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Thermochemical data and modelling for ex-vessel corium behaviour during a severe accident

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Abstract

A project on thermochemical data and modelling of ex-vessel corium behaviour has been conducted as part of the European Commission Fourth Framework Programme on Nuclear Safety. The objective of the study was to provide critically assessed thermochemical data and develop calculational methods for use in assessment studies of the consequences of melt–basemat interactions. The work programme comprised the critical assessment and experimental determination of thermochemical data for species of key importance to melt interactions. A database has also been developed to model the multi-component system and calculations performed to predict the melt composition. Experimental determinations for silica and zirconia-based systems, for example measurements of the chemical activities of Ce and La in melts, have been performed to assist in the validation of the models. The sensitivity in the predictions of the effects of ex-vessel interactions to factors such as accident conditions (e.g., temperature, concrete type) and, uncertainties in the thermochemical and kinetic data has also been assessed.

1. Introduction

The ex-primary vessel progression of a severe accident in a light water reactor involves the molten core debris being ejected onto the reactor basemat (either concrete or protective material). Although some freezing of the melt could occur, particularly if a water pool is present initially, at some stage molten core debris would interact thermally with the basemat resulting in ablation and the release of species by vapourisation. The composition of the melt would change progressively as the basemat decomposition products are subsumed during the interaction. The major issues for consequence analysis of basemat attack are:

- Melt coolability and extent of basemat ablation; threat to containment integrity.
- The extent of fission product and fuel/actinide release to the containment; the releases of low-volatile species would add to the radiological risk.
- The production of CO and H_2; influence on speciation of fission products and risk of deflagration.
- Influence of non-radioactive aerosols from the interaction in mitigating the fission product release from the containment; production of aerosols.
- The relative timing of the containment and basemat failure.

Thermochemical methods, involving models and critically assessed thermodynamic data, play an increasing role in predicting fission product release and source term applications. The models are especially important in high temperature scenarios where only few or no experimentally determined release data exist and hence the thermochemical database is a vital part of the process for severe accident applications.

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Therefore a three year project on thermochemical data and modelling for ex-vessel corium behaviour and coolability (THMO) involving three European organisations was conducted. The work was part of the European Commission (EC) Fourth Framework Programme on Nuclear Safety and the objective of the study was to provide critically assessed thermochemical data and develop calculational methods for use in assessment studies of the consequences of melt–basemat interactions (MCCIs).

2. Work programme

The work programme for the THMO project was divided into four technical packages. The first involved the critical assessment of thermochemical data for pure substances and gases relevant to melt interactions, followed by the experimental determination of thermochemical parameters that were considered inadequate or were based on estimated data. The second package included the critical assessment of thermochemical data for phase diagrams and the development of models based on the assessed data; the calculation of equilibrium compositions for relevant systems was also included in this task. The third package comprised the validation of the models by experimental determinations for representative systems. The final work package was concerned with analysing the sensitivity in the predictions of the effects of ex-vessel interactions to factors such as accident conditions (e.g., temperature, concrete type, melt composition and stratification) and uncertainties in the thermochemical and kinetic data.

2.1. Task 1 – Experimental determination of thermochemical data

The code comparison exercises performed on the large-scale experiments of the advanced containment experiments (ACE) MCCI programme showed order of magnitude differences between predictions and experiments for the release of some of the fission products [1]. Based on an assessment of available data and the identification of key species, thermochemical parameters were determined for the following:

- Silicates and zirconates of the low volatile fission product elements, such as barium, strontium, lanthanum, and cerium (pure substances).
- Selected binary phase diagrams for systems comprising Fe_2O_3.
- Enthalpies of melting of binary and ternary oxides.
- Gaseous hydroxide species of U, Ba, Ce, La, Ru, Sr and Te.

2.2. Task 2 – Development of models and calculations

The work for this task was concerned with the development of models for the calculation of the chemical behaviour of fission products in the melts and release into the cavity atmosphere. In addition, the models can provide a description of the chemical composition of the oxide and metal phases of the melt which can be used in modelling the melt thermal behaviour. The models are based on data for the binary and ternary sub-systems and consider the phases of importance during the early and late stages of the interaction, that is the metal phase, metal–metal oxide interaction phase and oxide phase. The work required the extention of models currently available, which included

- Addition of Fe_2O_3 and UO_{2+x} to the 11 component oxide database UO_2–ZrO_2–SiO_2–CaO–Al_2O_3–MgO–BaO–SrO–La_2O_3–CeO_2–Ce_2O_3.
- Assembly of a metal database for the system U–Zr–Fe–Si–Sr–Ba–Ce–La–Te–Ru.
- Addition of Fe–FeO to the U–Zr–Si–O database.

2.3. Task 3 – Validation experiments

Experimental data are required to assess the models used for multi-component systems; for example, the activity of a component in the melt or extent of release of species from a simulated core melt at high temperature can be compared to the model predictions. Two sets of experiments have been conducted on silica containing systems to provide such data:

- The measurement of the chemical activities of key fission products in silica containing melts using Knudsen cell mass spectrometry.
- Experiments involving fission product simulants, siliceous material and zirconium or silicon metal to study the releases and potential interactions in the vapour phase between the simulants and SiO(g).

2.4. Task 4 – Sensitivity calculations

The objective of this task was to analyse the sensitivity of the predictions of melt behaviour, including fission product vapourisation, with respect to specific ex-vessel conditions (e.g., partial solidification, changing melt compositions due to dissolved basemat or protective material) and the uncertainties in the thermochemical and kinetic data. The work involved studies of the distribution of heat-generating radionuclides between the metallic and oxide melt phases and the releases of radiologically relevant fission products from melt pools. The work was divided into three parts:

- Assessments of possible melt configurations and corresponding distribution of fission products.
- Modelling of MCCI with thermochemical equilibrium codes.

- Sensitivity studies based on anticipated uncertainties in the thermochemical data.

3. Results

3.1. Experimental determination of thermochemical data

Pure and well-defined samples of the following solid compounds were prepared; $Sr_3MgSi_2O_8$, $BaCeO_3$, $SrCeO_3$, $La_2Zr_2O_7$, $BaZrO_3$, $SrZrO_3$, Ce_2O_3, $SrFe_{12}O_{19}$, $Sr_7Fe_{10}O_{22}$, $Sr_2Fe_2O_5$, $Ba_2Fe_2O_5$, α-$BaFe_2O_4$, $BaFe_{12}O_{19}$ and $SrBaFe_4O_8$. Measurements of the low-temperature heat capacity, enthalpy increment and enthalpy of formation were performed on these samples using calorimetry or EMF cell methods. Data for $C°p$(298.15 K), S°(298.15 K), $\Delta_f H°$(298.15 K) and the variation of H°(T)-H°(298.15 K) with temperature were determined for the compounds.

Phase diagram determinations were also conducted using differential thermal analysis (DTA) for the pseudo-binary systems La_2O_3, SrO and BaO with Fe_2O_3. Experiments were performed using a Bähr 701 DTA apparatus with platinum crucibles as container material in an oxygen/argon atmosphere (20/80 vol.%) to prevent reduction or oxidation of the iron containing compounds. The maximum temperature which was obtained with the DTA furnace was 2015 K.

A review of enthalpy of fusion data for binary and ternary oxides of relevance to MCCIs was conducted and recommended experimental values provided for the binary oxides MgO, CaO, SrO, BaO, Al_2O_3, SiO_2 and ZrO_2, and for the ternary oxides $MgSiO_3$ and $CaSiO_3$. The enthalpies of fusion of the low volatile fission product oxides $SrSiO_3$, $BaSiO_3$, $La_2Si_2O_7$, $Ce_2Si_2O_7$, $BaZrO_3$ and $SrZrO_3$ were estimated. Values for the entropies of fusion of the binary and ternary oxides were also proposed; however, with strong interactions in the liquids leading to stable ionic complexes, such as in the case of SiO_2-rich liquids, much lower values could be found.

The importance of the gaseous hydroxides and oxyhydroxides of reactor materials and fission products in safety analyses has been emphasised in many studies: for many elements they are predicted to be the dominant species over oxides in contact with steam/hydrogen mixtures at high temperatures [2]. A number of these gaseous species are likely to exist, particularly for elements such as ruthenium and the actinides which have a number of valencies even in the solid state. However, there are few experimental data for the thermodynamic stability of these species, and accident analysis has relied heavily on estimates. A review was conducted of the estimation procedures used by Jackson [3] for the stabilities of the mono and di-hydroxides and by Krikorian [4] for the stabilities of the gaseous hydroxides and oxyhydroxides of Y, Zr, Nb, Ru, Rh, Ce, Pr, Eu, U, Pu, Am and Cm. Revised estimated data for these species were determined.

Gas phase transpiration experiments were conducted on samples of La_2O_3, RuO_2, CeO_2 and UO_2 at temperatures up to 2248 K. Carrier gas of either Ar/steam or Ar/O_2 of the same controlled oxygen potential was introduced at the base of the vessel and passed over the oxide; the weight change of the oxide was measured and material condensed from the vapour phase analysed by scanning electron microscopy (SEM). The results from the tests at 2123 K indicated that there was an increase in the overall releases for lanthanum, cerium and ruthenium in the case of the Ar/H_2O test but that the differences were small. The largest weight losses were measured for the ruthenium samples, with slightly higher releases in the steam atmosphere. In the case of the urania samples, weight losses of approximately 5% were determined; slightly higher releases were found for the Ar/O_2 case. The results from the analysis of the coupons indicated low deposition rates in all the experiments; it is possible that the aerosol particles were very small and were carried off in the gas flow. In the case of Ce, La and Ru the deposition levels were below the detection limits of the ICPOES analysis method. The mass of uranium recovered from the coupons was less than that expected from the mass loss data. Two uranium tests were conducted at a higher temperature (2248 K) and the results showed a small increase in the release for the steam atmosphere test. The results indicated that, for the test conditions studied, the overall releases were not enhanced significantly by gaseous hydroxide species.

3.2. Development of models

The existing oxide database comprising UO_2–ZrO_2–SiO_2–CaO–Al_2O_3–MgO–BaO–SrO–La_2O_3–CeO_2–Ce_2O_3 was extended to include Fe_2O_3; this required the inclusion of 11 binary oxide systems to the database. In order to calculate fuel hyperstoichiometry, UO_{2+x}, new data were also incorporated into the urania fuel model. Uranium dioxide oxidises at high temperatures in steam to form hyperstoichiometric urania where x can range from 0 to 0.25. Increasing fuel stoichiometry has a large impact on reducing the fuel solidus and liquidus temperatures. In addition the retention of more oxygen in the melt will lead to an increase in the oxygen potential of the melt; this in turn can have a strong influence on the vapour pressures of key fission product species.

A literature review of the thermodynamic data for all the binary and ternary metal sub-systems was performed and where available the published phase diagrams assessed. Since no experimental work was planned in this area during the current programme, the assessments were limited to existing published data. Due to the lack

of data available for many of the stoichiometric compounds, in particular the intermetallic phases, assessments were performed to estimate these data. In view of the amount of effort required, emphasis was placed on the fission product interactions with the main core and concrete elements (i.e., U, Zr, Fe and Si). Less detailed work was conducted for the inter-fission product systems, since the concentrations are probably sufficiently low that these interactions will not be important. The sub-system U–Zr–Si–Fe was fully assessed first and a database with the optimised interaction terms compiled. The addition of each of the fission product systems Ba, Sr, La, Ce, Ru and Te with the four component system (U–Zr–Si–Fe) was then performed. Typically, the binary systems of the Ba–Sr–La–Ce system are characterised by regions of either large immiscibility and no intermetallic compounds (e.g., Ba–Ce) or ideal solutions (Ce–La and Ba–Sr). The tellurium and ruthenium systems however tend to contain many intermetallic phases; the exceptions to this are the Ba–Ru and Sr–Ru systems which are immiscible.

The extension of the metal–oxide system, U–Zr–Si–O, to include Fe–FeO has required the addition of three metal–oxide systems. The Fe–FeO system is an important component during the initial oxidation phase of the structural material in a severe accident and may play a significant role in fixing the oxygen potential of the melt due to the large amount of steel available. Unlike the oxide database which assumes fully oxidised Fe_2O_3, the metal–oxide database also has to consider the Wustite phase and the composition range (i.e., FeO_{1+x}). The interactions between liquid Fe and the liquid oxides of the U–Zr–Si system were assumed to be immiscible.

Calculations using sub-sets of the databases were performed to assess the uncertainties in the method and to estimate the chemical behaviour of melts during MCCI. A set of calculations to estimate the releases from the melt pool compared the use of an ideal pure substance database and non-ideal solution models in order to estimate the uncertainties associated with the two different approaches. The results showed that although the agreement between the databases for some of the elemental components was reasonable there were discrepancies in cases where complex interactions in the melt were predicted for certain concrete types but which were not treated in the simplified models.

Predictions were also made using the non-ideal oxide solution database of the behaviour of a proposed basemat sacrificial layer during MCCI. The evolution of the solidus and liquidus temperatures and, the weight fraction and compositions of the solid and liquid phases were determined for temperatures between the solidus and liquidus.

Thermodynamic calculations of material volatility were performed to investigate the trends shown in the amount of aerosol released during the validation experiments described in Task 3. The results were in broad agreement with those determined in the experiments; for example, Si was predicted to reduce SiO_2 to form SiO(g), whereas $CaSiO_3$ was more stable in the presence of Si or Zr. Temperature was also seen to be a very important parameter affecting the amount of SiO(g) predicted to be formed. The calculated releases of Ba and Sr for the tests performed at temperatures less than 1650°C were low in agreement with the experimental results. An increase in the partial pressures of the Ba and Sr species of approximately two-orders of magnitude were determined on increasing the temperature from 1650°C to 2000°C.

3.3. Validation experiments

3.3.1. Knudsen effusion experiments

In order to obtain more data on the behaviour of cerium oxide and lanthanum oxide in mixtures with silica, the thermodynamic activities in the systems Ce_2O_3–SiO_2, La_2O_3–SiO_2, and Ce_2O_3–SiO_2–ZrO_2 were studied by means of Knudsen effusion mass spectrometry in the temperature range 1800–2350 K.

The tungsten Knudsen effusion cell consisted of three separate parts: a lid with a conical Knudsen effusion orifice, a bottom part with cylindrical bore for the thermocouple wires and a central cylindrical container for the sample. Temperature measurement was performed with a W, Re thermocouple, with ice-water as the cold junction reference. In order to minimise the interaction between the samples and the tungsten components, the sample to be investigated was contained in an iridium crucible in the Knudsen cell. The stream of particles effusing from the Knudsen cell passes through a hole in the heat shield and a water-cooled diaphragm before entering the ionisation chamber of the mass spectrometer; the latter (Fisons Instruments, SXP Elite 400) is capable of detecting ions (charged 1+) up to 400 amu. The non-ionised fraction of the effusing species condenses on a cryogenic trap (N_2) above the ionisation chamber of the mass spectrometer.

The activity data obtained in each case were fitted with a Redlich–Kister equation to describe the deviation from ideal mixing behaviour. In the case of the $CeO_{1.5}$–SiO_2 system at 2250 K, the activity data were fitted to a second-order Redlich–Kister function. However, at 1900 K, a third-order function was needed to obtain a satisfactory fit. Interestingly, the nearly horizontal section (from $x CeO_{1.5} \approx 0.06$ to 0.17) in the resulting prediction for the activity function strongly suggested a miscibility gap in the liquid phase. For the $LaO_{1.5}$–SiO_2 system the activity data at 2070 K were fitted to a third-order Redlich–Kister function and the nearly horizontal part in the fit again suggested a miscibility gap. However, the relatively poor reproducibility of the measurements (compared to the cerium silicate series) and inconsistent data at 2300 K indicated that

either the La_2O_3–SiO_2 samples needed more time to arrive at equilibrium or that a surface layer formed on the sample due to interactions with the Knudsen cell materials. The activity data for the SiO_2–$CeO_{1.5}$–ZrO_2 series showed good reproducibility and that chemical equilibrium in the iridium crucible was established. The replacement of some of the silica in a silica–ceria mixture by zirconia apparently increases the activity of ceria.

3.3.2. Silica aerosol experiments

The experiments were conducted in a facility in which the samples were contained within a silica vessel which had various ports and branches for the entry of thermocouples and sampling tubes, and for the attachment of thermal gradient tubes as required. Each sample was loaded into a zirconia crucible which was positioned concentric with an iridium susceptor, which in turn was concentric with an alumina tube to aid insulation. For the tests performed at the highest temperatures the sample was contained in a dual crucible-susceptor of high density graphite. The powdered mixtures were heated to temperatures between 1460°C and 2000°C for periods of 5–11 min in flowing helium gas. After each test any mass loss from the sample was determined and the filters inspected using SEM to establish if any aerosol was transported by the gas flow. The matrix of experiments comprised tests with Si and Zr metal as the reducing component of the system. Barium and strontium species were used as the simulant low volatile fission products; one test was conducted using caesium to compare the release of a high volatile species.

In general, the aerosol generated from these tests was very uniform in term of its primary particle size and morphology. Primary particles were spherical and agglomeration had occurred to generate strings and complex structures in tests involving large concentrations of particles. Size analysis of the collected particulate indicated an average geometric primary particle size of 0.07 μm with high uniformity. The exceptional test involved Cs during which a large quantity of material was vapourised at low temperature (~700°C); so much so that the pyrometer was obscured and the walls of the vessel became coated with a metallic film.

SEM/EDS analysis of the aerosol material from the tests at temperatures $\leqslant 1630$°C confirmed that the composition was mainly Si. In tests involving Ca, Ba, Sr and Zr, their levels were below the detection limit for EDS. Bulk chemical analyses of the aerosol indicated the material was predominantly silicon with minor quantities of other elements present (excluding oxygen). Ba and Sr were detected but at very low levels compared with Si (<0.1%). The release of aerosol was small for tests involving Zr metal foil as the surface appeared to have been pacified by an adherent layer of zirconium silicate.

Three tests were conducted at sample temperatures of ~2000°C. The first test involved zirconium powder and barium silicate but had to be aborted due to an energetic reaction. The other two tests were performed successfully and aerosol samples were taken during the course of each test; the particles comprised predominantly silicon with up to 10 wt% of strontium or barium.

3.4. Task 4: Sensitivity studies

In order to perform the sensitivity studies, the boundary conditions for the calculations were defined first. This involved a review of core configurations in the reactor cavity for concrete and sacrificial basemats and an analysis of melt stratification between oxidic and metallic phases for different scenarios. The chemical constitution of the fuel and fission products in the core-concrete melt and their distribution in the different melt phases were also assessed and confirmed by the results from available experimental studies. A selection of radiologically relevant fission products for release calculations was performed based on radiotoxicity and fraction in the core; this selection was in good agreement with a previous study [5].

The sensitivity of fission product release predictions to the various parameters was assessed by scoping calculations using the stand-alone thermochemical equilibrium code CHEMSAGE [6]. A pure substance thermochemical data file was compiled, EUMCCI, comprising phases of the most important radiologically relevant fission products; data for the liquid oxide and metal phases were included but these were treated as ideal. Sensitivity calculations for different MCCI scenarios were carried out to assess the influence on the releases during MCCI of various parameters for example, temperature, concrete type and melt layering [7]. The results showed which species of the pure substance database are relevant for fission product release calculations and hence should be studied further in sensitivity calculations investigating the influence of uncertainties in the thermochemical data. The modelling of the dissolution of a total amount of concrete can be performed within a single thermochemical equilibrium calculation or as a sequence of separate thermochemical calculations, each involving a part of the total concrete. The results for calculations of a core melt with initially 40%Zr oxidation confirmed that at least three steps of concrete dissolution were required.

Sensitivity calculations were also performed using RELOS.MOD2 incorporating the module version of CHEMSAGE, named CHEMAPP, and the EUMCCI database [7]. These calculations showed a significant influence of the Gibbs energy variations on the fission product releases; the specific relevance of individual species also differed significantly. For example, whereas a 10% decrease of the Gibbs energy for the gaseous

species $CeO_2(g)$, and thus a stabilisation of this species, resulted in an increase of the total Ce release by a factor of 600, a similar stabilisation of CeOH(g) showed no influence on the total Ce release. A reduction of the Gibbs energy uncertainty from 10% to 3% decreased considerably the deviations of the total Sr release from up to four to generally one-order of magnitude. As expected, a decrease of the Gibbs energy of a species in the gas phase resulted in an increase of the total release of the species considered. Correspondingly, a decrease of the Gibbs energy of liquid phase species reduced the total release. The distribution of the different species contributing to the total Ru release was a function of the Gibbs energy variation of the Ru gas phase species. In contrast, Gibbs energy variations of the liquid phase Ru species showed no effects on the speciation of the Ru species released but did influence the total Ru releases. However, for a complete evaluation of the influence of Gibbs energy variations and the relevance of the individual species, the specific boundary conditions have to be taken into account since these could influence significantly the speciation of the releases. The results of this sensitivity study were valid only for scenarios with high oxygen potentials, in particular for late-phase MCCI situations. At lower oxygen potentials, for example in core melts with significant amounts of metallic Zr (early stages of MCCI), the metallic fission product species are more important and this could have a significant influence on the results of a corresponding sensitivity study.

4. Conclusions

The THMO ex-vessel project on thermochemical data and modelling has provided new experimental data and critically assessed thermodynamic parameters for use in assessment studies of the consequences of melt–basemat interactions during a severe accident. Methods have also been developed to calculate the chemical equilibrium of the multi-component melt and hence provide data on the constitution of the condensed and gas phases. The sensitivity in these predictions for ex-vessel interactions to factors such as accident conditions (e.g., temperature, concrete type, melt layering) and the uncertainty in the thermochemical and kinetic data has also been investigated.

The development of data and modelling techniques for investigating core–basemat interactions has wider applications, for example, in the assessment of candidate sacrificial materials which could be used in the reactor cavity to dilute the core melt and improve the heat conducting properties. The modelling of the thermal-hydraulic behaviour of high temperature melts is also dependent on the changes in chemical composition and the relative amounts of the solid and liquid phases. The development of the calculational tools will assist in reducing some of the uncertainties, in particular the prediction of the evolution of melt temperatures, which in turn influences the extent of fission product releases during a severe accident.

An in-vessel thermochemical database has also been developed within the Fourth Framework Programme to study core degradation phenomena, for example the interaction of the core melt with the steel primary vessel and stratification effects in the melt. As part of the EC Fifth Framework Programme this database will be combined with the ex-vessel database and further extended to produce a common European database for application in severe accident codes. The work will be performed as part of the ENTHALPY project and supported by experiments to validate the models.

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