{(NBO-BO)_i} = \frac{n_{(O-M)_i}}{n_{Total-O}} = 3\left(c - \sum_i b_i\right) \Big/ \left(2a + \sum_i b_i + 3c\right) \qquad \sum_i b_i < c \qquad (5)$$

The volume fraction of precipitated solids affects the effective viscosity of the slag. Mostafaee [4] gives following correlation for the effective viscosity:

$$\mu = \mu_0 \left(1 + 5.5V_f\right) \qquad (6)$$

where μ_0 is the viscosity of the slag phase and V_f is the volume fraction of solids in the system.

Viscosity model does not require SiO_2 or Al_2O_3 to be present. For example if they both are zero then $N_{(NBO+FO)i}$ is simplified to mole fraction of B_i.

Viscosity model (without UO_2 and ZrO_2) has been validated with chromium and non-chromium containing slags for steel making. Figure 12 shows the correlation between measure and calculated viscosities.
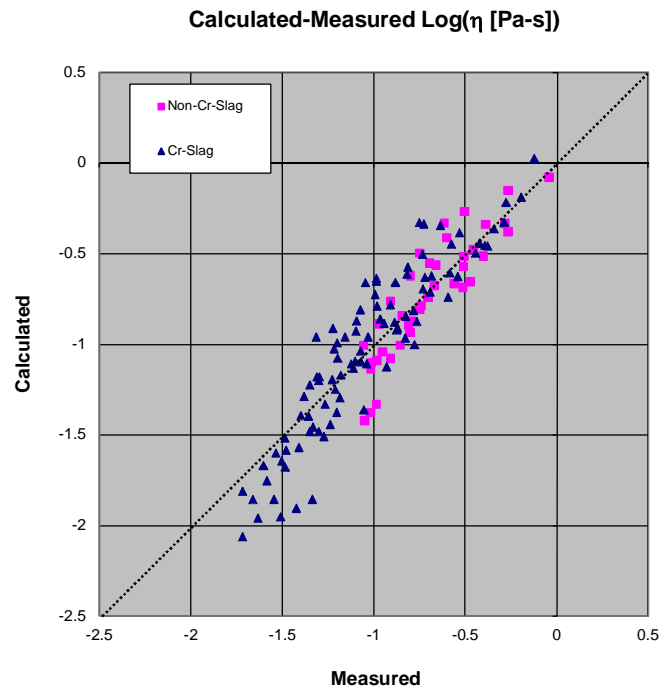

Figure 12. Correlation between measured and calculated viscosity data.

3 Results

3.1 HECLA experiments

ChemSheet model was made to calculate the **HECLA** experiments with **NUCLEA** system. Table 3 shows the input compositions taken from **HELCA** report [5].

Table 3. Chemical compositions of HECLA experiments used with ChemSheet.

Compound	Mass/kg			
	HECLA2	HECLA3	HECLA4	HECLA5
SiO_2	57	38	52.5	44.4
Fe_2O_3	3.54	34.7	31.7	32.1
FeO	0	0.66	0.1	0.2
CaO	10.2	6.4	6.1	9.8
$CaCO_3$	2.32	1.9	2.84	3.04
$Ca(OH)_2$	1.6	3	1.1	1.2
Al_2O_3	12.6	7.04	1.9	3.2
MgO	1.89	1.15	0.27	0.33
H_2O	3.4	3.27	2.46	4.09

Note that of the main components SO_3, K_2O, Na_2O and TiO_2 are not included as elements S, K, Na and Ti were not present in the thermodynamic system. Especially K_2O and Na_2O might affect the melting temperature.
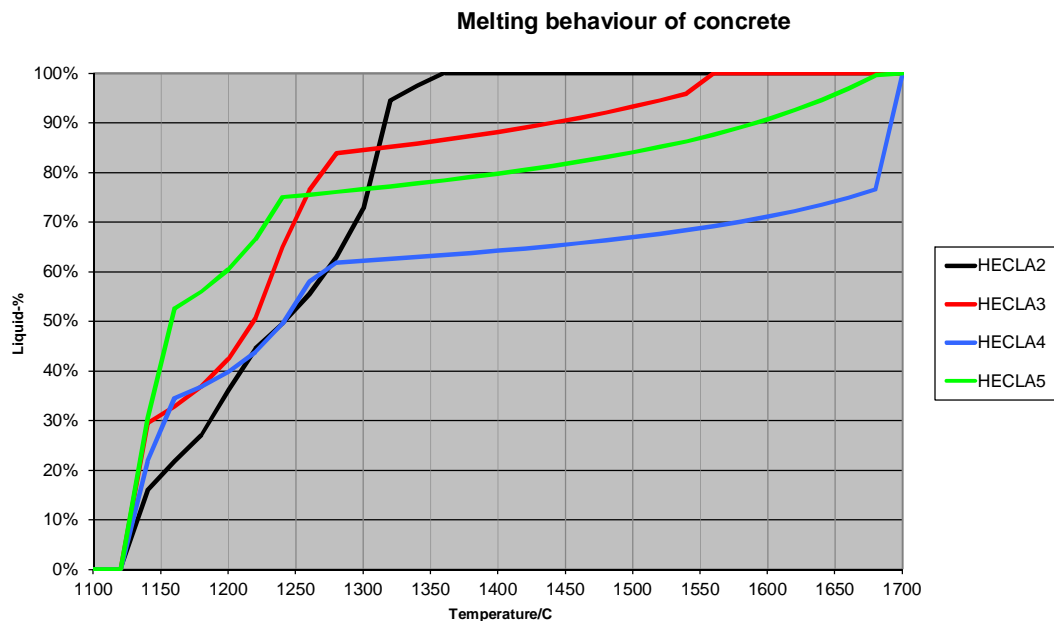

Figure 13. Melting behavior of the concrete, calculated with ChemSheet.

Table 4 shows the melting temperatures for each experiment.

Table 4. Melting temperatures of the concretes, calculated with ChemSheet. The ablation temperature corresponds to the point where 50 % of the mass is molten.

| Experiment | Temperature/C | | |
	Solidus	Ablation	Liquidus
HECLA2	1120	1240	1360
HECLA3	1120	1220	1560
HECLA4	1120	1240	1700
HECLA5	1120	1160	1680

There are some differences between these results and those that were calculated with **GEMINI2** program, especially for liquidus temperature. The NUCLEA database used with **ChemSheet** was older on so that might have affected the results (from 2005 as the one used with **GEMINI2** was from 2009).

Figure 14 shows the calculated viscosities for HECLA experiments. These have been calculated for the LIQUID#1 phase composition that was earlier calculated with **ChemSheet**. In most cases LIQUID#2 phase also existed at equilibrium but its mass fraction was much less than that of LIQUID#1. Viscosities were calculated so that the volume fraction of solid phases was taken into account (although in this case it was probably not needed as the concrete that was still solid didn't exist as precipitated particles)
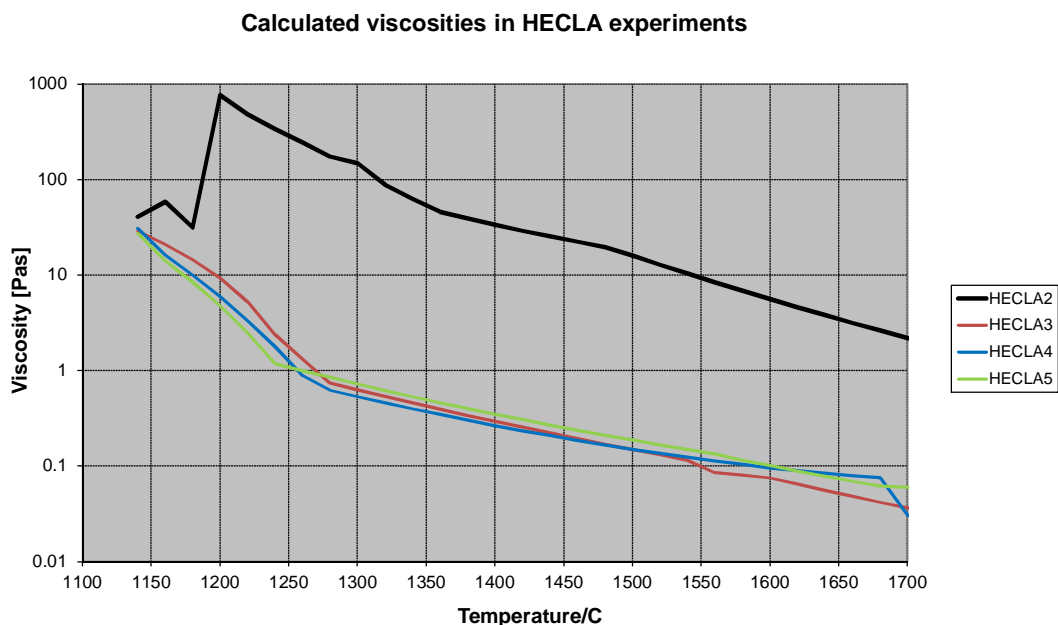

Figure 14. Viscosities in HECLA experiments: calculated with CSFoam using the composition of LIQUID#1 phase calculated by ChemSheet.

Viscosity for HECLA2 experiment shows very high values compared to others. It has the highest SiO_2 and lowest Fe_2O_3 content and the viscosity is very dependent especially on the silica content, but it still might be out of valid range of the model and in that case the result is not reliable.

3.2 Corium

Theofanous et al. [6] give following correlation for ZrO_2:

$$\eta = 1.5 \times 10^{-4} \exp\left(\frac{10430}{T}\right)$$ (7)

and for UO_2:

$$\eta = 1.61 \times 10^{-4} \cdot \exp\left(\frac{11130}{T}\right)$$ (8)

Figure 15 shows the viscosities of pure ZrO_2 and UO_2 calculated with these correlations and with CSFoam. Fitted α parameters for ZrO_2 and UO_2 components are shown in Table 2.
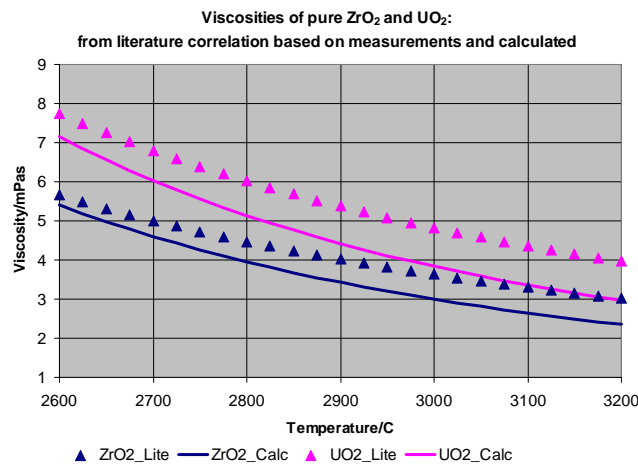

Figure 15. Viscosities of pure ZrO₂ and UO₂: from literature correlation based on measurements and calculated with CSFoam.

Sudreau and Cognat [7] have modelled the viscosity of corium above its liquidus temperature. They show also measured viscosities for corium with 77.8 m-% UO_2 and 22.2 m-% ZrO_2. Figure 16 shows the measured and calculated viscosities (with CSFoam) for corium.
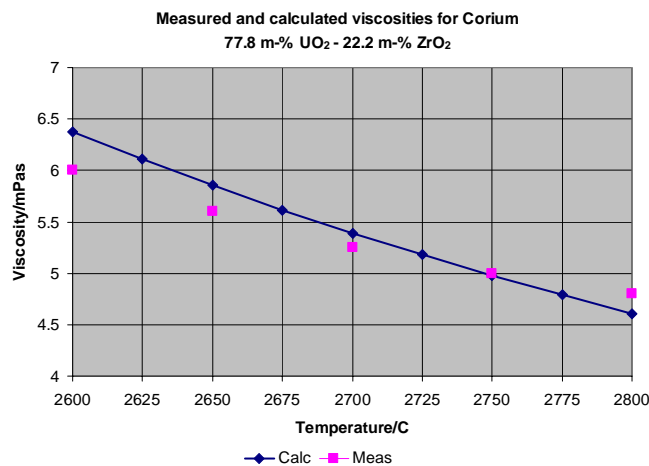

Figure 16. Measured and calculated viscosities for corium.

Equilibrium calculations were made for system containing UO_2, ZrO_2 and concrete, which had the same composition as in HECLA3 experiment. Concrete mass fraction was varied from 0 to 50 mass-%. Mass ratio between UO_2 and ZrO_2 was kept constant (78 m-% : 22 m-%). Table 5 shows the used absolute masses.

Table 5. Masses of Concrete, UO_2 and ZrO_2.

Conc/kg	UO2/kg	ZrO2/kg
0	78	22
5	74.1	20.9
10	70.2	19.8
15	66.3	18.7
20	62.4	17.6
25	58.5	16.5
30	54.6	15.4
35	50.7	14.3
40	46.8	13.2
45	42.9	12.1
50	39	11

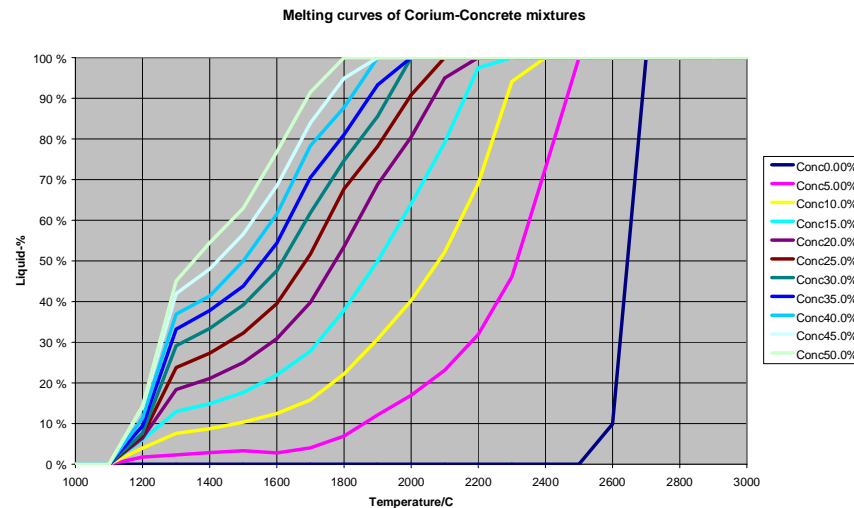Figure 17 shows the calculated melting curves of corium-concrete mixture.

Figure 17. Melting curves of Corium-Concrete mixture.

Figure 18 shows the calculated viscosity curves of corium-concrete mixture.
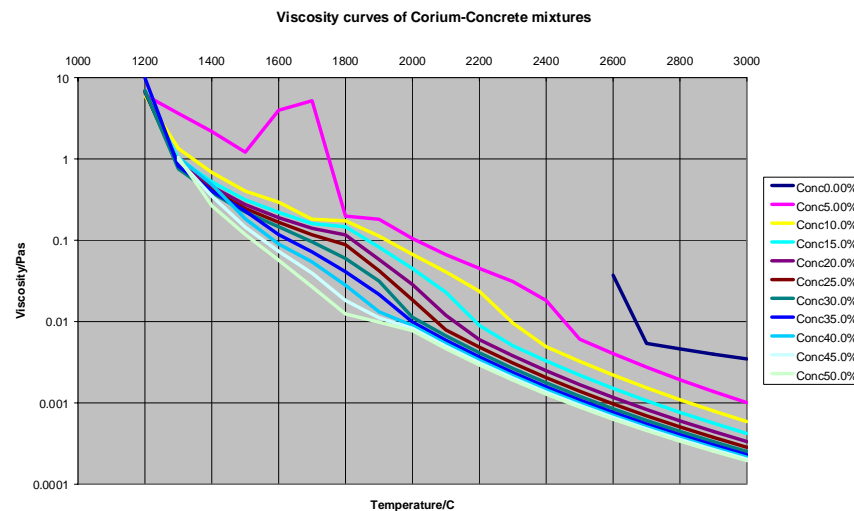

Figure 18. Viscosity curves of Corium-Concrete mixture.

4   Conclusions

ChemSheet with **NUCLEA** database and **CSFoam** are very efficient tools to calculate and study different severe accident scenarios that involve the melting of the core and the formation of different partly molten oxide and metals phases.

5   Summary

NUCLEA database (**TDNucl/2005**) for in- and ex-vessel applications containing 18 + 2 elements was acquired for FactSage database program. This database is same as the one used in **GEMINI2** program from **ThermoData**. It covers the entire field from metal to oxide domains. It allows the user to calculate the thermochemical equilibrium state at any step of a severe accident and to use the results of the thermodynamic approach for improving the predictions of thermo-hydraulic or other accidents codes. A thermodynamic system of selected elements and phases was created from this database for calculations with **ChemSheet** tool in Excel. First melting temperatures of different concretes were calculated (from **HECLA** project experiments). Then melting behaviour of corium with different amount of steel and concrete were calculated at different temperatures. In the reactor pressure vessel the molten corium forms two immiscible liquid phases, the lighter oxide rich phase is formed on top and the heavier metal-rich phase on bottom. Then these hot molten phases will penetrate the steel wall of the reactor vessel and flow to concrete cavity causing concrete ablation. **CSFoam** tool was used to calculate the viscosity of this mixture at various temperatures and compositions. **CSFoam** tool contains correlations to calculate the viscosity, the surface tension and the density of molten oxide system. The calculation of the viscosity is based on the bonding state of oxygen in molten silicate and the flow mechanism of melts with a network structure. The needed viscosity parameters for uranium oxide (UO_2) and zirconium oxide (ZrO_2) were assessed and added to **CSFoam** tool.

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