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THE EFFECT OF METAL CONTENT IN MOLTEN CORIUM ON CONCRETE ABLATION: NEW INSIGHTS WITH THERMOCHEMICAL APPROACH

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1. ABSTRACT

Nuclear power is a reliable source of energy; however, nuclear accidents could lead to catastrophic environmental effects. According to the current level of knowledge, the Molten Corium Concrete Interaction (MCCI) analysis is still uncertain and the effect of metal content on ablation rate has not been thoroughly investigated. This study therefore investigates the effect of metal content such as zirconium, chromium, and iron on concrete ablation and damage from a practical perspective. This research presents the use of CORQUENCH with related chemical reactions to model the molten corium, its various compositions, and its interaction with concrete using different heat transfer models. The developed model was validated against experimental data and the results showed that the metal content of corium has a pronounced effect on the depth of ablation and reactor integrity. In addition, it was found that corium containing zirconium was very reactive, partly because it produces significant quantities of carbon monoxide/hydrogen and partly because of the inhibition of the formation of ablation controlling gases such as carbon dioxide. It was found that the concrete ablation mechanisms during MCCI are highly case-dependent on the concrete solidus, liquidus and ablation temperatures, respectively.

2. INTRODUCTION

A nuclear accident could lead to the formation of molten corium in the nuclear reactor. This molten corium is a high-temperature mixture of molten nuclear fuel, cladding, and structural elements. The molten corium could penetrate through the nuclear reactor pressure vessel and could cause concrete ablation basement melt-through, via the process of molten corium concrete interaction (MCCI). The intensity of nuclear accidents and the release of nuclear radiation into the environment are controlled by the characteristics and composition of molten corium. The properties and composition of corium vary due to several factors such as sources of heat in the molten corium, heat generated by the fission reaction, the heat produced by the oxidation of molten corium, and the heat emitted by the chemical reactions. Admittedly, a lot of work has been done in past on the nuclear thermal hydraulics [1]-[6]. However, mitigating measures towards reactor safety and contingency plans have not received the much-needed attention. The development of strategies to mitigate MCCI and different factors affecting it is thus a relatively newer field compared to neutronics and thermal-hydraulics.

In this study, our objective is to develop a model and determine the effect of corium composition. Using different chemical reactions and their related equilibrium constants according to the composition of molten corium. This approach assisted to develop the relationship between the mass flow rate of oxidizing agents (CO_2, H_2O, and SiO_2) from concrete into the molten corium. To model the different chemical reactions, it is assumed that first zirconium oxidizes, and then it is followed by silicon, chromium, and iron respectively. The molten corium energy conservation equation includes the terms of energy source and sinks to the concrete and overlying atmosphere [7] as shown below:

$$e = \frac{1}{m}\{-me + \chi_{m,UO_2} m q_{dec} - A_b h_t (T_m - T_t) - A_b h_b (T_m - T_b) - A_s h_s (T_m - T_s) + E_{H_2O}\left(A_b m_{b,H_2O} + A_s m_{s,H_2O}\right) + E_{CO_2}\left(A_b m_{b,CO_2} + A_s m_{s,CO_2}\right) + E_{SiO_2}\left(A_b m_{b,SiO_2} + A_s m_{s,SiO_2}\right) + \left(1 - \chi_{cng,g}\right)\rho_{con} e_{con,d}(A_b \eta_b + A_s \eta_s) - A_b(\langle \rho e \rangle_b \delta_b + \langle \rho e \rangle_t \delta_t) - A_s \langle \rho e \rangle_s \delta_s - X_c \rho_{sol,m} e \eta_s (\delta_b + \delta_t) - \langle \rho e \rangle_s A_C H_m - A_b m_{ent} e + m_{core} e_{corem}\} \quad (1)$$

During the interaction of molten corium with the concrete, the viscosity of the molten corium increases due to the inclusion of concrete into the molten corium. The viscosity of the molten corium during MCCI can be calculated by the numerical models given as follows [8].

$$\mu = \mu_c \left(1 - \frac{V_{sol}}{V_{sol,max}}\right)^{-2.5 V_{sol,max}\left(\frac{\mu_{d+0.4\mu_c}}{\mu_d + \mu_c}\right)} , \quad (2)$$

$$\mu = \mu_c \left[\frac{1+\frac{1}{2}\frac{V_{sol}}{V_{sol,max}}}{\left(1-\frac{V_{sol}}{V_{sol,max}}\right)^4} \right] \quad , \tag{3}$$

Where $V_{sol,max}$ is the maximum fraction of solid-phase packing and it is used to determine molten corium viscosity, μ is the viscosity, μ_c is the viscosity of continuous phase, μ_d is the viscosity of dispersed phase and they can be estimated as

$$\mu_c = V_{liq,m}\mu_m(T) + V_{liq,o}\mu_o(T), \tag{4}$$

$$\mu_d = V_{sol,m}\mu_m\left(T_{sol,m}\right) + V_{solo,o}\mu_o(T_{sol,o}). \tag{5}$$

It is important to mention that high viscous molten corium decreases the volume and rate flow of corium that could leak through the cracks formed in the crust. Consequently, the cooling efficiency would be reduced. After the injection of water for quenching the molten corium. The molten corium could be quenched by the following cooling mechanisms such as ingression of water, corium bulk cooling, crust breach, and corium eruption. The bulk cooling of molten corium is a process that will take place before the formation of crust on the surface of the corium. Because as soon as the gas is produced, the MCCI would escalate and the increase in boiling would increase the interface area between corium and water. For extreme conditions, Farmer *et al.*, [9] estimated the overall rate of heat transfer by the injected water and the radial heat transfer coefficient and it is given as

$$q^{"}_{wat} = A\left(\frac{k_e \Delta T_{sat}}{\delta_{fb}} + q^{"}_r\right) \quad , \tag{6}$$

$$h_r = \sigma_{stef}\varepsilon_m\left(T_{t,I}^2 + T_{sat}^2\right)(T_{t,I} + T_{sat}) \quad , \tag{7}$$

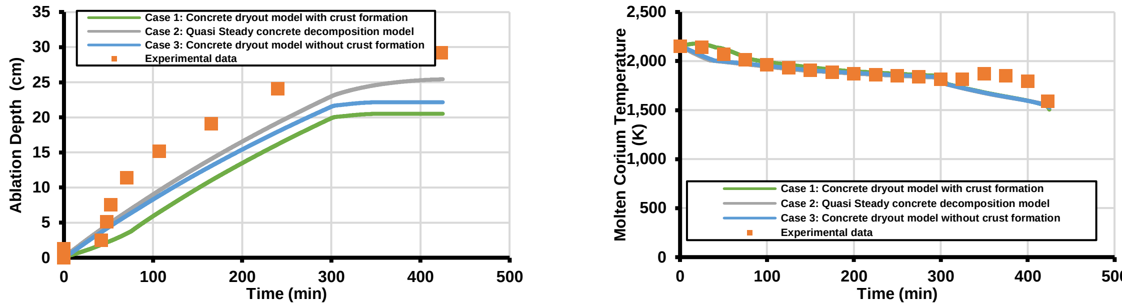
Where δ_{fb} stands for the gas film thickness composed of water vapours and various non-condensable gases produced during corium-concrete interaction, and ΔT_{sat} is the superheat surface comparative to the water saturation point. With the constant flow of water, a stable crust would form and the molten corium will remain insulated under the surface of the crust. Thus, molten corium could only be quenched if the injected water flows through the crust cracks. Thus with the production of gas, the crust would break apart, the ingression of water would become extremely active, and the layer (thermal boundary) between molten corium and crust becomes thinner. This process might improve the transfer of heat due to conduction. Lomperski et al., [10] determined that the overall heat flux for such cases is given as

$$q^{"}_{c,dry} \geq k_{t,c}\frac{(T_{t,frz}+T_{sat})}{\delta_t} + \frac{(Q_{t,c}\delta_t)}{2}\rho_v h_{lv} j \quad . \tag{8}$$

3. MODEL VALIDATION/JUSTIFICATION

In this research, the results of the developed model were compared with experimental work performed on concrete ablation with molten corium used in CCI-2 by [8]. The various concrete and corium properties for each CCI-2 are taken from [7]. The CCI-2 part of the OECD/MCCI test was performed for a 50×50 cm limestone/common sand concrete, with 400 kg of the total mass of the molten corium. The test was executed for 300 minutes and then water was injected. Figure 1 presents the comparison of CCI-2 experimental data with developed models. Figures 1(a) & (b) present the location of the ablation front and the molten corium temperatures respectively. Here the ablation depth is used for comparison, as it determines the integrity of the reactor containment. From Figure 1(a), it can be observed that the abrupt concrete ablation takes place as soon as molten corium interacts with concrete at the beginning of the cycle (during the dry phase). However, the developed model shows that ablation takes place from zero and increases linearly to 25.4, 22.2, and 20.5 cm by quasi-steady concrete decomposition model, concrete dry-out model with crust formation, and concrete dry-out model without crust formation, respectively. It can be observed that the results predicted by quasi-steady concrete decomposition model are closer to the experimental data. It is also evident from the behaviour of all the models that the ablation levels off to a plateau with the injection of water.

The model results are also compared with the experimentally measured temperatures of corium as depicted in Figure 1 (b). The latter figure shows an overall good match between experimental and simulation results, especially with the concrete dry-out model with crust formation gives the best match. The analysis of experimental data of CCI-2 shows that molten corium temperature drops promptly in the initial phase that is, its temperature drops sharply after 75 min till water is injected. It can be observed from experimental data presented in Figure 1(b) that corium temperature during the CCI-2 experiment does not decrease sharply after the injection of water. Thus, as soon as the water is injected the temperature of corium drops to 1818k at 300 minutes, and then it increases to 1873 at 350 minutes (for experimental data only). However, the developed model shows that corium temperature drops sharply with the injection of water at 300 minutes to 1818 K, and at 350 minutes it further drops to 1670 K. The analysis of results revealed that the heat flux from the molten corium to water increases rapidly with the injection of water.



(a) (b)

Figure 1: Comparison of (a) axial ablation experimental data (CCI-2) with model prediction, (b) molten corium temperature experimental data (CCI-2) with model prediction

4. RESULTS AND ANALYSIS

An integrated model is developed to simulate the interaction of molten corium and concrete as described in the previous sections of this study. After validation of the developed model, a detailed sensitivity analysis is executed for the composition of molten corium. It should be noted that the composition of molten corium is taken from Farmer [7]. It is important to mention that to analyse the effect of molten corium, we took the experimental corium data from CCI experiments because the composition of corium is the most uncertain parameter as it defines the corium temperature, sources of heat including the generation of heat by fission reaction, generation of heat corium oxidation. Thus, the use of hypothetical data is considered erratic hence one must consider the experimental data for the existence of certain species at high-temperature conditions.

Table 1. Chemical reactions (Oxidation) utilized in CORQUENCH [7]

Chemical Reactions (oxidation)	Constituent i	$\gamma_i^{SiO_2}$	$\gamma_i^{H_2O}$	$\gamma_i^{CO_2}$
Zr+2H₂O→ZrO₂+2H₂ *Zr+2CO₂→ZrO₂+2CO*	*Zr*	$-\frac{F_{cond}(1+F_{Si})M_{Zr}}{2M_{SiO_2}}$	$-\frac{F_{Zr}M_{Zr}}{2M_{H_2O}}$	$-\frac{F_{Zr}M_{Zr}}{2M_{CO_2}}$
	ZrO₂	$\frac{F_{cond}(1+F_{Si})M_{ZrO_2}}{2M_{SiO_2}}$	$\frac{F_{Zr}M_{ZrO_2}}{2M_{H_2O}}$	$\frac{F_{Zr}M_{ZrO_2}}{2M_{CO_2}}$
Gas Phase: *Si+2H₂O→SiO₂+2H₂* *Si+2CO₂→SiO₂+2CO* Condensed Phase: *Zr+SiO₂→ZrO₂+Si(l)* *(T≤2784)* *Zr+2SiO₂→ZrO₂+2SiO(g)* *(T >2784)*	*Si*	$\frac{F_{cond}F_{Si}M_{Si}}{2M_{SiO_2}}$	$-\frac{F_{Si}M_{Si}}{2M_{H_2O}}$	$\frac{F_{Si}M_{Si}}{2M_{CO_2}}$
	SiO₂	$-F_{cond}$	$\frac{F_{Si}M_{SiO_2}}{2M_{H_2O}}$	$\frac{F_{Zr}M_{SiO_2}}{2M_{CO_2}}$
2Cr+3H₂O→*Cr₂O₃+3H₂* 2Cr+3CO₂→*Cr₂O₃+3CO*	*Cr*	0	$-\frac{2F_{Cr}M_{Cr}}{3M_{H_2O}}$	$-\frac{2F_{Cr}M_{Cr}}{3M_{CO_2}}$
	Cr₂O₃	0	$\frac{F_{Cr}M_{Cr_2O_3}}{3M_{H_2O}}$	$\frac{F_{Cr}M_{Cr_2O_3}}{2M_{CO_2}}$
Fe+H₂O→*FeO+H₂* Fe+CO₂→*FeO+CO*	*Fe*	0	$-\frac{F_{Fe}M_{FeO}}{M_{H_2O}}$	$-\frac{F_{Fe}M_{Fe}}{M_{CO_2}}$
	FeO	0	$\frac{F_{Fe}M_{FeO}}{3M_{H_2O}}$	$\frac{F_{Fe}M_{FeO}}{2M_{CO_2}}$

The detailed effect of molten corium composition on ablation depth, surface heat flux, molten corium temperature, molten corium viscosity, hydrogen production, carbon-monoxide production, and carbon dioxide production is shown in Figure 2. The analysis of the composition of different corium in various CCI experiments shows that only CCI-4 has zirconium and iron metal 13.82 kg and 8.97 kg respectively. Figure 2(a) depicts the effect of corium composition on ablation depth during molten corium concrete interaction. It can be observed from the results that the ablation becomes uncontrollable, with the corium composition of CCI-4. The ablation reaches the depth of 139 cm with CCI-4 molten corium composition as shown in Figure 2(a). It is worth mentioning that all the corium as shown in Table 2 contains chromium but the adverse effect occurred only in CCI-4 which contains zirconium and iron. This illustrates that the use of chromium is safe as compared to zirconium and iron. Moreover, water injection has no quenching effect on molten corium. However, for CCI-2, CCI-3, and CCI-5 the ablation reaches the depth of 18.4 cm and stops/plateaus after the injection of water at 300 minutes. These findings confirm that the metal content of molten corium plays a very important role during MCCI. If we design the reactor in such a way, the zirconium and iron don't become a part of molten corium. Then it will be easy to contain the spread of molten corium and control MCCI. It is evident that the presence of 13.82 kg of zirconium and 8.97 kg of iron metal in molten corium increased the ablation depth from 18.4 m to 139 m, around 7.55 times increase in ablation. Thus, it is very important to control/design the reactor with minimum content of iron and zirconium.

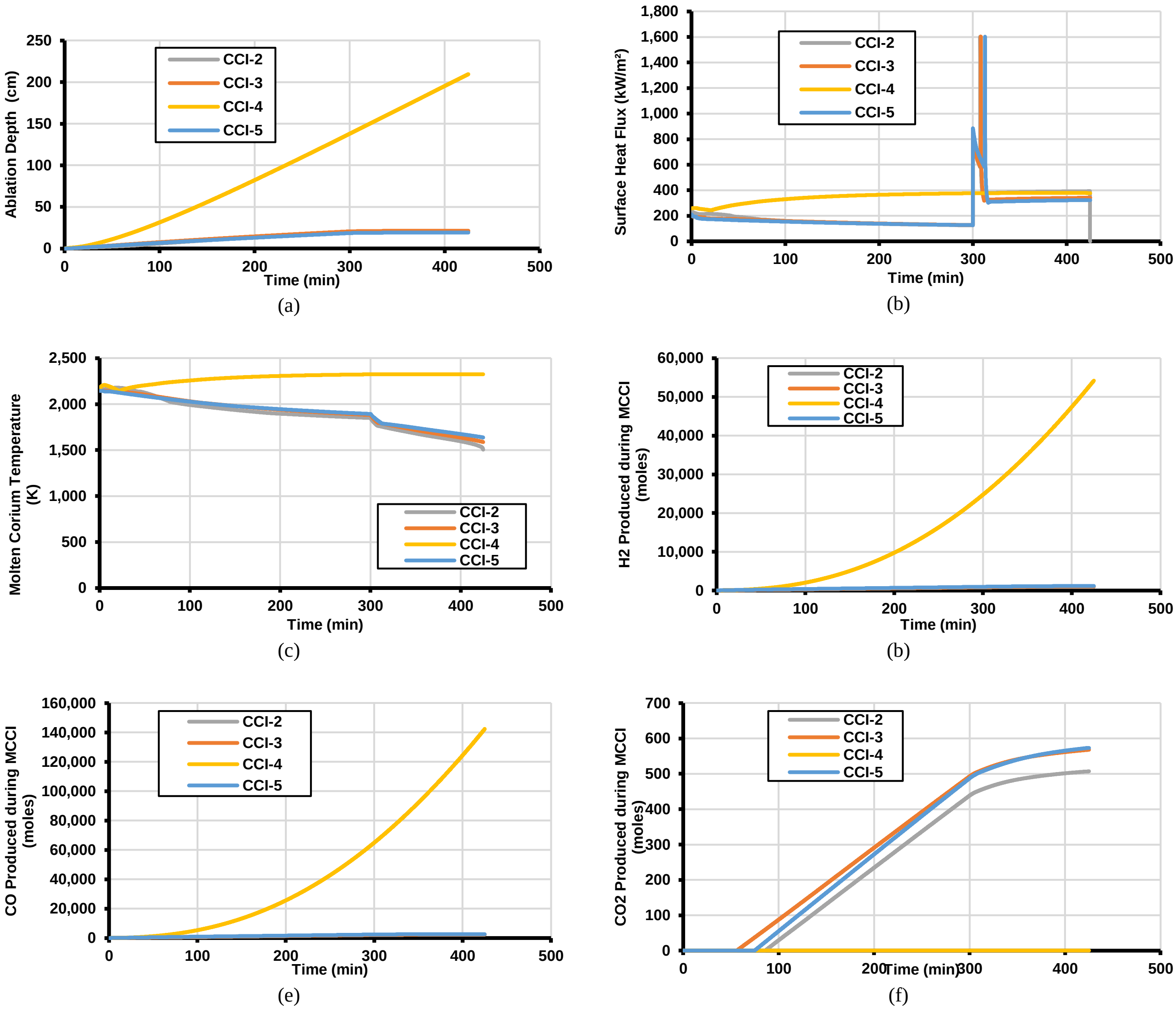


Figure 2: Effect of (a) corium composition on ablation, (b) corium composition on surface heat, (c) corium composition on molten corium thermal behaviour, (d) corium composition on Hydrogen gas produced, (e) corium composition on carbon monoxide (CO) gas produced, (f) corium composition on carbon dioxide gas produced.

Figure 2(b) describes the effect of corium composition on surface heat flux during molten corium concrete interaction. It can be observed from the results that after the injection of water at 300 minutes, we can perceive a peak in surface heat flux for CCI-2, CCI-3, and CCI-5. The surface heat flux peak for CCI-2, CCI-3, and CCI-5 are 1600 kW/m^2. However, for CCI-4 there is no change/peak in the profile of surface heat flux even after the injection of water, and it remains constant at 376 kW/m^2. This behaviour shows that for CCI-2, CCI-3, and CCI-5 the injected water-cools the molten corium instantaneously. However, for CCI-4 the injected water instead of quenching the corium molten augments the temperature of corium by facilitating exothermic reactions and generates a great number of explosive gases such as hydrogen and carbon monoxide as shown in Table 1.

The effect of corium composition on molten corium thermal behaviour is shown in Figure 2 (c). It can be observed that with the increase in time the temperature of molten corium decrease for CCI-2, CCI-3, and CCI-5. However, for CCI-4 the temperature of molten corium increases continuously. The reason behind this increase in molten corium temperature is the thermo-chemical reactions that take place continuously in CC1-4 corium due to the presence of zirconium and iron. It is also evident from Figure 2(c) that after the injection of water the temperature of molten corium for CCI-2, CCI-3, and CCI-5 drops significantly from 1870 K to 1500 K but for CCI-4 it increases from 2320 K at 300 minutes to 2330 K at 350 minutes. This increase in the temperature of molten corium is the reason behind increased ablation as shown in Figure 2(a).

Table 2. Composition of different corium used in CCI experiments

S.No	Constituent	Mass(kg)			
		CCI-2	CCI-3	CCI-4	CCI-5
1	UO_2	242.48	211.41	169.36	332.29
2	ZrO_2	99.6	86.82	64.51	136.47
3	SiO_2	13.56	41.92	12.15	65.9
4	Al_2O_3	1.64	2.4	1.47	3.78
5	MgO	4.56	0.45	4.08	0.7
6	CaO	12.52	8.31	11.23	13.04
7	Cr	25.64	24.06	14.08	37.82
8	Zr	0	0	13.82	0
9	Fe	0	0	8.97	0
	Total	400	375.37	299.67	590

Figure 2(d) through (f) shows the production of various gases and their production during molten corium concrete interaction. It is worthy to mention that all the other parameters such as design, initial corium temperature, concrete composition, and other parameters were kept constant, only the composition of corium is changed as shown in Table 2. It is observed that the presence of metals such as zirconium and iron has a significant effect on the production of explosive gases (H_2 and CO) and explosion/fire-controlling gases (CO_2). Figure 2(d) shows that for CCI-2, CCI-3 and CCI-5 corium composition produces only 1,200 moles of hydrogen. However, for CCI-4 corium composition, 54,000 moles of hydrogen are produced. This significant amount of hydrogen is produced only due to the presence of zirconium and iron. Similarly, Figure 2(e) presents that for CCI-4 corium 142,000 moles of carbon monoxide are produced. However, CCI-2, CCI-3, and CCI-5 produce only 2,710 moles of carbon monoxide. Figure 2(f) displays the production of carbon dioxide with various coriums. The CO_2 is generally considered as the gas that mitigates the concrete ablation, decreases the temperature of molten corium, and increases the viscosity of corium. It can be observed that CCI-2 corium produces 500 moles of CO_2, for CCI-3 and CCI-5 corium, 573 moles of carbon dioxide are produced. However, for CCI-4 the amount of CO_2 generation is zero moles. It occurred because the produced carbon dioxide is converted into carbon monoxide gas (the reactions are described in Table 1). Therefore, our details and comprehensive analysis show that the presence of zirconium and iron is very critical in the conversion of explosion/fire controlling gas (CO_2) to explosive gas (CO). Therefore, it is important to design the reactor such that the concentration of metals especially zirconium and iron is reduced to the minimum.

The results presented in the previous paras showed that the composition of corium would cause a severe nuclear accident if the corium has a composition of CCI-4. The analysis showed that Case-3 has two extra metals in comparison to other metals. These metals are zirconium and iron. Therefore, here section the effect of zirconium and iron presence in the molten corium and its influence on MCCI is obtained through running the developed model in Corquench while considering four different scenarios:

1) With Fe and Zr
2) Without Fe and Zr
3) With Fe and without Zr
4) Without Fe and With Zr.

The different parameters analysed that would be affected by MCCI includes ablation depth, molten corium temperature. These parameters would affect the surface heat flux, corium viscosity, production of hydrogen, carbon monoxide, and carbon dioxide. The determined parameters these four scenarios and the results are depicted in **Fig. 3**. It is worth mentioning here that all the melt/corium cases presented in **Figs. 3** contain iron. However, the confrontational effect occurred only in those events of CCI-4 which contains zirconium. This shows that the use of iron is a safe as compared to zirconium. Moreover, from this figure, one can note that the results of simulation with considering zirconium and those without Fe are the almost same where the profiles of all parameters for these two scenarios (1 and 3) are in a match. Nevertheless, the case without zirconium while considering iron shows a normal profile. These results show that molten corium concrete interaction in the presence of zirconium could cause more severe consequences than containing iron. Therefore, it is essential to consider the presence of zirconium in safety assessment for MCCI.

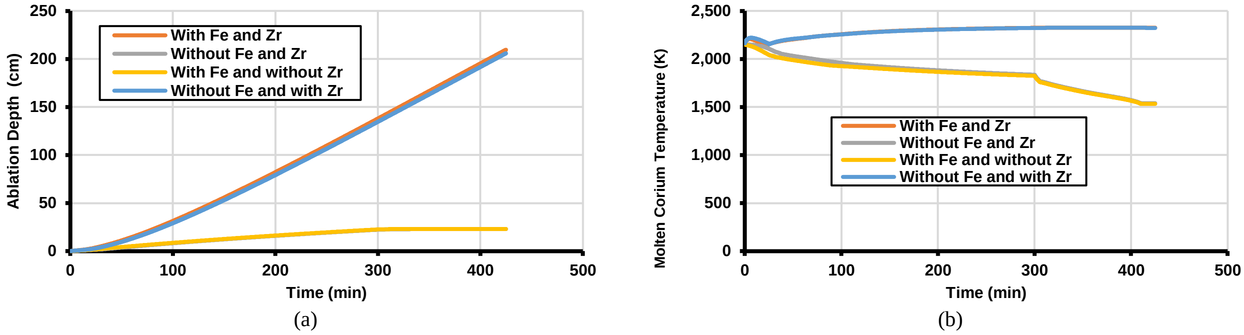


Figure 3: Effect of Fe, Zr, and their various combinations on (a) ablation depth during MCCI at 425 minutes (b) molten corium temperature during MCCI at 425 minutes

5. SUMMARY AND CONCLUSIONS

This work emphasized the importance of thermochemical modeling to characterize the effect of corium composition, especially its metal content. A model was developed and benchmarked against experimental data. It was shown that the model could be used as an effective tool to predict the molten corium concrete interaction before and after a nuclear plant accident. We analysed the change in ablation depth in response to the effect of melt/corium composition with four different corium constituents and performed a detailed sensitivity analysis of two metals in the worst-case scenario. It is concluded that the corium composition is the foremost controlling factor to mitigate ablation in case of nuclear accidents. Our findings show that the concentration of zirconium has a significant role during molten corium concrete interaction. The percentage of zirconium was limited to just 4.6 % of the total mass of corium. However, its presence increased the ablation depth from 18.5 cm to 139 cm. Moreover, the viscosity of corium remains low in the presence of zirconium due to the constant increase in corium temperature because of continuous thermochemical reactions.

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REFERENCES

[1] Benhamadouche, S., Afgan, I., Manceau, R., 2020. Numerical Simulations of Flow and Heat Transfer in a Wall-Bounded Pin Matrix. Flow Turbu. Comb. 104 (1): 19-44.
[2] D'Auria, F., Hassan, Y., 2021. Challenges and concerns for development of nuclear thermal-hydraulics. Nucl. Eng. Desig. 375: 111074.
[3] Kahil, Y., Benhamadouche, S., Berrouk, A. S., Afgan, I., 2019. Simulation of subcritical-Reynolds-number flow around four cylinders in square arrangement configuration using LES. Euro. J. Mech. - B/Fluids. 74: 111-122.
[4] Khurshid, I., Amidu, M. A., Addad Y., Imran, A., 2021. A New Insight into Molten Corium Concrete Interaction with Concrete Ablation Analysis for Mitigation Scheme. International Conference on Nuclear Engineering 85253, V002T07A028
[5] Kim, H.J., Kwon, S., Choi, J., Cho, J.Y., 2019. Concrete ablation analysis for molten corium-concrete interaction mitigation strategy. Ann. Nucl. Ener. 132: 615-627.
[6] Khurshid, I., Imran, A., Amidu, M. A., Addad Y., 2022. Influence of corium temperature, concrete composition and water injection time on concrete ablation during MCCI: New insights. Progress in Nuclear Energy, 144, 104102.
[7] Farmer, M.T., 2018. The CORQUENCH Code for Modeling of Ex-Vessel Corium Coolability under Top Flooding Conditions, Code Manual – Version 4.1b, No. ANL-18/22. Argonne National Lab, Argonne, IL, United States.
[8] Farmer, M.T., Lomperski, S., Basu, S., 2005. The results of the CCI-2 reactor material experiment investigating 2-D core-concrete interaction and debris coolability. In 11th Int. Topical Meeting on Nucl. Reactor Thermal-Hydraulics (NURETH-11), Avignon, France.
[9] Farmer, M.T., Spencer, B.W., Kilsdonk, D.J., Aeschlimann, R.W., 2000. Results of MACE core coolability experiments M0 and M1b. No. ANL/RE/CP-100964 at Argonne Natl. Lab.
[10] Lomperski, S., Farmer, M.T., Basu, S., 2006. Experimental investigation of corium quenching at elevated pressure. Nucl. Eng. Des. 236: 2271-2280.