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Modeling of Viscosity and Heat Transfer of Complex Oxidic Melts in WECHSL

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Institut für Angewandte Thermo- und Fluidodynamik

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Abstract

Attention has recently been drawn to the importance of the melt properties, among others the viscosity of oxidic melts, on the heat transfer and on the melt behaviour during MCCI. In the first step the viscosity model in WECHSL was changed in order to achieve a better agreement between the available viscosity measurements and the viscosities predicted by WECHSL. In order to account for the variation of the viscosity with temperature an iterative scheme was used in WECHSL to determine the temperature gradient of the pool bulk temperature to the interface temperature. In addition, the influence of porosity of the melt on the thermal conductivity was modeled. Finally, the so-called property ratio method was implemented for correcting the Nusselt number. A validation matrix which consists of the ACE and SURC experiments was used to assess the improvements of the code.

Viskosität und Wärmeübergang von komplexen oxidischen Schmelzen in WECHSL

Zusammenfassung

Neulich wurde der Einfluß der physikalischen Eigenschaften von Kernschmelzen, insbesondere der Viskosität von oxidischen Schmelzen, auf den Wärmeübergang und das Verhalten von Kernschmelzen während der Wechselwirkung mit Beton erkannt. Zunächst wurde das in WECHSL verwendete Modell zur Berechnung von Viskositäten komplexer oxidischer Schmelzen so abgeändert, daß eine gute Übereinstimmung mit verfügbaren Meßergebnissen erreicht wurde. Zusätzlich wurde die Abhängigkeit der Zähigkeit von der Temperatur in die Berechnung der Grenzschichttemperatur zwischen der oxidischen Schmelze und Beton einbezogen. Der Einfluß der Porosität auf die Wärmeleitfähigkeit wurde ebenfalls berücksichtigt. Danach wurde eine Korrektur der entsprechenden Nusselt-Zahl, die den Einfluß temperaturabhängiger Zähigkeit beschreibt, durchgeführt. Anhand von Nachrechnungen der ACE- und SURC-Experimente wurde das verbesserte WECHSL-Rechenprogramm getestet.

Contents

1.	Introduction	1
I.	Viscosity modeling in WECHSL	1
I.1	Modification of the D_i coefficients	6
II.	Thermal conductivity of porous materials	10
III.	Effects of temperature varying viscosity on heat transfer	11
IV.	Assessment matrix and WECHSL results	12
IV.1	Determination of the exponent n	12
IV.2	Results for ACE-L2	15
IV.3	Experiment ACE-L7	17
IV.4	Experiment ACE-L8	19
IV.5	SURC-1 experiment	21
IV.6	SURC-2 experiment	23
V.	Conclusions	25
	References	25

1. Introduction

The WECHSL code has been validated using a comprehensive set of experiments selected from the BETA, SURC and ACE test matrices. This assessment has shown that the code has a wide range of applicability with respect to corium composition, concrete type and level of the internal power.

The WECHSL results are in quite good agreement with the findings of the experiments for the metallic melt experiments (BETA). Particularly, the model used in WECHSL according to which the heat transfer is determined by gas superficial velocity and crust formation was found to be adequate to represent a large variety of physical conditions ranging from liquid metal attack to crust-dominated ablation. The WECHSL code was also used to analyse oxidic experiments (SURC, ACE) to assess predictions for plant applications in an intermediate time period. Serious discrepancies were observed between the behaviour of complex oxidic melts in the experiment and that predicted by the WECHSL code. However, there are still open questions concerning the reliability of the experimental data and the understanding of the physics of this type of melts.

Attention has recently been drawn to the importance of the melt properties, among others the viscosity of oxidic melts, on the heat transfer and on the melt behaviour during MCCI. In the first step the viscosity model in WECHSL was changed in order to achieve a better agreement between the available viscosity measurements and the viscosities predicted by WECHSL. In order to account for the variation of the viscosity with temperature an iterative scheme was used in WECHSL to determine the temperature gradient of the pool bulk temperature to the interface temperature. In addition, the influence of porosity of the melt on the thermal conductivity was modeled. Finally, the so-called property ratio method was implemented for correcting the Nusselt number. A validation matrix which consists of the ACE and SURC experiments was used to assess the improvements of the code.

I. Viscosity modeling in WECHSL

Calculations of the viscosity of complex oxidic melts in the WECHSL code are based on the method proposed by Bottinga and Weill [1]. According to their correlation, the dynamic viscosity of a magmatic melt is calculated by the following formula:

$$\eta = 0.1 \cdot \exp \left\{ \sum_{i \in \{O\}} X_i D_i \right\}, \quad (1)$$

$$\{O\} = \{UO_2, ZrO_2, FeO, SiO_2, CaO, Al_2O_3\}$$

where X_i are mole fractions of the melt components.

The $D_i, i \in \{O\}$ coefficients are given in [2] for the melt constituents with SiO_2 content between 35 mole% and 85 mole% and for discrete values of the temperature between 1473 K and 2073 K. The temperature dependence of the D_i coefficients is described by the following relation:

$$D_i = A_i + \frac{B_i}{T} \quad (2)$$

where A_i and B_i are Arrhenius coefficients. A_i and B_i can be easily calculated from the tabulated data for $D_i, i \in \{O\}$ in [2]. Then Eq. 2 is used to determine the D_i coefficients for arbitrary temperatures.

Because of the lack of data for UO_2 and ZrO_2 $D_{UO_2} = D_{ZrO_2} = D_{TiO_2}$ is used in [2] for a content of SiO_2 in the melt in the range of 35 mole% and 75 mole%. For a SiO_2 content in the melt between 75 mole% and 85 mole% D_{UO_2} and D_{ZrO_2} are defined to be $D_{UO_2} = D_{ZrO_2} = D_{CaO}$. For a SiO_2 content less than 35 mole% A_i and B_i are obtained by linear interpolation using the data for the pure corium with the composition given in Table 1. In the range above 85 mole% of SiO_2 content, the A_i and B_i coefficients are estimated by linear interpolation using the data for pure SiO_2 .

UO_2	ZrO_2	FeO	Cr_2O_3	NiO
52.1	19.1	23.0	3.9	1.9

Table 1: Core melt composition in weight%

Skoutajan et al. [3] experimentally investigated the viscosity of corium/silicate melts at temperatures between 1573 K and 1873 K. They found that the calculations from [2] give values far below the experimental data.

Using Eq. 1, WECHSL results are presented in Fig. 1 and Fig. 2, respectively, for the viscosity of the different types of corium (Tab. 1 and Tab. 2) and for different content of concrete with the composition given in Tab. 3.

UO ₂	ZrO ₂
78.1	21.9

Table 2: Core melt composition in weight%

SiO ₂	CaO	Al ₂ O ₃
68.99	13.47	4.04

Table 3: Idealised siliceous concrete composition in weight%

SiO ₂	CaO	Al ₂ O ₃
28.27	26.02	3.48

Table 4: Idealised limestone-sand concrete composition in weight%

Calculated results underestimate the measured data considerably (Fig. 1). For a concrete content below about 20 weight% (SiO₂ content below about 30 mole%) in the melt with composition given in Tab. 2 the WECHSL results show unphysical behaviour, i.e. an increase of the viscosity with increasing temperature (Fig. 2).

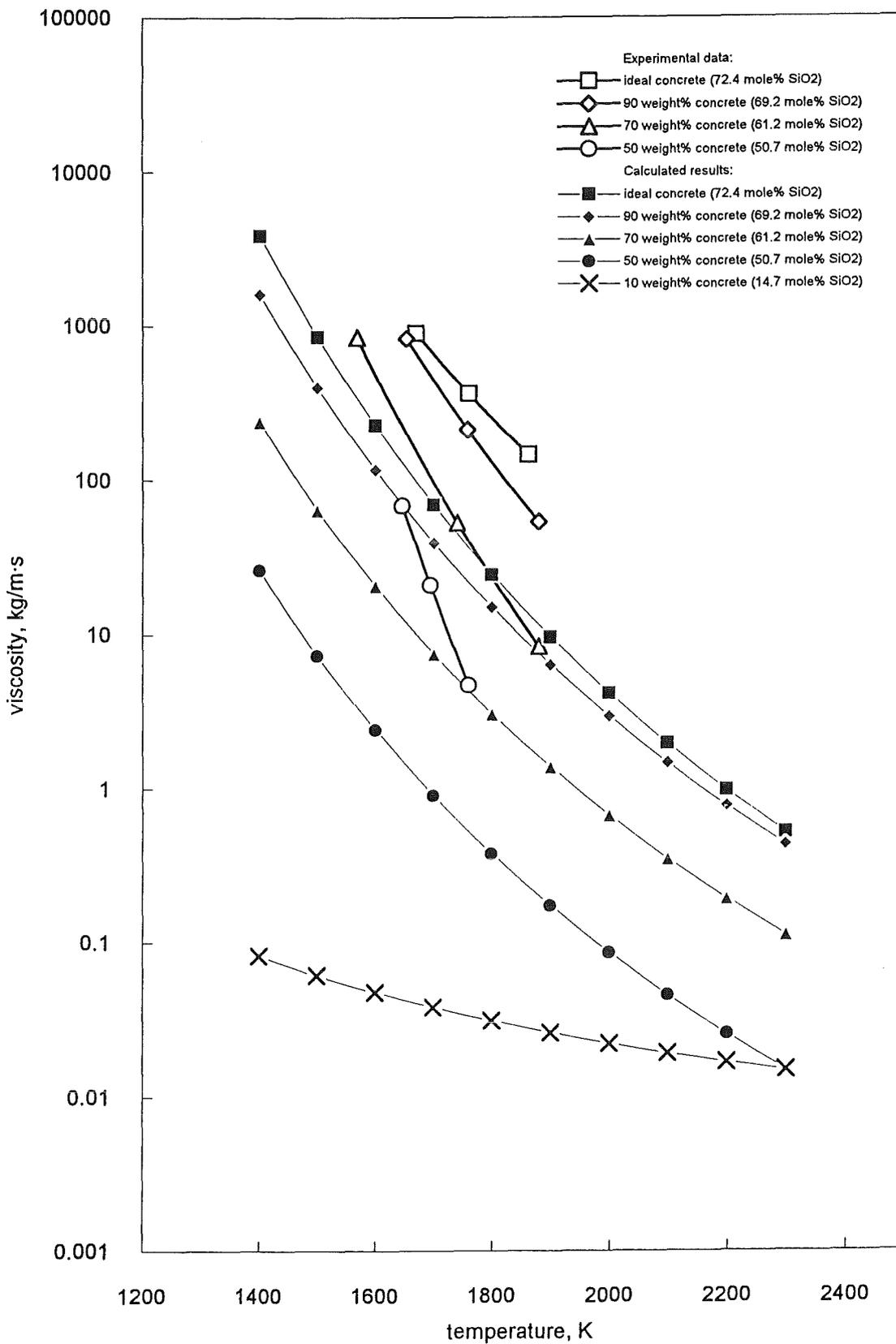


Fig. 1: Viscosity dependence on temperature for different concrete content in the melt (Tab. 1) - using old WECHSL data

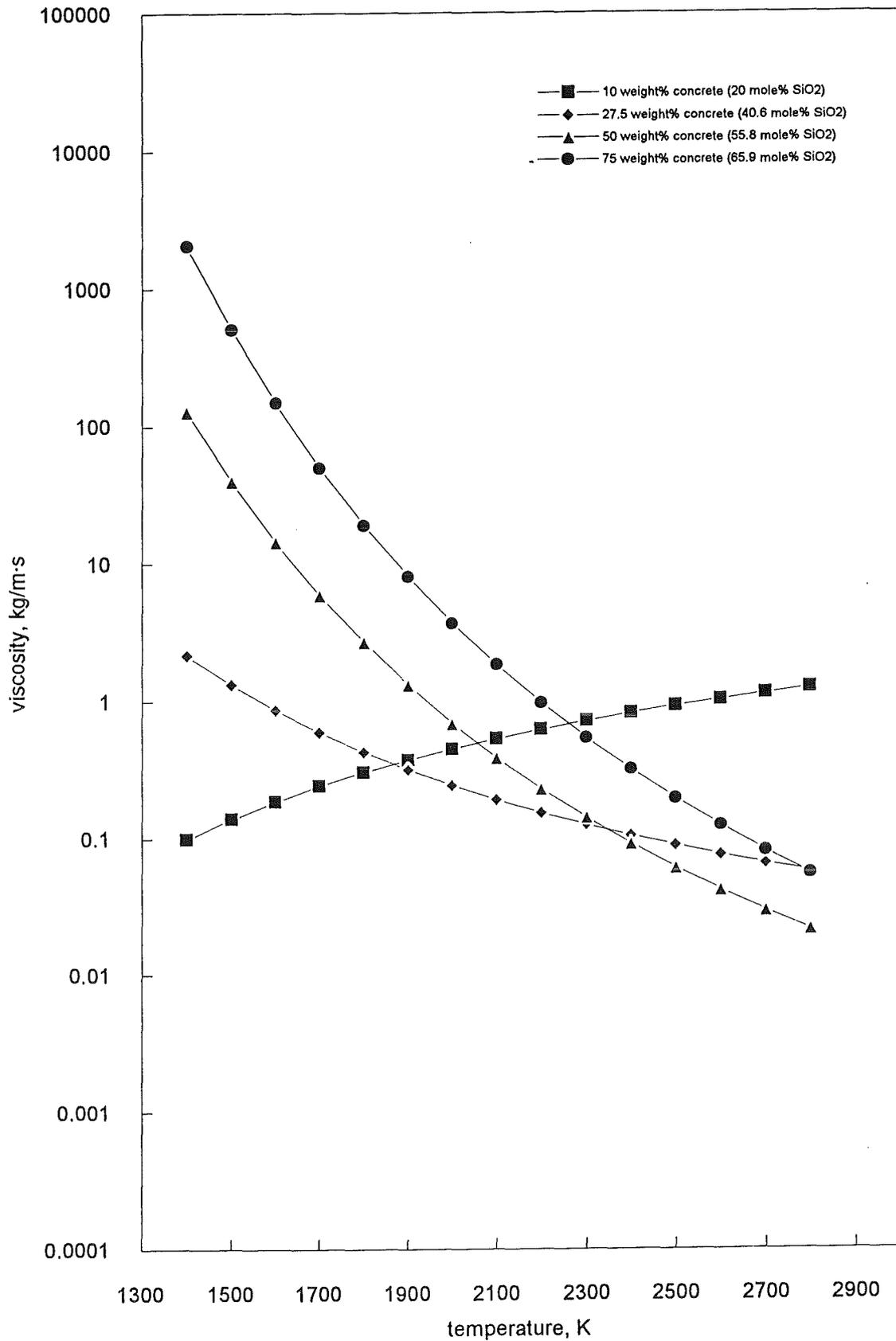


Fig. 2: Viscosity dependence on temperature for different concrete content in the melt (Tab. 2) - using old WECHSL data

I.1 Modification of the D_i coefficients

For SiO_2 content above 50 mole% in the melt (Table 1) only the D_{SiO_2} coefficients were changed in order to reproduce the experimental data given in [3]. For melt compositions containing less than 50 mole% of SiO_2 which are not accounted for in measurements [3], the D_{UO_2} and D_{ZrO_2} coefficients were modified in the following way. First, for the melt with 0 mole% SiO_2 content $D_{\text{UO}_2} = D_{\text{ZrO}_2}$ is calculated from Eq. 1 using the viscosity of pure UO_2 given in [4]. In the range of SiO_2 content in the melt between 0 mole % and 50 mole%, the D_{UO_2} and D_{ZrO_2} coefficients were obtained by linear interpolation using the old WECHSL D_{UO_2} and D_{ZrO_2} coefficients given in [2] for 50 mole% of SiO_2 in the melt and those for pure UO_2 , which have been determined in the previous step.

For the melt used in the calculations given in Fig. 1, WECHSL results using new estimated coefficients are depicted in the following figure (Fig. 3). In accordance with experimental data (SiO_2 content in the melt higher than 50 mole%) calculated results show a steep gradient of the viscosity compared to previous results (Fig. 1). The same tendency is observed for the SiO_2 content in the melt less than 50 mole%, but with lower viscosities than calculated in Fig. 1.

The viscosities for different concrete content in the melt (see Tab. 2 for composition) are shown in Fig. 4 and Fig. 5 for the siliceous and the limestone-sand concrete, respectively. Due to the described changes of the D_{UO_2} and D_{ZrO_2} coefficients, the unphysical behaviour does not occur for a SiO_2 content in the melt below about 30 mole%. However, the results in this range seem to be questionable.

Recently, viscosity measurements for a melt composed of 56.6 weight% of UO_2 , 15.9 weight% of ZrO_2 and 27.5 weight% of both siliceous and limestone-sand concrete (see Tab. 3 and Tab. 4) were performed by Roche [5]. These measurements show a non-Newtonian behaviour which was not observed in [3]. In the viscosimeter with a rotating spindle, the viscosity of the melt near the freezing temperature was observed to increase when the rotational velocity decreases from 200 to 10 revolutions per minute. The data measured at 40 RPM are also given in Fig. 4 and Fig. 5. The WECHSL results underestimate measured viscosities by about three orders of magnitude.

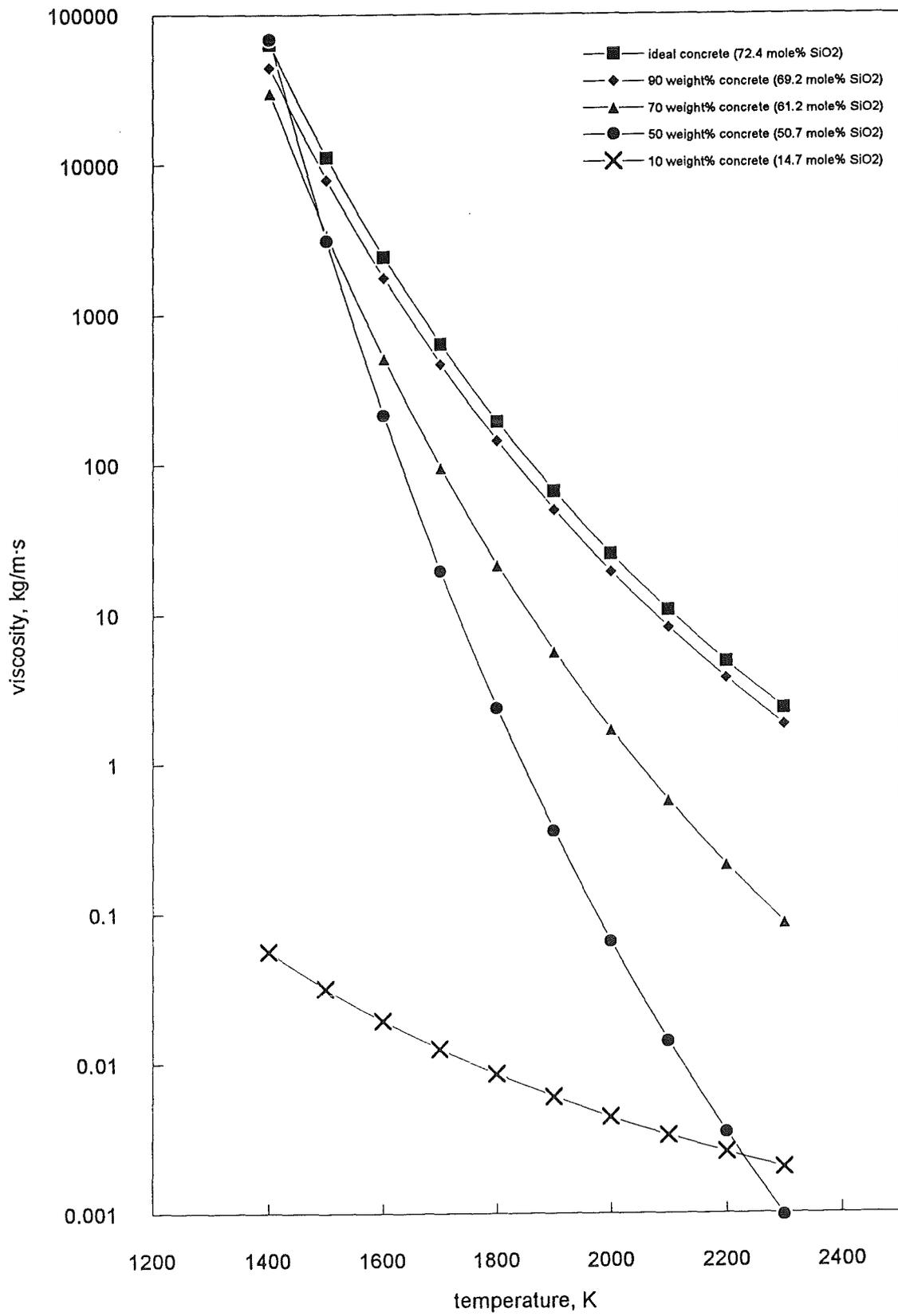


Fig. 3: Viscosity dependence on temperature for different siliceous concrete content in the melt (Tab. 1) - using new WECHSL data

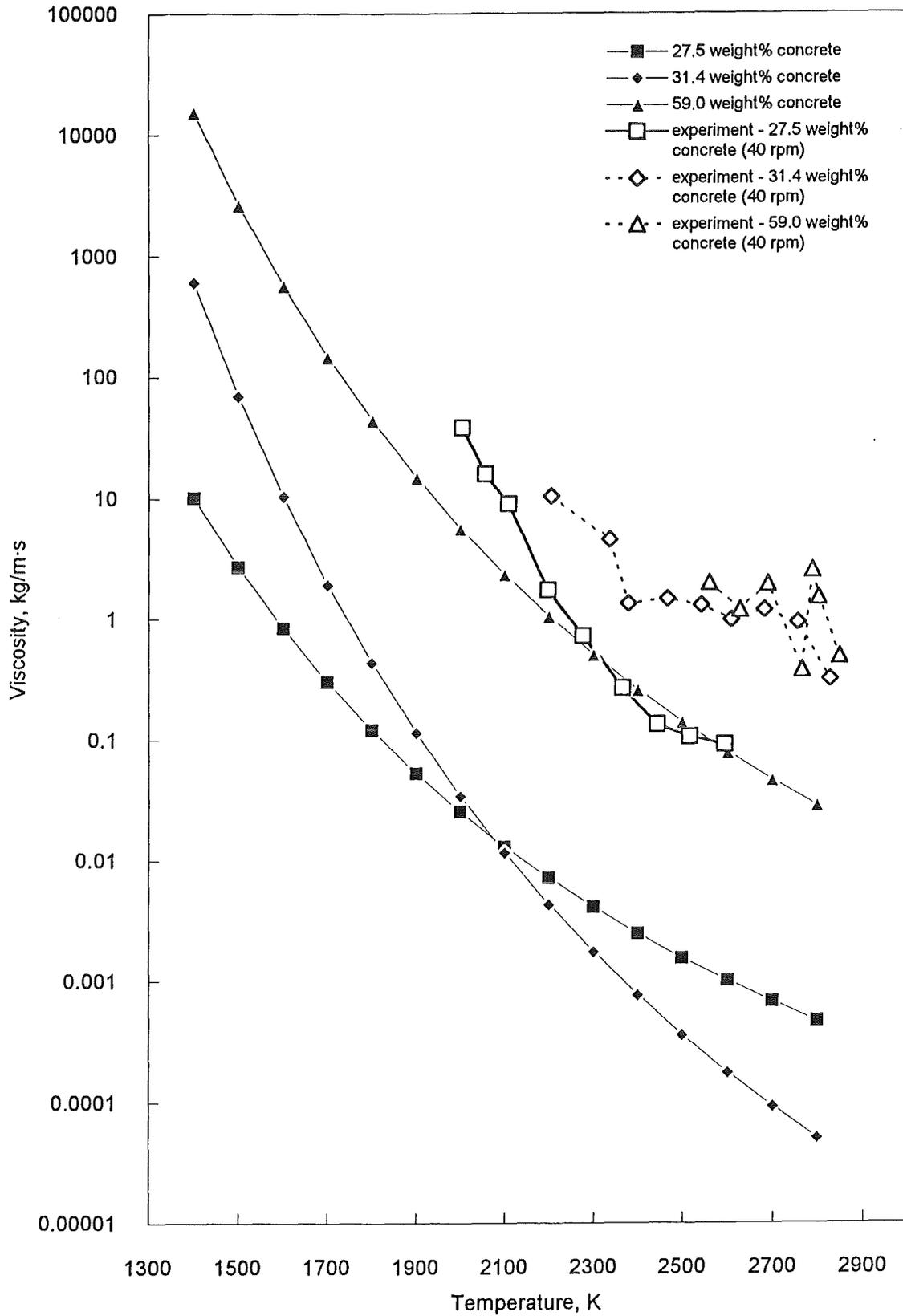


Fig. 4: Viscosity dependence on temperature for different siliceous concrete content in the melt (Tab. 3) - using new WECHSL data (experimental data for 31.4 and 59.0 wt% concrete are considered to be unreliable by the authors [5])

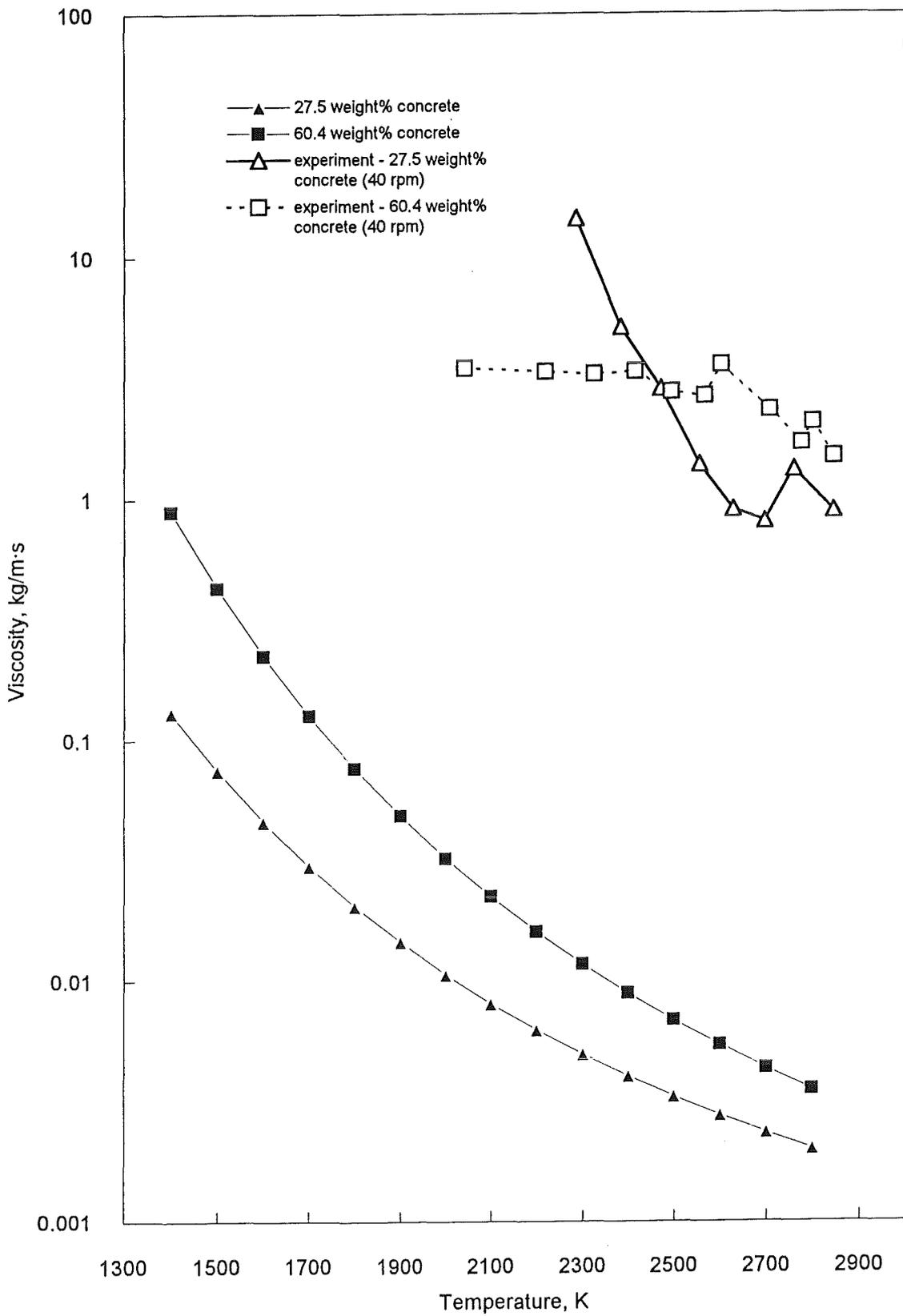


Fig. 5: Viscosity dependence on temperature for different limestone-sand concrete content in the melt (Tab. 4) - using new WECHSL data (experimental data for 60.4 wt% concrete are considered to be unreliable by the authors [5])

A comparison of WECHSL results with other available viscosity data is given in the following table (Tab. 5):

melt composition	viscosity, Pa·s		
	measurements [3]	old WECHSL	new WECHSL
UO ₂ 100w% T=3073 ÷ 3273 K	4·10 ⁻³ ÷ 8·10 ⁻³ 5.87·10 ⁻³ ÷ 5.46·10 ⁻³ [4]	3.67·10 ⁻³ ÷ 3.07·10 ⁻³	5.95·10 ⁻³ ÷ 5.68·10 ⁻³
30 mole% SiO ₂ 70 mole%, UO ₂ T=2663 K	5·10 ⁻²	6.91	2.3·10 ⁻³
UO ₂ 58.2 w% ZrO ₂ 21.7 w% FeO 13.8 w% Cr ₂ O ₃ 4.2 w% NiO 2.1 w% T=2073 K	4.3·10 ⁻³	4.96·10 ⁻³	3.39·10 ⁻³
UO ₂ 28.9 w% ZrO ₂ 11.1 w% FeO 41.5 w% Cr ₂ O ₃ 12.5 w% NiO 6.0 w% T=2673 ÷ 2773 K	2·10 ⁻³ ÷ 5·10 ⁻³	2.53·10 ⁻⁴ ÷ 2.08·10 ⁻⁴	2.63·10 ⁻⁴ ÷ 2.21·10 ⁻⁴

Table 5: Comparison of WECHSL results with measurements

Except for the viscosity of a corium with high content of FeO, the corrected modeling in WECHSL gives a satisfactory agreement with the available data.

II. Thermal conductivity of porous materials

The porosity of the melt which is caused by the gases released from the decomposing concrete will decrease the thermal conductivity of melt. Under the assumption of spherical pores the porosity correction of the thermal conductivity is given in [6] as follows:

$$\lambda_p = \lambda_o(1-p)^{3/2} \quad \text{for } p \leq 0.1 \quad (3)$$

and

$$(1-p)(\lambda_o - \lambda_p) \left[\frac{4}{\lambda_p + \lambda_o} + \frac{1}{\lambda_p} \right] = 5p \quad \text{for } p > 0.1 \quad (4)$$

where p is the porosity,

λ_p and λ_o are the thermal conductivities of the porous and dense melt, respectively. Eqs. (3) - (4) give a satisfactory agreement with available data [7], [8].

III. Effects of temperature varying viscosity on heat transfer

In all flow regimes of the gas film model modeled in WECHSL [9], the temperature difference between the temperature of the melt at the interface with the gas film and the decomposition temperature of the concrete determines the amount of heat transfer. In the bottom region of the pool, a thin boundary layer driven by microconvection between bubble release sites is assumed to exist. Along the inclined walls, a boundary layer is created by drag forces exerted by the gas flowing in the film. Both boundary layers result in a temperature gradient of the pool bulk temperature to the interface temperature. Because of the high thermal conductivity and the low viscosity of the metallic phase the temperature drop across the boundary layer is only of the order of a few degrees. For an oxidic melt, however, the temperature drop across the boundary layer is quite significant because of the low thermal conductivity and the high viscosity of the oxides.

The boundary layer and the heat transfer models in WECHSL were based on the assumption that the viscosity remains constant. This assumption is obviously not compatible with available viscosity data. In order to account for the variation of the viscosity with temperature an iterative scheme is used in WECHSL to determine the interface temperature between the melt and the concrete. This reduces considerably the total heat transfer from the molten pool to the concrete. In the second step, the so-called property ratio method is implemented for correcting the appropriate non-dimensional parameter, i.e.

$$Nu = Nu_o \left[0.645 \left(\frac{\mu_i}{\mu_b} \right)^n + 0.355 \right]. \quad (5)$$

Nu is the corrected Nusselt number and Nu_o the constant property solution. The viscosity μ_i is the viscosity at the interface temperature, while μ_b is evaluated at bulk temperature [10], [11]. The exponent n is a fit parameter.

IV. Assessment matrix and WECHSL results

The changes implemented in the WECHSL code are assessed on the basis of a comprehensive set of experiments selected from the SURC (SNL, USA) and ACE (ANL, USA) test matrices. The following table (Tab. 6) shows the matrix of the tests selected for the validation procedure.

Experiment	SURC		ACE			
Test	SURC-1	SURC-2	L2	L6	L7	L8
Cavity (1-dimensional)	Ø 40 cm		50 x 50 cm			
Concrete	Limestone	Basaltic	Siliceous	Lime- stone/ common sand	Lime- stone	
Zr	Yes		Yes			

Tab. 6: Validation matrix

IV.1 Determination of the exponent n

The ACE-L6 test [12] is used to fit the exponent n in Eq. (5). This is done using a WECHSL computation for a one-dimensional cavity consisting of siliceous concrete with 7 cm thick concrete/metal inserts and a melt consisting of one oxidic layer containing the dispersed metallic phase. The value of n was chosen to give a good prediction of the final erosion depth which was obtained in the experiment. The determined value is $n = -14$. In all ACE experiments the data were recorded after the concrete/metal inserts were ablated.

The former WECHSL version significantly overpredicts the final erosion depth of the ACE-L6 test (35 cm at 80 min instead of about 20 cm in the test (Fig. 6)). The fitted curve as well as the experimental data are given in Fig. 6. Despite of the

good agreement of the final erosion depth there is a strong discrepancy of the erosion as a function of time. In the time period of the ablation of the concrete/metal inserts the position of the erosion front was not determined.

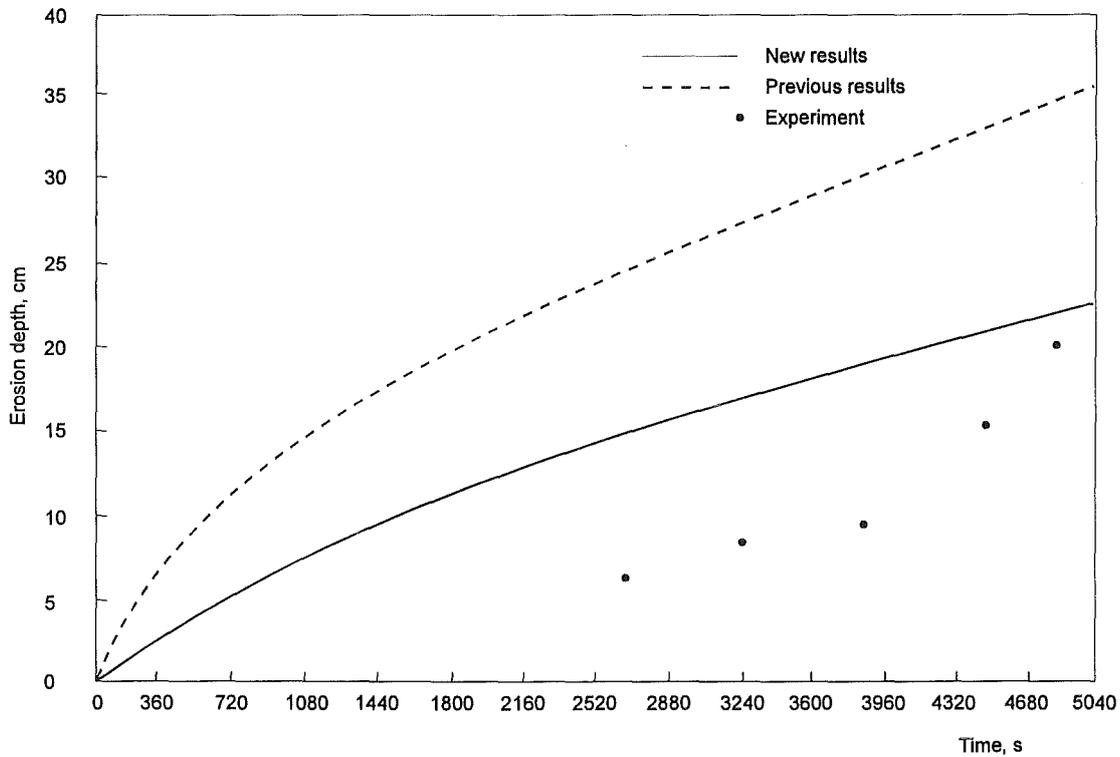


Fig. 6: Erosion depth

Compared to the previous WECHSL results (Fig. 7) the new model predicts a higher temperature level (about 150 K - 200 K) which is still too low in comparison with the measured data (Fig. 8).

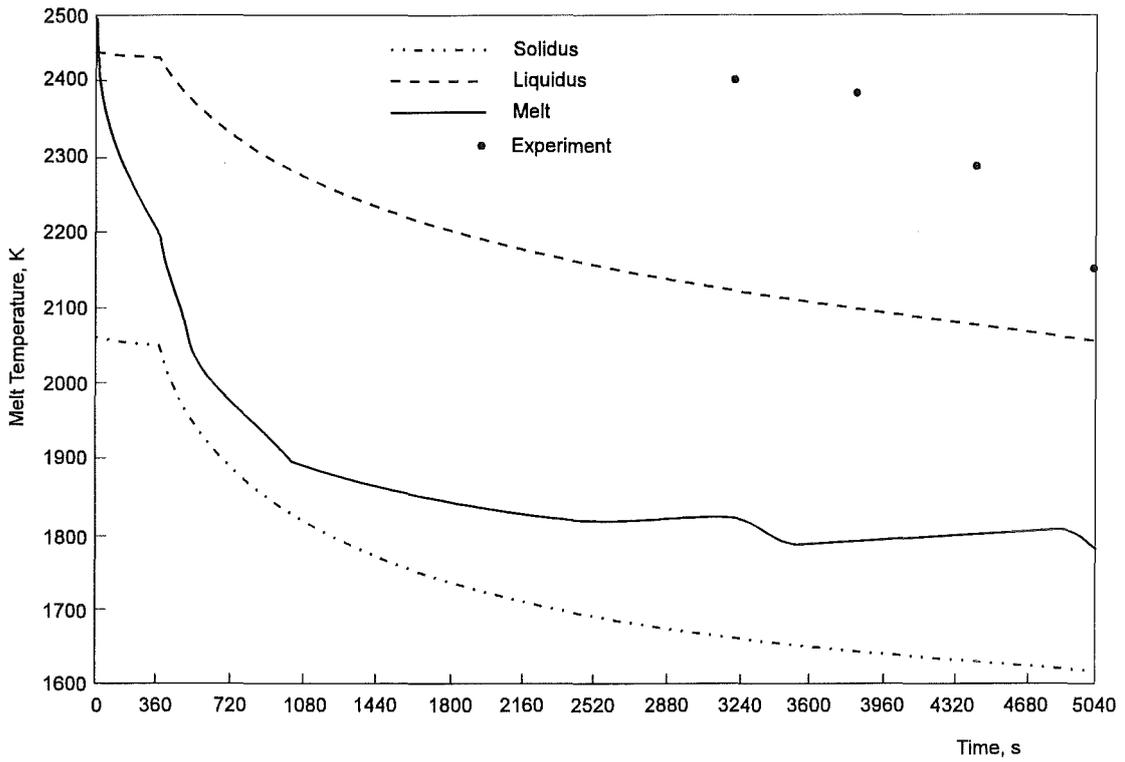


Fig. 7: Melt temperature (previous results)

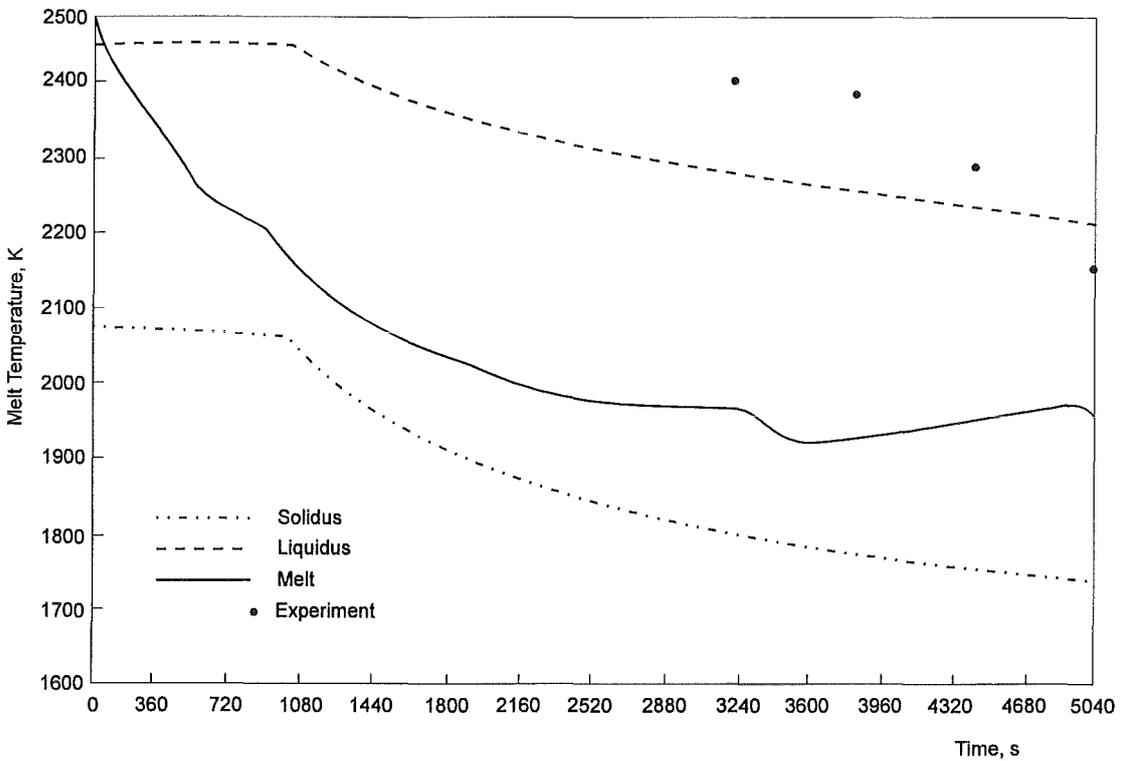


Fig. 8: Melt temperature (new results)

IV.2 Results for ACE-L2

All subsequent results are obtained using the previously determined exponent $n = -14$ in Eq. 5. The siliceous crucible consists of 1.5 cm thick concrete metal inserts which contain 59 % of the Zirconium metal in the corium inventory. The predicted erosion curve for the ACE-L2 test [13] obtained with the new WECHSL version agrees very well with experimental results whereas the concrete erosion is grossly overestimated by the old WECHSL version (Fig. 9).

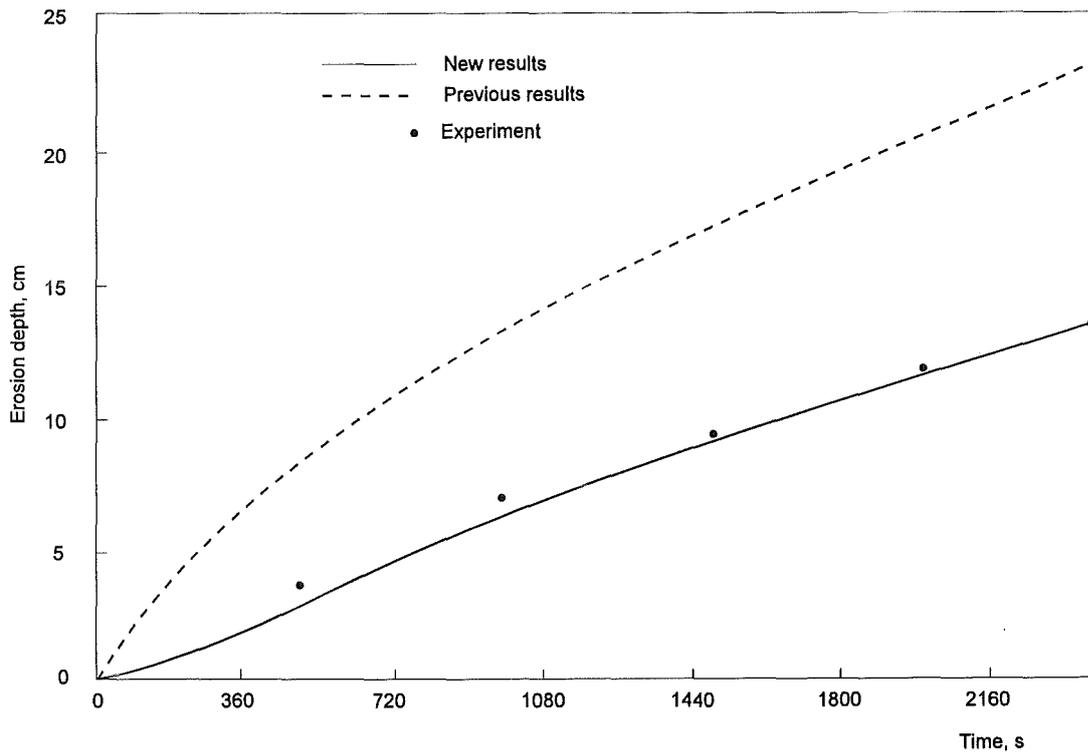


Fig. 9: Erosion depth

Even though there is a good agreement for the erosion, the predicted melt temperature still differs significantly from the measurements (Fig. 10) but some improvement is obtained compared to the previous results (Fig. 11).

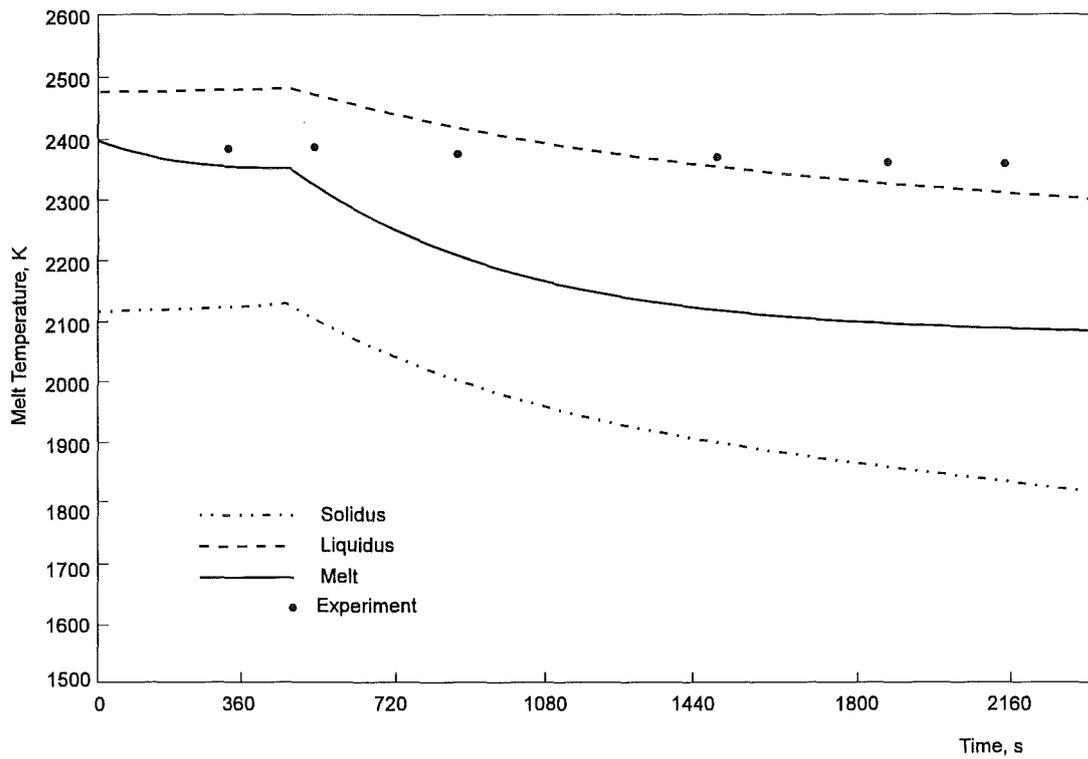


Fig. 10: Melt temperatures (new results)

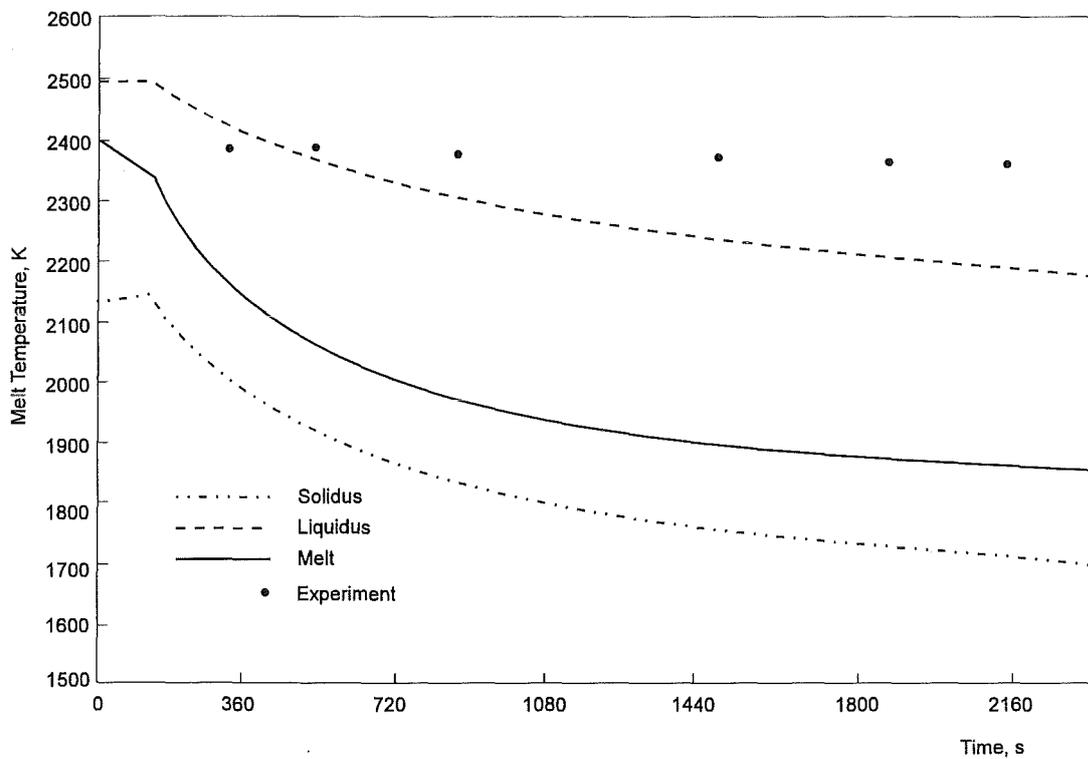


Fig. 11: Melt temperatures (previous results)

IV.3 Experiment ACE-L7

The ACE-L7 [14] calculations of the limestone/common sand basemat erosion which include 5.7 cm thick concrete/metal inserts are shown in Fig. 12.

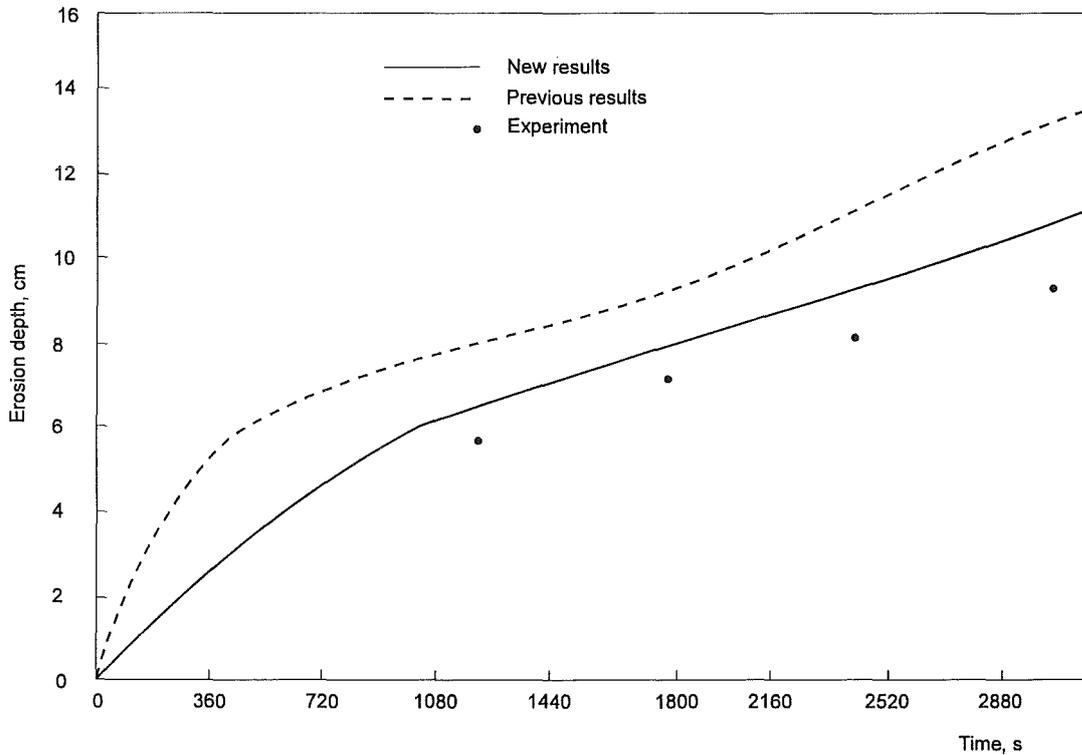


Fig. 12: Erosion depth

The new WECHSL version predicts a basemat erosion which is in good agreement with the experiment. The calculated values for the melt temperature differ again significantly from the measurements (Fig. 13, Fig. 14). but the discrepancy between the temperature levels is reduced in the new results.

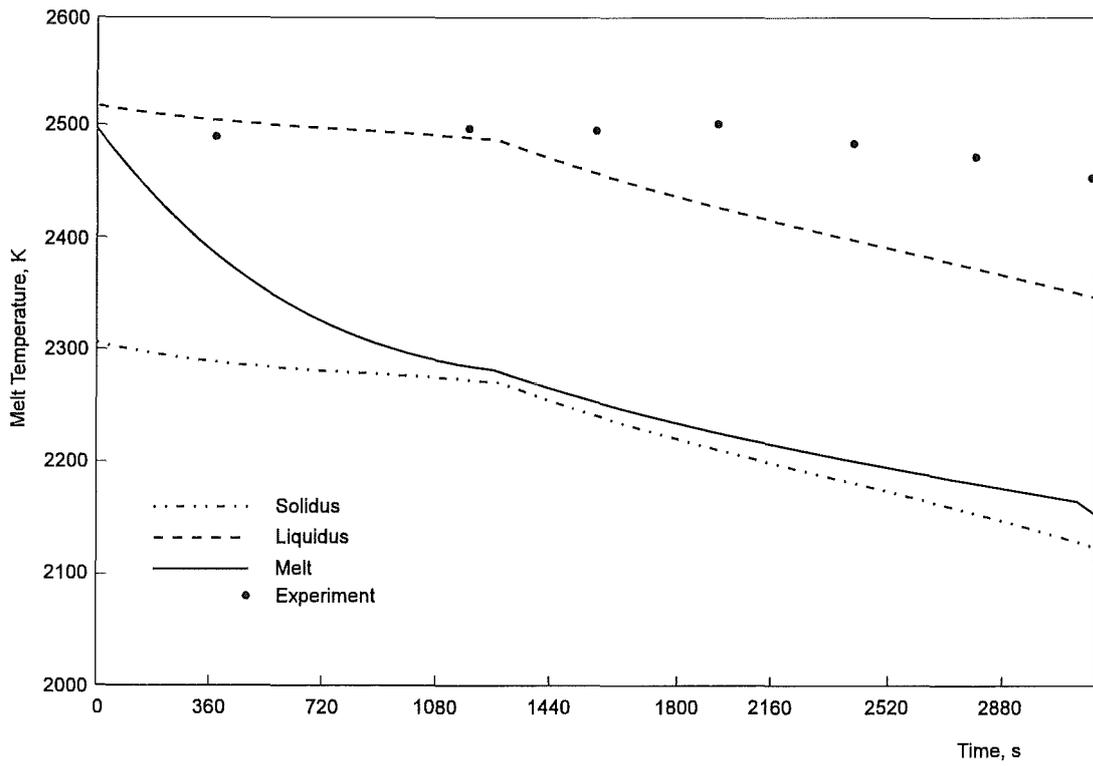


Fig. 13: Melt temperature (new results)

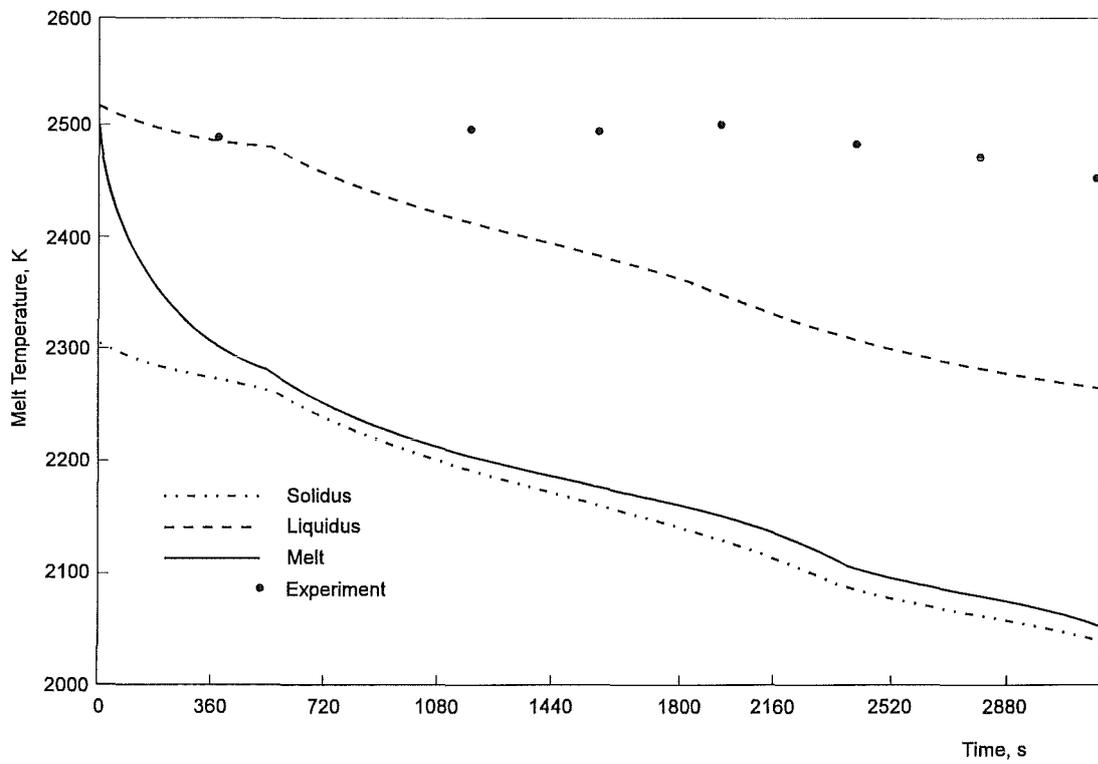


Fig. 14: Melt temperature (previous results)

IV.4 Experiment ACE-L8

The crucible of the test under consideration [15] consists of a limestone concrete. The metallic Zr is included in so-called concrete/metal inserts which are 4.3 cm thick in this case. The behaviour of the limestone concrete under a thermal attack is completely different from the behaviour of a siliceous type of concrete which serves as a basis for the modeling WECHSL. Nevertheless, it is possible to perform a WECHSL simulation.

Both WECHSL versions give a good agreement of the calculated evolution of the concrete erosion (Fig. 15) with experimental findings. The previous WECHSL version slightly overpredicts the erosion at the beginning of the interaction and is in accordance with the final experimental erosion depth. The new version underestimates the final erosion depth by about 20 %.

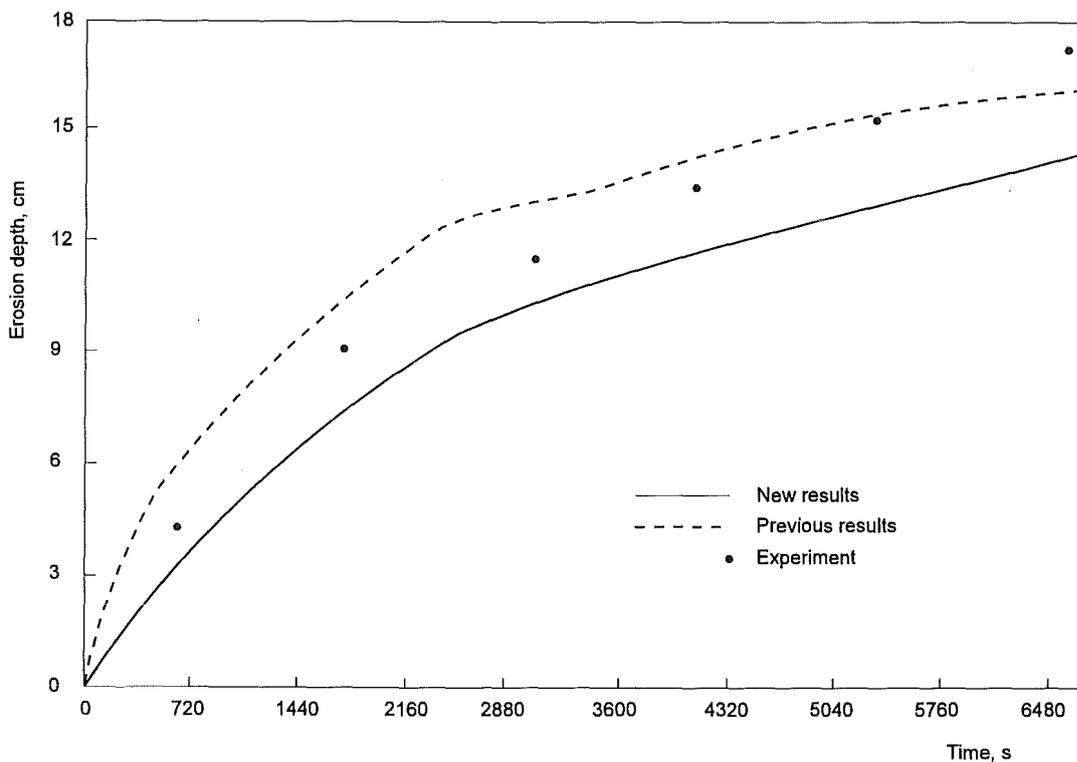


Fig. 15: Erosion depth

There is the same tendency to underpredict the measured melt temperature as in the cases considered above (Fig. 16 and 17).

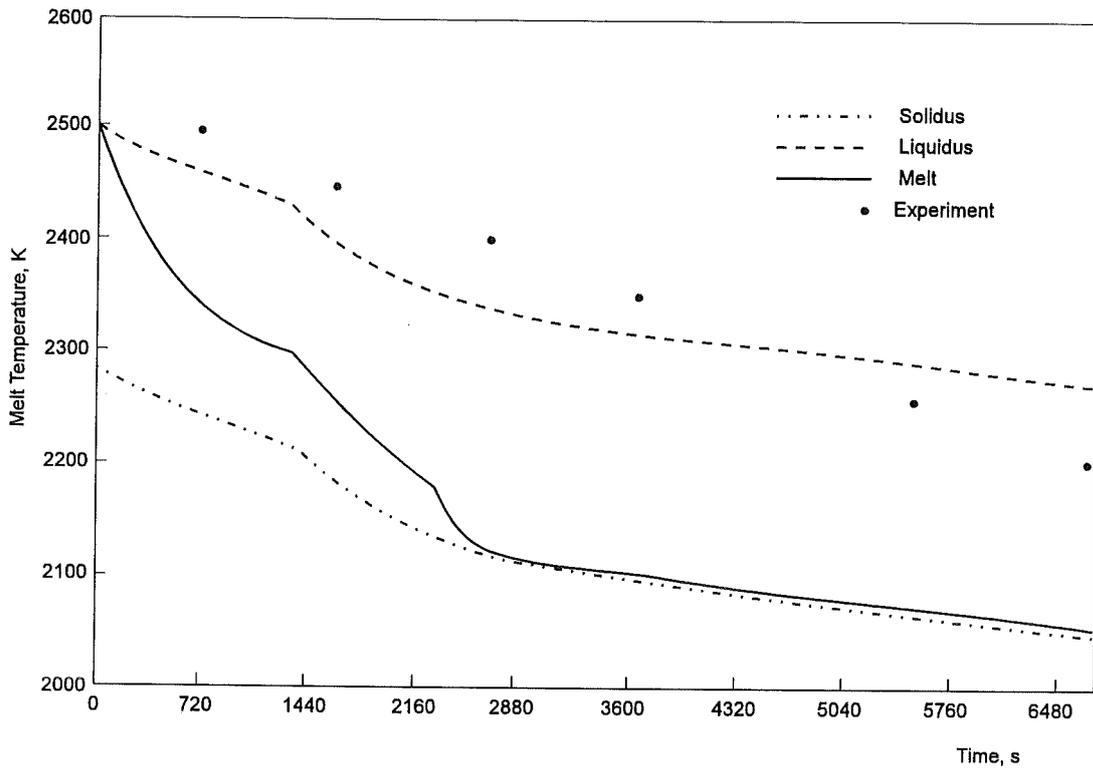


Fig. 16: Melt temperature (new results)

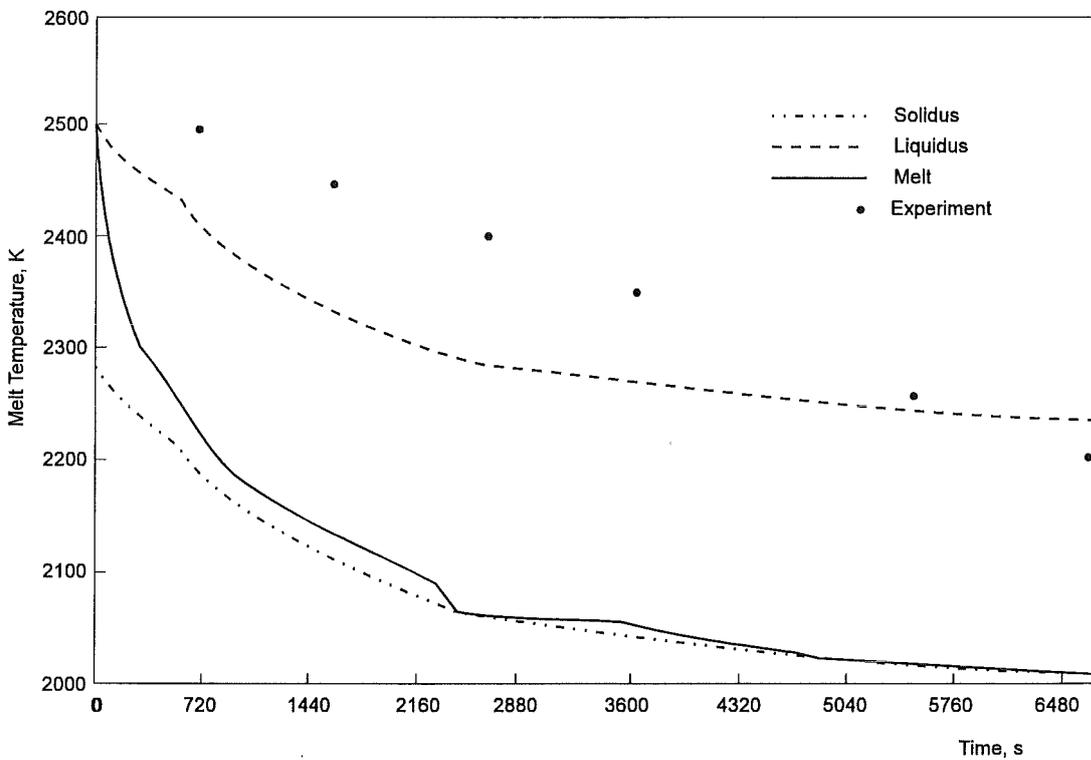


Fig. 17: Melt temperature (previous results)

IV.5 SURC-1 experiment

The crucible of this experiment [16] consists also of a limestone concrete. Both WECHSL versions grossly underpredict the erosion depth during the first 3600 s of the melt/concrete interaction. After this time period the previous prediction follows the measurements and the new results slightly underestimate the experimental data (Fig. 18).

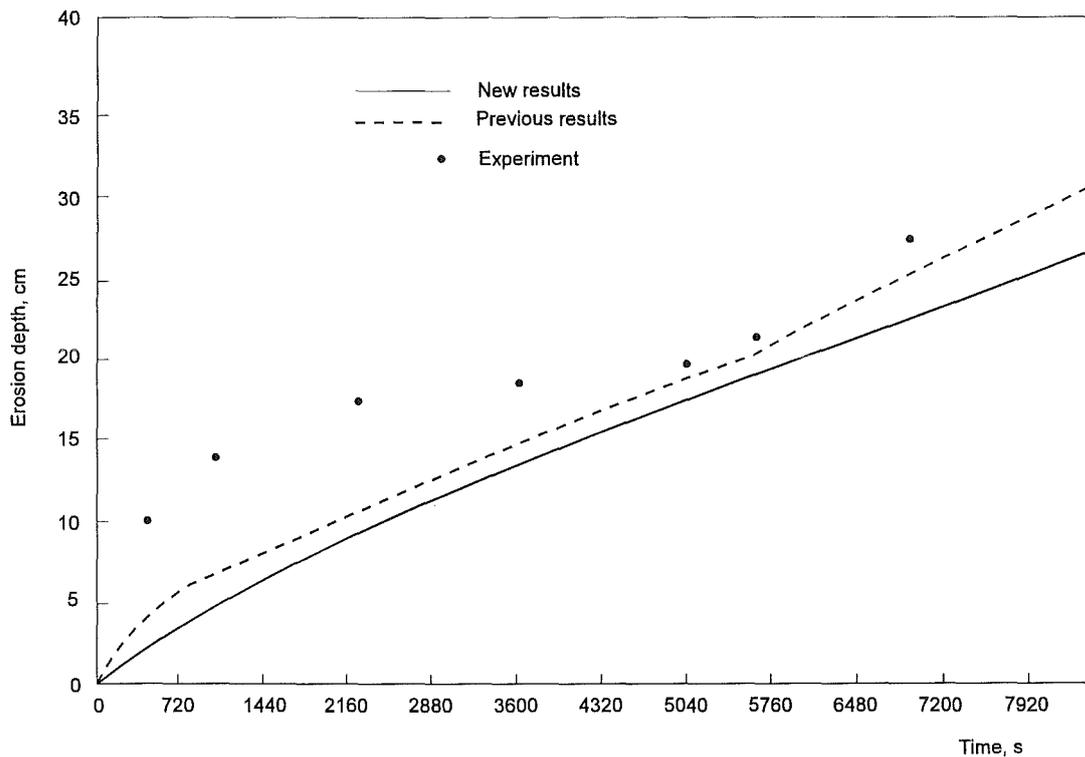


Fig. 18: Erosion depth

The WECHSL calculations for the SURC-1 experiment differ from the predictions of the melt temperature for the ACE experiments. Both WECHSL versions underestimate the melt temperature during the first 5000 s of the interaction but predict the final melt temperature of the experiment very well (Fig. 19, Fig. 20).

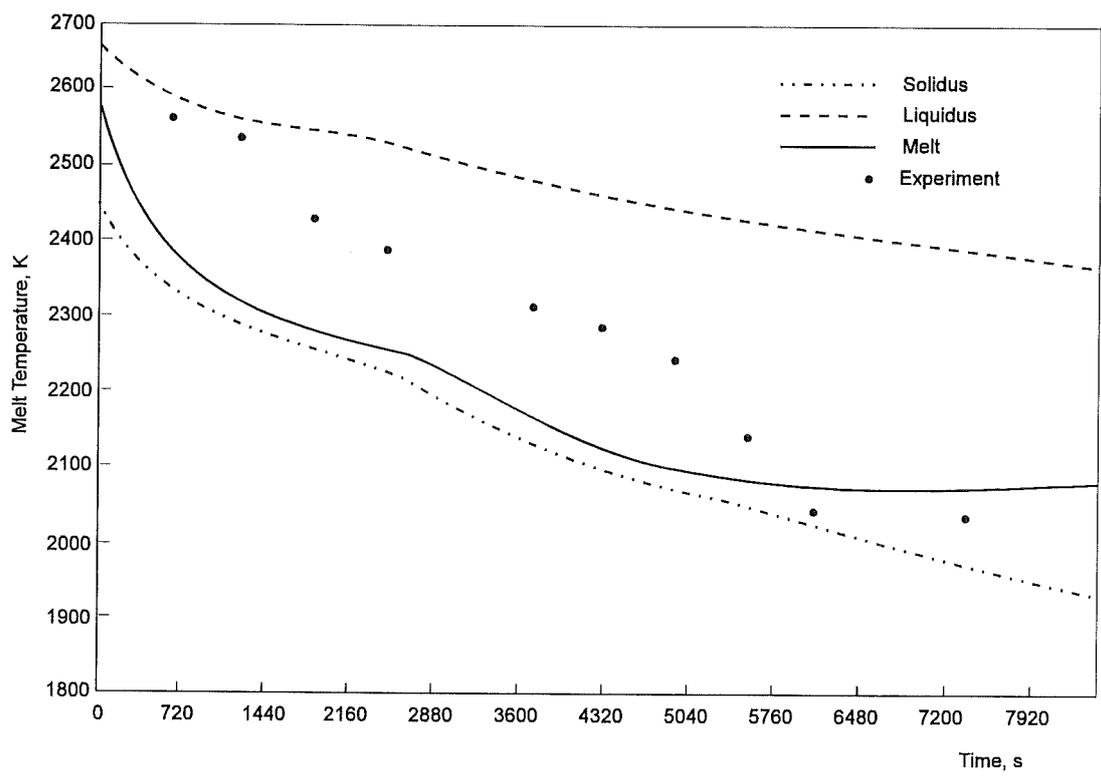


Fig. 19: Melt temperatures (new results)

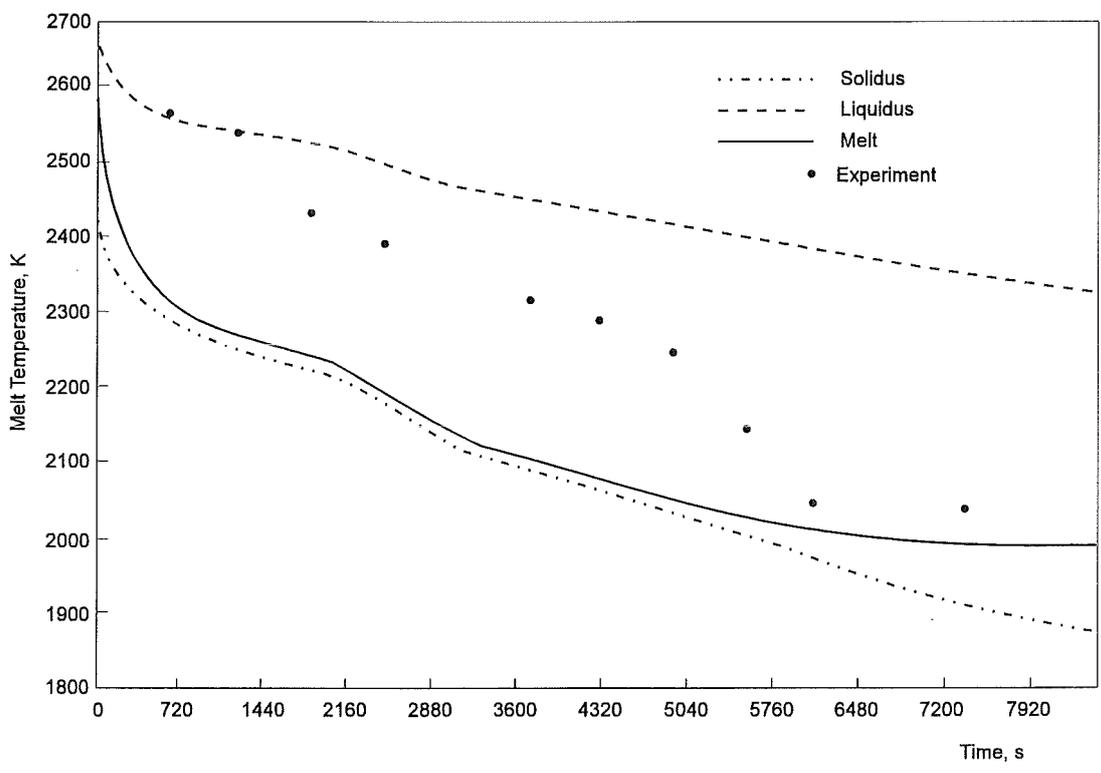


Fig. 20: Melt temperatures (previous results)

IV.6 SURC-2 experiment

The erosion of the basaltic SURC-2 [17] concrete is predicted very well during the first hour of the interaction but the final erosion depth is overpredicted (Fig. 21).

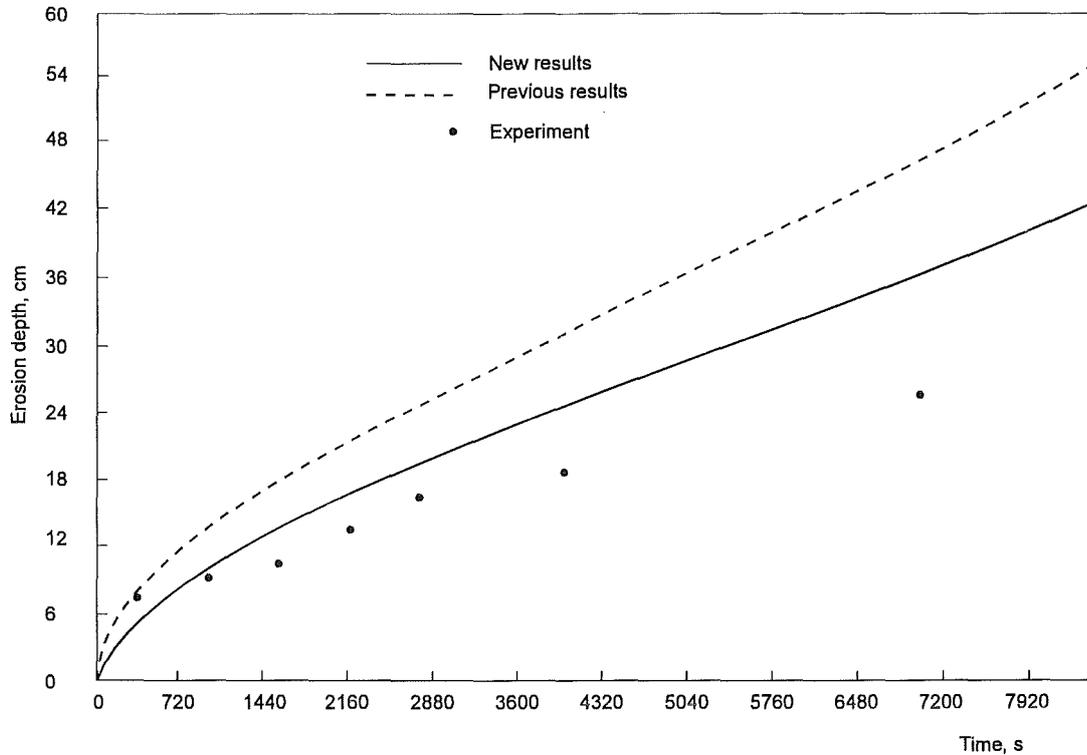


Fig. 21: Erosion depth

The new prediction of the melt temperature agrees very well with experimental data during the whole time period of the melt/concrete interaction whereas the former results underpredict the measurements considerably (Fig. 22, Fig. 23).

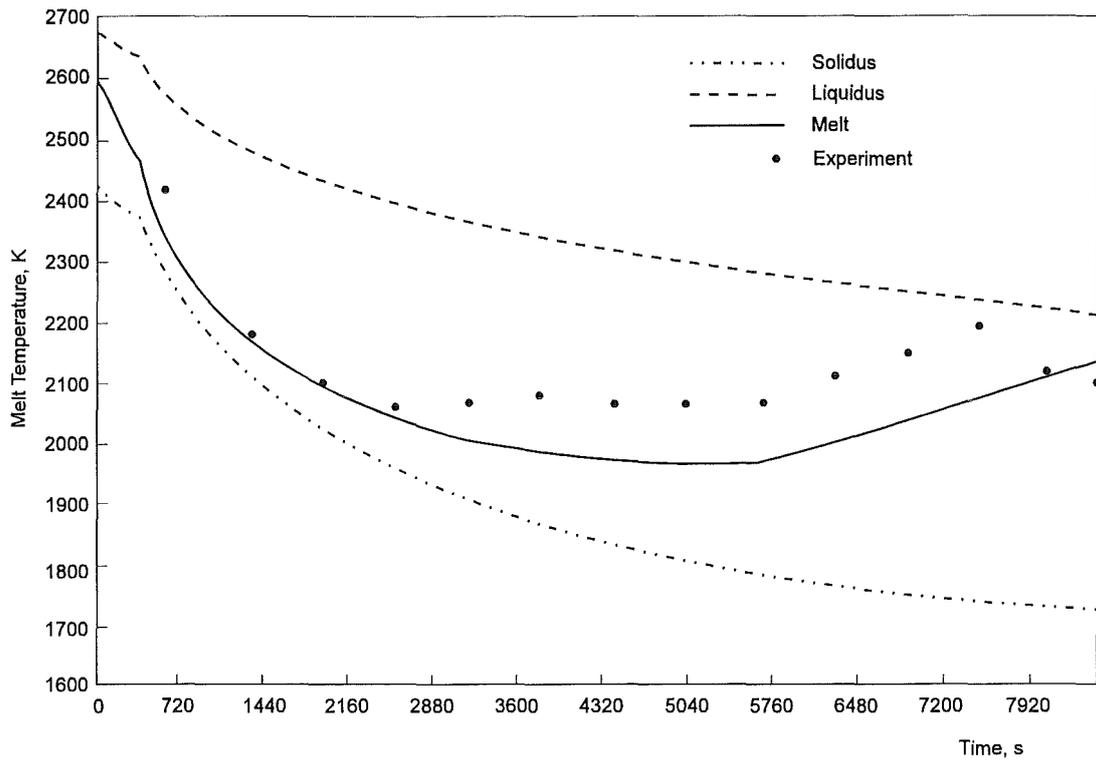


Fig. 22: Melt temperatures (new results)

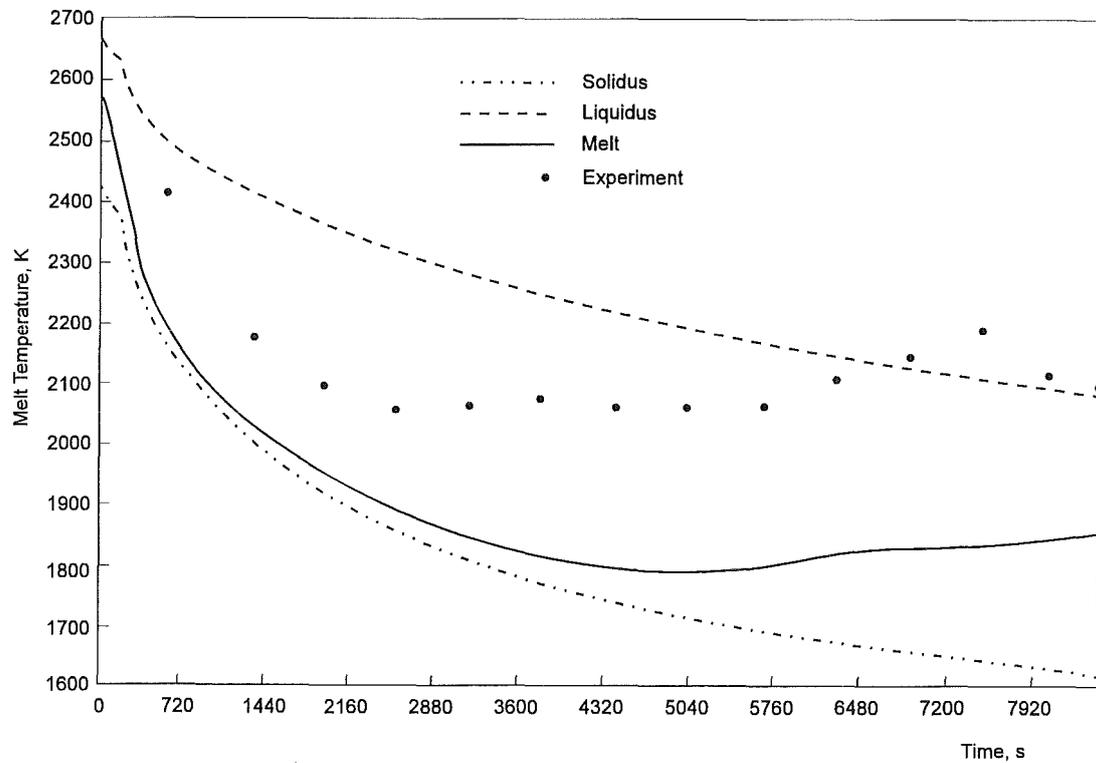


Fig. 23: Melt temperatures (previous results)

V. Conclusions

The WECHSL analysis of oxidic melt MCCI experiments has been improved by including the effect of the melt temperature on the viscosity dependence of the heat transfer modeling together with a correlation which reduces the thermal conductivity of the oxidic melts to allow for the porosity of the debris. A satisfactory prediction was derived for the concrete erosion for all experiments. Good estimates for the melt temperatures were obtained for the SURC experiments whereas the predicted temperature levels for all ACE experiments are too low compared to experimental data. However, there are still open questions concerning the reliability of the experimental data (net power input to the melt, beginning of the metal/concrete inserts ablation, accuracy of the temperature measurements).

Viscosity data which have become available recently [5], exceed the values predicted by the improved viscosity model in WECHSL. These data showed also a non-Newtonian behaviour which leads to additional difficulties in viscosity modeling. Such a behaviour was not observed in former viscosity measurements [3], therefore, this point needs further clarification.

It is hoped that the recently established ACE extension project will clarify these uncertainties and lead to a better understanding and consequently, to further improvements in the modeling of complex oxidic melts.

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