CALCULATION OF DECOMPOSITION PROPERTIES OF SILICATE CONCRETE AND APPLYING THEM IN SIMULATIONS OF SEVERAL EXPERIMENTS WITH AC²-COCOSYS

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ABSTRACT

As part of the research project “Analysis and externel validation of the AC² model basis (AVAMO)”, simulations of Molten Core-Concrete Interactions (MCCI), are conducted. In this work, simulations of the BETA, MOCKA and OECD-CCI tests, performed with COCOSYS 3.0, will be presented.

The focus of this work is the calculation of the decomposition parameter of silicate concrete and the modelling of the heat transfer between melt and concrete. An approach for calculating the decomposition parameters of concrete is presented and the influences of the individual components are shown. In addition, a distribution factor D_{AXRA} is implemented, which can be used to map the erosion behaviour. Depending on the calculated decomposition properties of the concrete and the heating power, the effective heat transfer coefficients can be determined.

INTRODUCTION

In a postulated severe accident scenario in a light water reactor (LWR) a failing of the decay heat removal might lead to degradation of the reactor core. After the reactor pressure vessel has failed, the core debris will relocate to the reactor cavity region and interact with the structural concrete, known as Molten Core - Concrete Interaction (MCCI). MCCI is mainly associated with the following three issues, that threaten the integrity of the containment. Firstly, the pressure build-up in the containment up to overpressure failure caused by the release of non-condensable H_2 during the decomposition of the concrete. Secondly, the composition of the containment atmosphere, resulting from releases of gases and aerosols, can potentially lead to accumulation of hydrogen with a subsequent possible hydrogen combustion. And thirdly, the propagation of the melt front as a consequence of the erosion of the concrete basement. How strong the presented issues are, depend on several parameters, including the composition of the concrete, composition of the melt and the respective decay heat.

The validation work presented focuses on several experiments, such as the BETA, MOCKA and CCI tests [1, 2, 3, 4], with silicate concrete and shows simulations with the severe accident analysis code COCOSYS 3.0 in AC² 2019. All simulations conducted mainly relate to the most significant parameters in COCOSYS for MCCI, identified in the frame of an Uncertainty and Sensitivity Analysis (UaSA) [5]: The decomposition temperature and decomposition enthalpy of concrete as well as the effective heat transfer coefficients (HEFF) from melt to concrete. The decomposition enthalpy of the concrete will be calculated as a function of the temperature based on the essential constituents. With adjustments of the HEFF values for the respective layers in the melt, the experimentally measured erosions are attempted to be reproduced for each so called decomposition temperature of the concrete. In addition a distribution factor D_{AXRA} is implemented, which can be used to map the erosion behaviour. The main objective is to reduce the user-influence of the parameters named above, related in particular to concrete erosion, and to extend the current approach for heat transfer between concrete and melt using constant effective

heat transfer coefficients. In previous investigations at PSS, a relationship between HEFF and heating power has already been shown, which is taken up again in this paper by consideration of the BETA experiments [6]. The calculation of the decomposition variables of concrete and their influence on the effective heat transfer coefficients, as well as the influence of the heating power are presented.

MODELLING

The focus in the description of the modelling of the investigated experiments is on the determination of the already mentioned essential parameters of an MCCI. The basic part of the modelling has been taken from previous investigations at PSS [6]. Changes in the composition of the concrete always involve a change in density, decomposition temperature and decomposition enthalpy of the concrete. These values, especially the decomposition enthalpy, can often only be determined experimentally [7]. In this approach, it is calculated using the concrete composition. For a classification of the determined values, they are compared with values from literature.

In a first step the most important components of the concrete are determined. The main concrete compositions of the considered experiments are given in Table 1, which had to be rescaled to 100% in some cases. Silicate concrete consists, in addition to the components listed in Table 1, of compounds such as $CaCO_3$ and $Ca(OH)_2$. The fractions of these two compounds are usually not directly given in the concrete compositions and must therefore be calculated from the concrete compositions. In addition, $CaCO_3$ and $Ca(OH)_2$ require a significant amount of energy to be dissolved due to the high enthalpies of formation, wherefore the fractions have to be determined. The following equation gives the amount of energy the respective compounds must receive to dissolve, which results from the differences of the enthalpy of formation. $Ca(OH)_2$ dissolves between 673 - 873 K and $CaCO_3$ between 873 - 1173 K [8, 9].

$$CaCO_3 + 1778.45\frac{kJ}{kg} \rightarrow CaO + CO_2 \qquad Ca(OH)_2 + 1475.18\frac{kJ}{kg} \rightarrow CaO + H_2O \text{ [9]}$$

$CaCO_3$ can be calculated from the fraction of CO_2 in the concrete, as CO_2 only exists in combination with $CaCO_3$ [10]. The remaining part of CaO, which is not required for the formation of $CaCO_3$, is therefore available for the formation of $Ca(OH)_2$, provided that sufficient H_2O is present in the concrete. The calculated fractions can be found in Table 2. In the MOCKA 5.4 experiment, the fraction of $CaCO_3$ is given with 7.8 wt-%, which agrees very well with the calculation of 7.71 wt-%.

Table 1: Main constituents of concrete in the considered experiments [1, 2, 3, 4]

Main concrete constituents	BETA V1.5 - V1.8 / V2.1	MOCKA 5.1 / 5.5	MOCKA 5.4	OECD-CCI 1	OECD-CCI 3
Al_2O_3 [wt-%]	0.0416	0.1217	0.1383	0.0079	0.0367
CaO [wt-%]	0.1405	0.1148	0.1304	0.0873	0.1748
CO_2 [wt-%]	0.0440	0.0299	0.0339	0.0092	0.1020
Fe_2O_3 [wt-%]	0.0104	0.1200	0.0000	0.0081	0.0155
H2O [wt-%]	0.0383	0.0612	0.0696	0.0381	0.0384
MgO [wt-%]	0.0073	0.0329	0.0374	0.0061	0.0088
SiO_2 [wt-%]	0.7179	0.5196	0.5904	0.8433	0.6237

Table 2: Calculated compounds of concrete in the considered experiments

Main concrete compounds	BETA V1.5 - V1.8 / V2.1	MOCKA 5.1 / 5.5	MOCKA 5.4	OECD-CCI 1	OECD-CCI 3
$CaCO_3$ [wt-%]	0.1001	0.0679	0.0771	0.0210	0.2320
$Ca(OH)_2$ [wt-%]	0.1116	0.1014	0.1152	0.0999	0.0592

In addition to the enthalpies of formation, the enthalpies of the constituents of the concrete are calculated up to a temperature of 2000 K using the Shomate equations, taking into account the temperature-dependent transformations and reactions of the respective components [11].

The enthalpies multiplied by the respective fractions are summed to obtain a total enthalpyas a function of temperature. It is assumed that the resulting H_2O and CO_2 from the dissolution of $CaCO_3$ and $Ca(OH)_2$ do not remain in the concrete. They no longer make an additional contribution to the enthalpy at increasing temperatures. An important point is to define at which temperature the concrete is decomposed. The exact definition of the so called decomposition temperature still has to be clarified in further works. Due to information provided by the experimenters and current developments, which assume decomposition temperatures of up to 1800 K, a temperature range of 1400 K - 1800 K was selected for the decomposition temperature [5, 12]. Table 3 shows the resulting decomposition enthalpies in the defined temperature range for the considered experiments and the experimenters' data as a comparison . It can be seen that the calculated decomposition enthalpy values agree well with the values given by the experimenters from the OECD-CCI experiments.

Table 3: Calculated decomposition Enthalpy of the given concrete compositions

Decomposition Enthalpy [kJ/kg]	BETA V1.5 - V1.8 / V2.1	MOCKA 5.1 / 5.5	MOCKA 5.4	OECD-CCI 1	OECD-CCI 3
at 1400 [K]	1674.03	1595.44	1678.38	1566.85	1783.61
at 1500 [K]	1782.18	1700.62	1785.65	1680.12	1883.82
at 1600 [K]	1890.95	1806.43	1893.59	1794.02	1984.63
at 1700 [K]	2000.28	1912.83	2002.12	1908.47	2085.97
at 1800 [K]	2114.92	2075.10	2111.21	2027.14	2194.95
specifications from the experimenters [kJ/kg]	-	2380.00 [3]	2380.00 [3]	1600.00 [4]	1720.00 [4]

After calculating the enthalpy of the concrete, nine different experiments are simulated with COCOSYS, using the combinations of decomposition temperature and associated decomposition enthalpy according to the concrete composition. Each experiment is simulated five times with the calculated decomposition parameters. With simultaneous adjustments of HEFF for the respective layers in the melts, the last experimentally measured erosions are attempted to be reproduced in each simulation. The erosion behaviour of the experiment is mapped using a distribution factor D_{AXRA} between axial and radial HEFF, as shown in Table 4.

Table 4: Determination of D_{AXRA} and the resulting erosion behaviour

D_{AXRA} [-]	Resulting erosion behaviour	EXAMPLE		
		D_{AXRA} [-]	$HEFF_{Axial}$ [W/(m²K)]	$HEFF_{Radial}$ [W/(m²K)]
<1	Axial erosion lower than radial erosion	0.25	1200	4800
=1	Axial and radial erosion are identical.	1.00	1200	1200
>1	Axial erosion greater than radial erosion	1.50	1200	800

In total 45 simulations are carried out and compared with each other in order develope a correlation on this basis. This is possible because the resulting decomposition enthalpies differ only slightly between the MOCKA, BETA, and CCI experiments. In the BETA experiments, different heating powers are also investigated, wherefor the approach here developed can be extended by the influence of the heating power.

RESULTS

Figure 1 shows, using the concrete composition of the BETA experiments as an example, the fraction of the components of the concrete in the total enthalpy (left axis) and the total enthalpy summed over the temperature (right axis). The considered decomposition range is marked. SiO_2 accounts for the largest amount with Al_2O_3 and CaO in this range with up to 80%. In addition, it becomes obvious whether $CaCO_3$ and $Ca(OH)_2$ have an influence on the total enthalpy. The dissolution of $CaCO_3$ is visible from 900 K on and the dissolution of $Ca(OH)_2$ from 1200 K on. They account for 14 - 17 % of the total enthalpy in the temperature range considered. Accordingly, these compounds must not be neglected in the calculation.

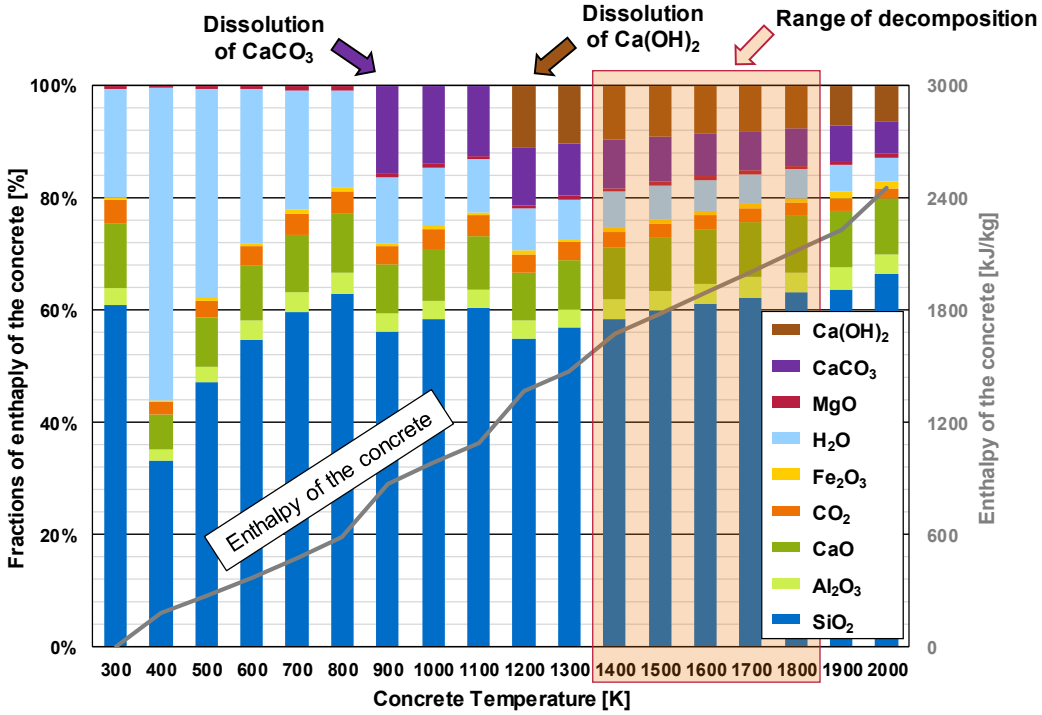

Figure 1: Fractions of the constituents to the calculated decomposition enthalpy of concrete and total calculated decomposition enthalpy over the temperature from BETA-Experiments

Figure 2 (a) shows the resulting axial values for HEFF of the metallic (left axis) and oxide layer (right axis) as a function of the decomposition temperature of all simulated experiments. It can be seen in each experiment that the HEFFs become higher as the decomposition temperature increases. At higher temperatures, more energy has to be added to the concrete so that it decomposes. The axial HEFFs of the metallic layer are between 315 and 16000 W/(m²K) and the HEFFs of the oxide layer are between 75 and 4500 W/(m²K). For the BETA V1.8 test, the highest values are obtained for HEFF. This can be explained by the highest average heating power of 1775 kW. Furthermore, the value of HEFF for the oxide layer of the CCI-1 experiment lies very close to the value of the BETA V2.1 experiment. In both experiments, a similar average heating power of 149 kW respectively 125 kW was injected into the melt. For the same heating power and decomposition enthalpies, identical HEFFs can be assumed, implying that the correlation can be applied to independent experiments. In the MOCKA experiments, the melt is heated through a continuous injection of thermite. Therefore, no heating power is specified by the experimentators. However, the maximum possible heating power can be determined using the enthalpies of the thermite at a throw-in temperature of approximately 2821 K. This results in a maximum heating power of 516 – 564 kW. According to this, the HEFF's of the MOCKA experiments should be above those of the BETA V1.5 experiment with an average heating power of 467 kW. Due to the fact that they are closer to the HEFF's of the BETA V2.1 test, it can be concluded that the effective heating power of the MOCKA tests is lower. One possible explanation is that a large part of the energy is dissipated through the upper surface and does not cause the concrete to heat up.

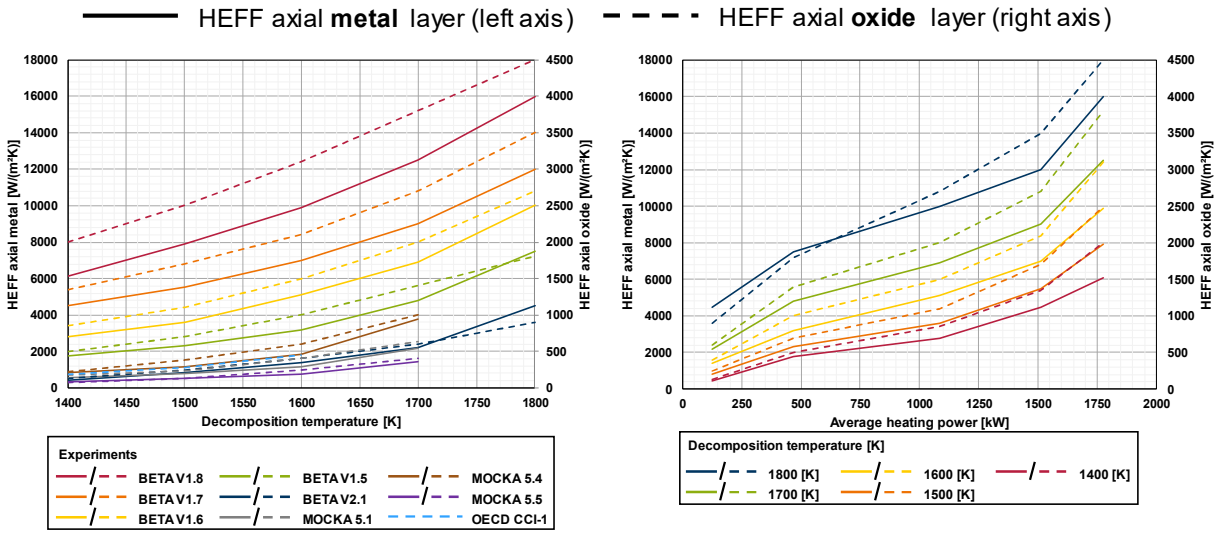

Figure 2: Effective heat transfer coefficients over the decomposition temperature

Figure 2 (b) shows the HEFF values plotted against the heating power to demonstrate the influence of the heating power regarding the BETA experiments at different decomposition temperatures. The other considered experiments are not shown here, as the used range of heating capacities is quite small. Basically, a slightly parabolic increase in HEFF can be seen. The increase can be seen in the metallic as well as in the oxidic layers. With low heating powers, the HEFFs are even closer together than with higher heating powers. At 1400 K, the difference between the HEFF values of the metallic layer is about 5650 W/(m²K) and that of the oxide layer 1865 W/(m²K). For 1800 K, the difference in the HEFF values of the metallic layer is about 11500 W/(m²K) and that of the oxidic layer 3600 W/(m²K).

D_{AXRA} determines the radial HEFF value range between 0.26 and 6.5. This also implies a dependency on the heating power. For the BETA V1.8 experiment with an average heating power of 1775 kW, the value is 6.5 compared to a value of 1.65 for the BETA V2.1 test with an average heating power of 125 kW. The axial erosion in the experiments is significantly greater than the radial erosion, resulting in D_{AXRA} greater than one. In the MOCKA experiments, the D_{AXRA} is between 1.0 and 1.65, which is due to the more uniform axial and radial erosion. The CCI-1 test shows a significantly higher radial than axial erosion, wherefor D_{AXRA}=0.26 results. The reasons for the different erosion behaviours are to be clarified in future works.

CONCLUSION

Depending on the concrete composition and the definition of the decomposition temperature of the concrete, it is possible to calculate a decomposition enthalpy of the concrete. The concrete constituents with the greatest influence on the decomposition enthalpy are SiO_2, CaO and Al_2O_3. In addition, the compounds $CaCO_3$ and $Ca(OH)_2$ must be taken into account, which can be calculated using the fraction of CO_2. The investigations also show that the recommended modelling of a MCCI with constant heat transfer coefficients, between melt and concrete, at different heating powers, is not adequate. With this approach, the effective heat transfer coefficients of the metallic and oxidic layer can be determined as a function of the heating power based on the decomposition temperature for different experiments. Increasing heating powers and decomposition temperatures result in higher heat transfer coefficients. The direct influence of decomposition enthalpy and concrete composition on heat transfer must not be neglected and still needs to be investigated. In addition, a distribution factor D_{AXRA} between the axial and radial HEFF is defined in the simulations in order to be able to map the different erosion behaviours. With this method, it is possible to significantly reduce the influence of the user using the presented correlation. Furthermore, the current approach in COCSYS can be maintained and easily expanded.

In future works, a definition of the decomposition temperature of the concrete has to be identified. Moreover, the reason for the different erosion behaviour is to be investigated in order to be able to define the highlighted connection even more comprehensively. The calculation of the enthalpy of decomposition currently only refers to silicate concrete. It is therefore of interest to evaluate the calculation for other types of concrete.

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