Implementation Status of CONTAIN-LMR Sodium Chemistry Models into MELCOR 2.1

David L.Y. Louie[1], Larry L. Humphries[1], Matthew R. Denman[1]

[1]Sandia National Laboratories, Albuquerque, New Mexico, U.S.A.

E-mail contact of main author: dllouie@sandia.gov

Abstract. Efforts to integrate the CONTAIN-LMR sodium physics and chemistry models into MELCOR 2.1 and CONTAIN 2 are in progress. Testing and results from this implementation of sodium properties are given here. The CONTAIN-LMR code was derived from an early version of the CONTAIN code and many physical models that were developed since this early version of CONTAIN are not captured by this early code version. Therefore, CONTAIN 2 is being updated with the sodium models in CONTAIN-LMR in order to facilitate verification of these models with the MELCOR code. Although CONTAIN 2, which represents the latest development of CONTAIN, now contains many of the sodium specific models, this work is not complete due to challenges from the lower cell architecture in CONTAIN 2, which is different from CONTAIN-LMR. This implementation should be completed in the coming year while sodium models from CONTAIN-LMR are integrated into MELCOR. Both spray fire and pool fire models from CONTAIN-LMR have been implemented into MELCOR. In the coming year, the atmosphere chemistry and sodium-concrete interaction models from CONTAIN-LMR will be finished. For testing, CONTAIN decks have been developed to verify and validate the models.

Regarding implementing the sodium models into MELCOR, a new sodium chemistry package (called NAC) was created. Both spray fire and pool fire models from CONTAIN-LMR have been implemented and implementation of the atmosphere chemistry model from CONTAIN-LMR has begun. ABCOVE AB-5, a spray fire experiment input deck, was created to test MELCOR. An input deck for testing the pool fire chemistry model is also being developed. The available results of the validations from these two models, including a code-to-code comparison with CONTAIN-LMR are provided here**.**

Key Words: MELCOR, CONTAIN, Sodium chemistry models, Sodium reactor safety.

1. Introduction

Licensing of sodium fast nuclear reactor (SFR) development in the United States will require accident analysis code(s) with capabilities to provide reactor designers and regulators with a means to perform containment and source term analyses for sodium reactors. Gap analyses of the ability for computer codes and models in the U.S. to support the licensing of SFRs identified gaps in the current capability to model source terms, sodium technology, accident sequences and initiators [1-3].

MELCOR [4-6] and CONTAIN [7], which have been employed by the U.S. Nuclear Regulatory Commission (NRC) for light water reactor licensing, have been traditionally used for Level 2 and Level 3 probabilistic analyses as well as containment design basis accident (DBA) analysis. In recent endeavours, in part due to increases in containment-reactor pressure vessel coupling through the use of passive safety systems [8-10]. Both codes were developed at Sandia National Laboratories (SNL) for the NRC. To meet future regulatory needs, new models are being added to the MELCOR code to simulate SFRs supported by U.S. Department of Energy. Existing models developed for separate effects codes are also being integrated into the MELCOR architecture. Sodium properties and equations of state (EOS),

such as from the SAS4A code [11-12], have been implemented into MELCOR to replace the water properties and its EOS as reported previously [13]. Due to a successful implementation, additional specific sodium-related models to address DBA can now be implemented into MELCOR. *FIG. 1* shows the sodium chemistry in the containment of a pool type SFR design. Much of the sodium chemistry phenomena (*see FIG. 1*) for the containment have been modelled in CONTAIN-LMR [14-15]. The discussions of the sodium models will be presented in this paper. The initial implementation of the sodium chemistry models from CONTAIN-LMR into MELCOR was successful. The models are currently being tested. To facilitate the code-to-code comparison, CONTAIN2-LMR, which is based on the latest CONTAIN code (CONTAIN 2), has been developed particularly for cases where there were no experiments for validation in addition to CONTAIN-LMR. Although some sodium models existed in CONTAIN 2, the sodium models were incomplete and were not functional. Therefore, an effort was made to upgrade CONTAIN2 code with the sodium models from CONTAIN-LMR (based on an older version of CONTAIN). Note the development of CONTAIN2-LMR is not complete due to challenges from the lower cell architecture in CONTAIN 2, which is different from CONTAIN-LMR. Once completed CONTAIN2-LMR should compare well with MELCOR since many containment models in both CONTAIN 2 and MELCOR are similar. All sodium chemistry models will be implemented into MELCOR and validated by the end of 2017. Note CONTAIN 2-LMR is not for general distribution, and is only used for the code-to-code comparison within SNL.

This paper discusses the progress of the sodium chemistry models from CONTAIN-LMR being implemented into MELCOR 2.1. Then, the paper discusses the progress of implementing the sodium chemistry models into MELCOR. A new package called “NAC” has been created for the sodium chemistry models in MELCOR. Lastly, the paper provides sodium chemistry model validations from various available experiments.

2. Sodium Chemistry Models

This section describes the sodium chemistry models being implemented into MELCOR 2.1 *(see FIG. 1)*. The models include spray fire, pool fire, atmosphere chemistry and sodium-concrete interaction models. Note that both spray and pool fire models have been implemented into MELCOR 2.1, but only the spray fire has been tested successfully. The atmosphere chemistry model has been partially implemented into MELCOR. The sodium concrete interaction model has not been implemented, but will be completed by the end of 2017, along with the rest of the atmosphere chemistry model.

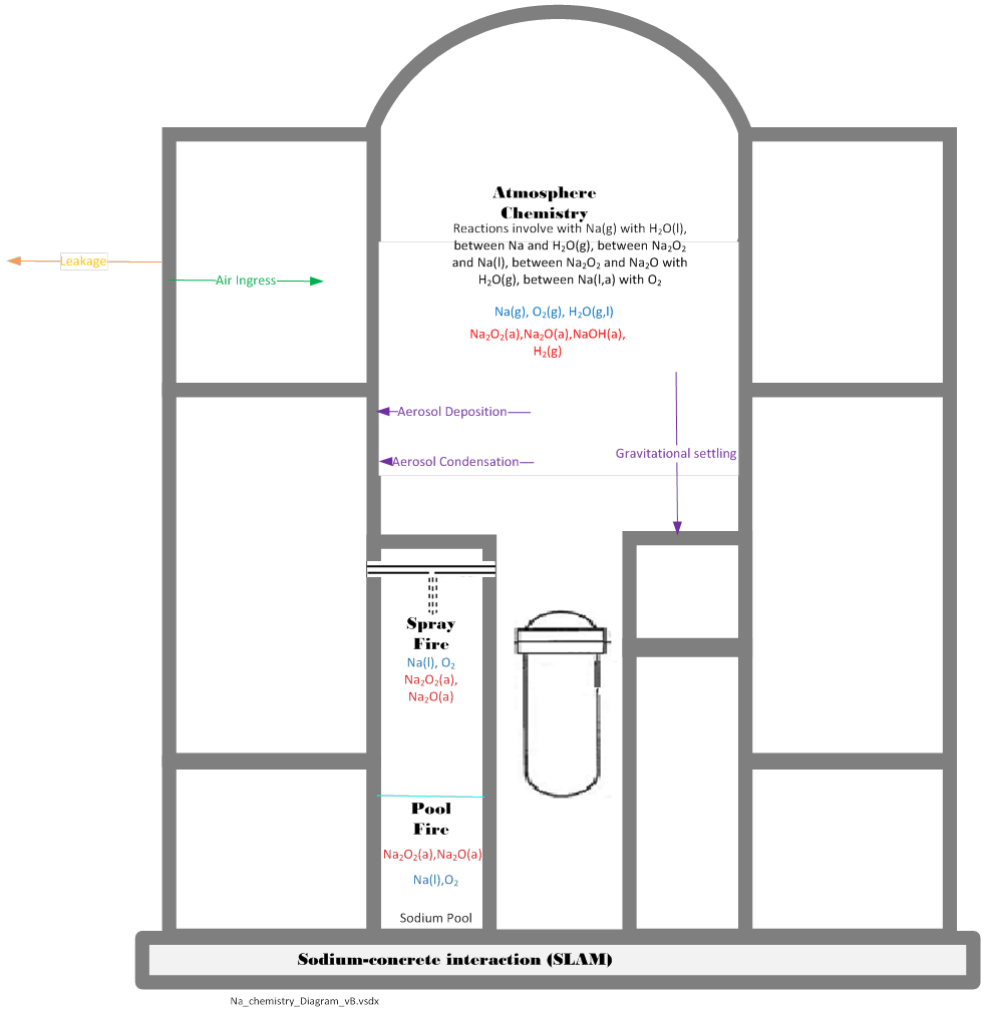


FIG. 1. Sodium chemistry models being implemented into MELCOR 2.1.

2.1.Spray Fire Model

The spray fire model simulates the leak of sodium in the air. This model is based on the phenomenological model used in NACOM, a code developed and tested at Brookhaven National Laboratory [16]. However, unlike NACOM, CONTAIN-LMR did not include the sodium reaction with water vapor. In the spray fire model, an initial size distribution is determined from a correlation using a specified mean droplet diameter. This correlation is based on the partitioning of the injected sodium spray source among eleven discrete droplet-size classes according to the Nukiyama-Tanasama correlation [16]. An assumption is used to state the trajectory of the droplets, which is assumed to have a downward flow with a terminal velocity. The combustion rate of the spray fire is integrated over the droplet's fall to obtain the total sodium burned mass (as functions of droplet size), fall velocity and atmospheric conditions. In the spray fire model, the two chemical reactions of sodium droplets and oxygen in the air are:

Monoxide: $2\ Na + 0.5\ O_2 \rightarrow Na_2O$ (1)

Peroxide: $2\ Na + O_2 \rightarrow Na_2O_2$ (2)

The combustion energy is computed based on the mole fraction of sodium ($F_{peroxide}$) to peroxide (Na_2O_2) as given by the following correlation:

$$S=(1.3478\cdot F_{peroxide})/(1.6957-0.3479\cdot F_{peroxide}) \quad (3)$$

Heat combustion, E_{spray} (J) is then calculated as

$$E_{spray} = (1-S)\cdot 9.1797\times 10^{6} + S\cdot 10.46\times 10^{6} \quad (4)$$

The duration of this sodium source and the available oxygen determines the combustion time and the amount of the by-products (Na_2O and Na_2O_2 as aerosols) and reaction heat to be generated. If a droplet of a given size is not predicted to burn completely, a temporal, numerical integration of the droplet fall is performed (based on droplet terminal velocity). The time increment for the integration is taken as 1/8 of the fall time initially determined. Following each time increment of integration for the combustion equation, a resulting droplet diameter is determined for a new droplet terminal velocity. The combustion heat is transferred to the atmosphere. The process continues until the droplet is either consumed or reaches the floor, forming a pool.

2.2.Pool Fire Model

Pool fire models the accumulation of the sodium on the containment floor in the air environment. This model was taken from the SOFIRE II code developed empirically from pool fire experiments [17]. Reactions (1) and (2) are also considered in this model. However, the model reaction is given as:

$$(1+f_1)\cdot 2\cdot Na + O_2 \rightarrow 2\cdot f_1 \cdot Na2O + (1-f_1)\cdot Na_2O_2 + q \quad (5)$$

Where f_1 = fraction of total O_2 consumed that reacts to form monoxide and q = 9.0454×10^{6} J/kg of monoxide and 1.09746×10^{7} J/kg of peroxide. The above reaction requires oxygen in the air to diffuse to the sodium pool. This diffusion is given by:

$$D = 6.4315\times 10^{-5}\, T_{film}^{1.823}/P \quad (6)$$

Where T_{film} = average temperature of the pool and atmosphere (K), and P = system pressure (Pa). Although the CONTAIN-LMR manual [14] describes the heat transfer model for the sodium pool, the appropriate implementation of this model into MELCOR is still being investigated; therefore, it is not documented here further. Similar to the spray fire model, the by-products of the pool fire model are the aerosols of Na_2O and Na_2O_2.

The pool fire model requires the allocation of the amount of the products and reaction energy to the pool and to the atmosphere layer of the cell. Thus, additional fractional inputs must be provided. The fractional inputs include:

- f_1 is the fraction of total O_2 consumed that reacts to form monoxide
- f_2 is the fraction of sensible heat from the reaction to the pool. The remainder will be directed to the atmosphere layer of the cell.
- f_3 is the fraction of Na_2O product that enters the pool after the fire. The remainder will be directed to the atmosphere as aerosols.
- f_4 is the fraction of Na_2O_2 product that enters the pool as a solid after the fire. The remainder will be allocated to the atmosphere as aerosols.

2.3.Atmosphere Chemistry Model

Atmosphere chemistry models the interactions of the sodium aerosols, vapors and deposits in the atmosphere. In addition to the reactions (1) and (2) above, the atmosphere chemistry model includes additional reactions with water:

$$Na + H_2O\,(l) \rightarrow NaOH + 0.5\cdot H_2 \quad (7)$$

$$2\ Na + H_2O\ (g) \rightarrow Na_2O + H_2 \quad (8)$$

Reaction (7) is assumed to occur only for liquid-phase water and sodium in contact with an aerosol particle, mingling aerosol deposits and condensate films on surfaces. Because the water is required to be liquid, the experimentally observed inhibiting effect of oxygen on reactions of water vapour and sodium is assumed to be inapplicable. This requirement assumes that either the temperature is relatively low (below the critical point of water) or the presence of liquid water is traceable to numerical effects and the amount is not significant. As shown in this reaction, hydroxide is expected to be the principal reaction product with water at low temperatures or with excess water. Conversion from hydroxide to monoxide is not modelled.

Reaction (8) is used when the phase of water is not liquid. It is appropriate at high temperatures with excess sodium. This reaction is also appropriate when water vapour is present, particularly when there is an excess of water vapour over oxygen. In this case, the water vapour is assumed to react not only with sodium vapour in the atmosphere, but also with sodium in aerosol form or in the form of aerosol deposits or films on surfaces. However, the reaction rate for this reaction at the surface with water is assumed to be limited by the evaporation rate of water from the surface.

Note that reactions (7) and (8) with water dominate in the atmosphere over the reactions (1) and (2) with oxygen. Two additional reactions are considered to occur in the atmosphere, which relate to the reactions of the sodium monoxide and peroxide with water vapour in the atmosphere to form sodium hydroxide:

$$Na_2O + H_2O\ (g) \rightarrow 2\ NaOH \quad (9)$$

$$Na_2O_2 + H_2O\ (g) \rightarrow 2\ NaOH + 0.5\ O_2 \quad (10)$$

Water vapour is assumed to react with aerosol particles and aerosol deposits in that order. Again, the user should note that while the hydroxide is expected to be the principal reaction product with water at low temperatures or with excess water, the possible subsequent conversion of the hydroxide to the monoxide is not modelled if conditions change. The chemical reaction models presented here assume that all reaction heat is retained only by the gases present or by the structures; the models ignore the increase in the heat content of the aerosols or aerosol deposits due to an increase in temperature above the temperature of the formation. The heat generated by the surface reactions is assumed to be deposited at surface nodes of the structures involved. This treatment is regarded as conservative.

2.4.Sodium-Concrete Interaction Model

Sodium-concrete interaction models the chemical reaction of the sodium with concrete. Although the concrete is normally lined with steel to protect against the direct contact of the sodium, there are heat transfers between the liquid sodium and the liners that could potentially heat up the concrete floor, which would cause the concrete to dry out. Both carbon dioxide and moisture released from the concrete can interact with sodium if the liner is penetrated.

This model is based on experiments done at SNL regarding the sodium limestone ablation model (SLAM) [18-19]. SLAM uses a nodalized representation of the concrete with models for heat transfer, water migration, water and CO_2 evolution, and chemical ablation of the exposed concrete surface (*see FIG. 2*). As shown in *FIG. 2*, SLAM consists of three regions. The top region is the pool region, but the nodalization is associated with the boundary layer where the ablation occurs. Below this region is the dry concrete region. As shown in this figure, a number of constituents can be included within SLAM, which includes SiO_2, H_2O,

Na, H_2, NaOH, Na_2SiO_3, Na_2CO_3, Na_2O, CaO, $CaCO_3$, CO_2, graphite, $MgCO_3$, MgO, inerts, steel and UO_2.

In SLAM, a variable, "δ", is the thickness of the boundary layer and dry concrete regions. This variable is subjected to change in terms of increasing or decreasing in the course of a problem. The initial δ is 0.003 m. The dry concrete region increases when the thermal penetration rate of the concrete exceeds the ablation rate and decreases when the converse is true. The bottom region is the wet concrete region where evaporable water may still be found in the concrete as shown in *FIG. 2*.

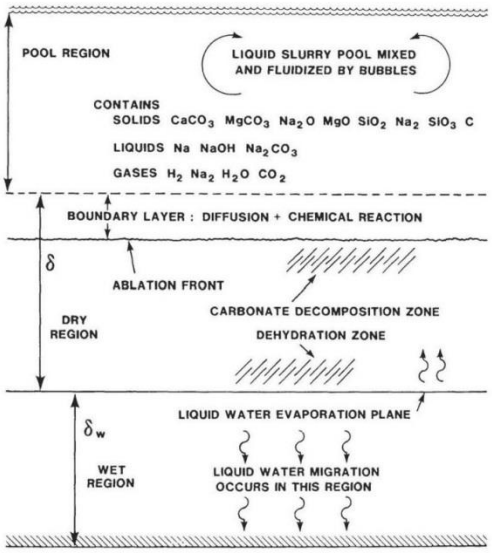


FIG. 2. Schematic Diagram of SLAM [18].

SLAM computes each region (shown in *FIG. 2*) as time passes and penetration occurs, during which each region will change its size and position. The coordinate system of SLAM uses the moving Eulerian system (see more details in [18]). A detailed discussion of this SLAM model can be found in a recent progress report on the implementation of MELCOR/CONTAIN LMR [20].

3. Sodium Chemistry (NAC) Package

To be more efficient and better manage the sodium-related models, a new package titled the "Sodium Chemistry" (NAC) package, has been added to MELCOR. The NAC package handles all sodium related chemistry models for sodium reactor safety applications. This package will utilize the CONTAIN-LMR subroutines from described in Section 2 of this paper. All NAC package subroutines will interface with various package variables for transferring chemistry related processes (both heat and mass), including sodium, oxygen, water and the creation of the by-products of sodium burn resulting from the reactions. A corresponding data structure for each of the implemented models has been created. Two models from CONTAIN-LMR have been integrated into MELCOR: sodium spray fire and the sodium pool fire. The atmosphere chemistry model is partially implemented.

There are several issues related to the implementation of the CONTAIN-LMR models into the NAC package in MELCOR:

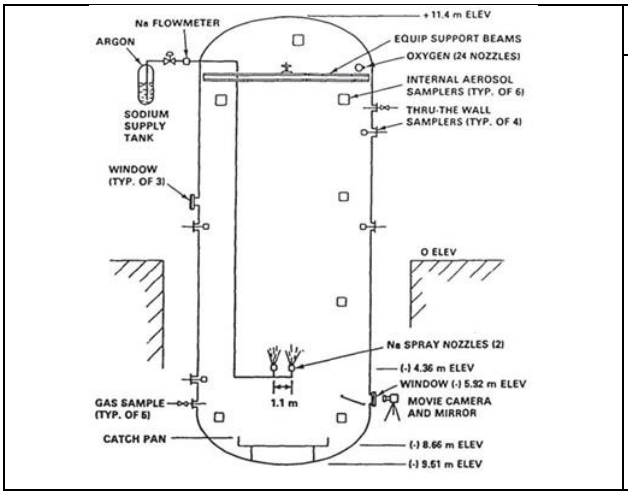
- When replacing the water coolant as sodium coolant, no other condensable can be modeled (i.e., water). Thus the two-condensable option from CONTAIN-LMR may not be easily implemented. Substantial modification to MELCOR architecture may be required.

- To treat the existence of water in the NAC, water is assumed to be a trace element (or aerosol) which does not affect the thermo-dynamic materials. Because the design of the water EOS in MELCOR in such a way, only liquid and vapor phases can be modeled. Thus the solid phase may not be easily implemented in the EOS. There is a similar situation for sodium as a coolant, since the melting temperature of sodium is 371 K. If the ambient atmosphere can be less than the sodium freezing temperature, it poses a challenge to MELCOR. The properties for the liquid phase are extrapolated for sub-frozen temperatures. Coding needs to be modified for 'small' sodium pool.
- Aerosol class re-assignment is required in MELCOR to model sodium as a coolant. Class 2 (Cs) includes Na as the list of elements included. Because water was replaced by sodium in MELCOR, now Class 14 (H_2O) becomes the sodium. A new water class must be created as H2OA.

The initial compilation of the NAC package is finished, including the full implementation of the spray fire and pool fire models. However, only the spray fire model is being tested as described in the next section.

4. Validations

To test the implemented sodium models as described in the previous section about MELCOR, a selection of the sodium chemistry experiments available was required. In addition, the code-to-code comparison, from CONTAIN-LMR/CONTAIN2-LMR would verify if the models were implemented correctly, particularly when no experiment is available for testing the models. The first model to be tested was the spray fire model. Utilizing an existing input deck from the MELCOR 2.1 assessment problems [6], the ABCOVE AB5 test was first [21]. The purpose of this experiment was to provide experimental data for validating the aerosol behavior generated by computer codes during a sodium spray fire scenario. This experiment was conducted at the Containment Systems Test Facility at Hanford Engineering Development Laboratory (see *FIG. 3* for the apparatus setup and spray data). Although the existing MELCOR model was intended for examining the aerosol behavior, rather than the sodium reactions, it can be modified easily to include the sodium spray model parameters. The CONTAIN-LMR model was also developed from this MELCOR model as the code-to-code comparison.

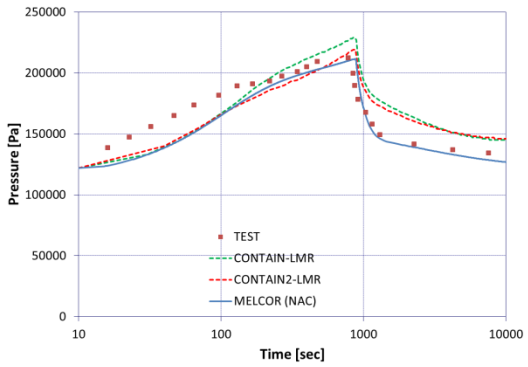


Sodium Spray Data
Na Spray rate (256±15 g/s)
Spray Start Time (13 s)
Spray Stop Time (885 s)
Total Na Sprayed (223±11 kg)
Na Temperature (836.15 K)
Spray Drop Size (1030±50 μm)
Spray size geom. standard deviation (1.4)

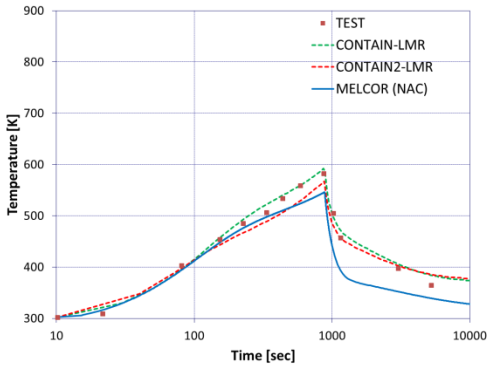
FIG. 3. Schematics and Spray Data of ABCOVE Sodium Spray Fire Test [21]

The initial sodium spray mass of 223 kg at 836 K was injected into a vessel of 852 m^3 filled with air and O_2 makeup. The validation goals were to observe the sodium combustion during

sodium spray, the calculated combustion energy, and aerosol generation. The effect of the pressure and temperature response in the vessel was also of interest. The initial test conditions were 302 K and 0.122 MPa. The sodium spray characteristics are provided in *FIG 3*. Note that the spray was pointed upward, so the current spray fire model will not correctly capture the sodium residence time since the spray points downward. Nonetheless, for this test a spray fall height was assumed to be 5.15 m from the vessel bottom. To sustain the combustion, a continuous flow of oxygen was provided from 60 to 840 s at a total 47.6 m^3. *FIG. 4* shows the preliminary MELCOR (NAC) results of the use of the spray fire model and comparison to CONTAIN-LMR and CONTAIN2-LMR. As shown in *FIG. 4 (a)*, MELCOR predicts the lower values (similar trends to both CONTAIN codes) than the test data to about 1000 s, and matches closely with the test data. As shown in *FIG. 4(b)*, MELCOR follows closely with the test data and both CONTAIN codes to about 600 s before decreasing lower than that of the test data and CONTAIN codes. More analyses will be done to investigate these differences. In terms of the suspended aerosols as shown in *FIG. 4(c)* MELCOR predicts well with the test data, while both CONTAIN codes predicts higher values than the test data after 1000 s. Note that the experiment result indicated that no monoxide was formed and only 60% peroxide and 40% hydroxide were obtained. Therefore, the spray fire input model only assumes 100% peroxide, and no NaOH is modeled, since the spray fire model only models reactions (1-2). Note that in order to model the experiment properly, the spray fire model needs to incorporate upward spraying and the terminal velocity of the droplet needs to be reflected in this change.



(a)



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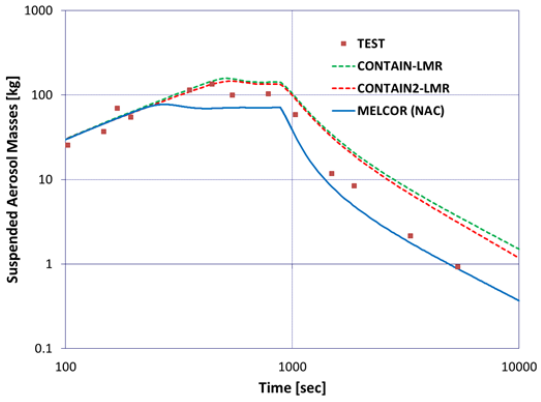

(c)

FIG. 4. Preliminary MELCOR Results

5. Summary and Conclusion

This paper documents the implementation status of the CONTAIN-LMR sodium chemistry models into MELCOR 2.1. The paper also describes the models from CONTAIN-LMR and the new package NAC for integrating these models in MELCOR. A preliminary validation test for the spray fire model using AB5 test is discussed. Additional validations need to be done for the spray fire model, particularly for the experiments that contain sprays pointing downward. Testing of the pool fire model implemented in MELCOR 2.1 is in progress and includes identifying experiments to validate the model. Incorporating the atmosphere chemistry model in the MELCOR model for both spray and pool fire model testing may be more appropriate since moisture may exist in the test apparatus. The moisture may enhance the formation of NaOH as reported in the AB5 test above as an example.

6. Acknowledgement

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