Development And Assessment Of An Extended MATPRO Materials Property Library For Accident Tolerant Fuel Materials

Chris Allison
Innovative Systems Software LLC
Idaho Falls, Idaho USA
issgml97@gmail.com

Marina Perez-Ferragut
Energy Software Ltd.
Barcelona, Spain
marina.pf@ensobcn.com

Sarah Khalil, Ayah Elshahat
Alexandria University
Alexandria, Egypt
sy.khalil@gmail.com, ayahabouelnaga@hotmail.com

Carlos Chavez
University of Mexico
Mexico City, Mexico
chm.carlos@gmail.com

Rawan Mustafa
Jordan Atomic Energy Commission
Aman, Jordan
rawan.mustafa@jaec.gov.jo

ABSTRACT

The MATPRO materials property library, developed originally by the U.S. Nuclear Regulatory Commission in the 1970s for use in their steady state and transient fuel behavior codes, FRAP-CON and FRAP-T, and extended in the 1980s for SCDAP/RELAP5, has been widely used to describe the behavior of typical LWR core and RCS structural materials for both design and beyond design basis accident conditions. In the mid-1990s, this library was extended by Innovative Systems Software (ISS) to include the Zr-Nb cladding materials used in many VVER fuel assemblies, allowing the RELAP/SCDAPSIM and new integral code, ASYST, to support the analysis of VVER-specific fuel bundle heating and melting experiments performed in the German CORA and QUENCH and Russian PARAMETER facilities for beyond design basis conditions. In 2021, it was proposed that this library be further extended to include a range of Accident Tolerant Fuel (ATF) materials to support an IAEA collaborative research project designed to extend the assessment and application of such materials. This activity is now well underway with support from ISS's collaborative partners from Mexico, Egypt, Spain, and Jordan as well as other collaborators in the IAEA project. This paper describes activities involved in this project, the development of material property models and reference correlations for one of the materials, FeCrAl, the implementation of these models and correlations into the MATSIM materials property library, and the status of the assessment and application of these new models and correlations.

***Keywords**: RELAP/SCDAPSIM, ASYST, PWR SBO analysis, Accident Tolerant Cladding, FeCrAl*

1 INTRODUCTION

The MATPRO materials property library, developed originally by the U.S. Nuclear Regulatory Commission in the 1970s for use in their steady state and transient fuel behavior codes, FRAP-CON and FRAP-T, and extended in the 1980s for SCDAP/RELAP5, has been widely used to describe the behavior of typical LWR core and RCS structural materials for both design and beyond design basis accident conditions (DBA and BDBA) [1]. In the mid-1990s, this library was extended by Innovative Systems Software (ISS) to include the Zr-Nb cladding materials used in many VVER fuel assemblies. This library of materials property correlations is now designated as MATSIM to distinguish it from the original SCDAP/RELAP5 MATPRO library. In 2021, it was proposed that this library be further extended to include a range of Accident Tolerant Fuel (ATF) materials to support an IAEA collaborative research project (CRP) designed to extend the assessment and application of such materials. This activity is now well underway with support from ISS's collaborative partners from Mexico, Egypt, Spain, and Jordan as well as other collaborators in the IAEA project.

The ISS, and collaborative, activities are described in Section 2 of the development and assessment activities. Section 3 provide a brief description of the approach used to develop and document the wide range of material property correlations needed for such a library. Examples of the review and assessment of proposed correlations currently available in the open literature, spreadsheet templates, and resulting correlations for FeCrAl are included in this section. Section 4 describes the approach used to implement the new material property correlation data base into MATSIM. Section 5 describes the status of the assessment and application of these new models and correlations.

2 OVERVIEW - DEVELOPMENT AND ASSESSMENT OF ATF MATERIALS PROPERTY LIBRARY

The IAEA ATF CRP is helping to extend the ATF data base and models for DBA and BDBA conditions. The project includes both separate effects (SE) and integral experiments (IE) that cover DBA ($T < 1500$ K) and the early stage of BDBA ($1500 \text{ K} < T < \sim 2000$ K). The SEs are focused primarily on cladding ballooning/rupture (LOCA-DBA) and DBA-BDBA oxidation and other material interactions for coated Zr cladding, FeCrAl, and SiC. The IE-related activities look at combined DBA/BDBA conditions related to the uncovery, heat up, and oxidation of representative fuel assemblies with FeCrAl or coated Zr cladding (and grid spacers). These activities include (a) the continued analysis of the Karlsruhe Institute of Technology (KIT) Quench 19 and associated material property SEs, and (b) the performance of new experiments in Japan and Hungary. The CRP also includes three modelling-related activities (a) fuel performance code improvements and uncertainty analysis, (b) system thermal hydraulic and Severe Accident (SA) code assessment and applications, and (c) material property data base model/correlation development, review, and assessment. The fuel performance code task is focused on small and large break evaluation and benchmarking for licensing and other DBA-related purposes. The system thermal hydraulic/severe accident code task is focused on the applications of the codes to representative plant calculations under DBA/BDBA conditions. ISS, and collaborators, are leading the activities related to task (c) as described in this paper [2].

The early FeCrAl experiments like Quench-19 and associated SEs have clearly demonstrated that FeCrAl cladding can significantly reduce the oxidation and associated hydrogen production under DBA conditions relative to Zr-based alloys. However, these experiments have also shown that the FeCrAl cladding behavior during BDBA conditions is much less clear. As shown in a recent paper published at the 2021 Top Fuel meeting [3], FeCrAl cladding starts losing its structural integrity when oxidized in steam under isothermal conditions at temperatures around 1620 K. The rate of oxygen uptake also increases dramatically around that temperature. Figure 1, which shows the oxidation parabolic rate coefficient computed from published data and correlations for two typical FeCrAl alloys, shows an increase of several orders of magnitude around the same temperature, significantly above that of Zircaloy in the same temperature range. The QUENCH 19 experiment, performed in 2018 with FeCrAl cladding, shroud, spacer grids, and associated SEs [4-6] had results consistent with the information shown in Figure 1 although this was a transient experiment terminated by quenching. The reported peak temperature for this experiment was ~1730 K. The measured hydrogen generation rates and integral hydrogen production for Quench 19 were somewhat inconsistent with what might have been expected based on a comparison with a similar experiment, QUENCH-15, that used ZIRLO. For example, as might be expected for FeCrAl, the integral hydrogen production in QUENCH-19 was almost a factor of 5 less than that of QUENCH-15. But there were sharp jumps in the hydrogen generation rate near the end of the QUENCH-19 experiment that were not well predicted using the recommended correlations for the FeCrAl alloys that were used (B136Y and KANTHAL APM). However, as might have been expected, the maximum hydrogen production rate measured (prior to quench) in QUENCH-19 was about 14% of the maximum rate in QUENCH -15. (The maximum cladding temperature in QUENCH-19 was also ~460 K lower than that of QUENCH-15, which could have contributed to the difference in hydrogen generation.) In reference to apparent problems in predicting the hydrogen production, it was noted in the reference [6] that hydrogen production in QUENCH-19 was apparently impacted by two different factors. First, a sharp increase in hydrogen production at temperatures above ~1650 K, was likely due to the disappearance of the protective Al_2O_3 coating on the surface of the material. Second, another sharp increase, approximately 800 s prior to reflood, may have been caused by the melting or reaction between the FeCrAl cladding and the steel cladding of the thermocouples in the upper region of the bundle.

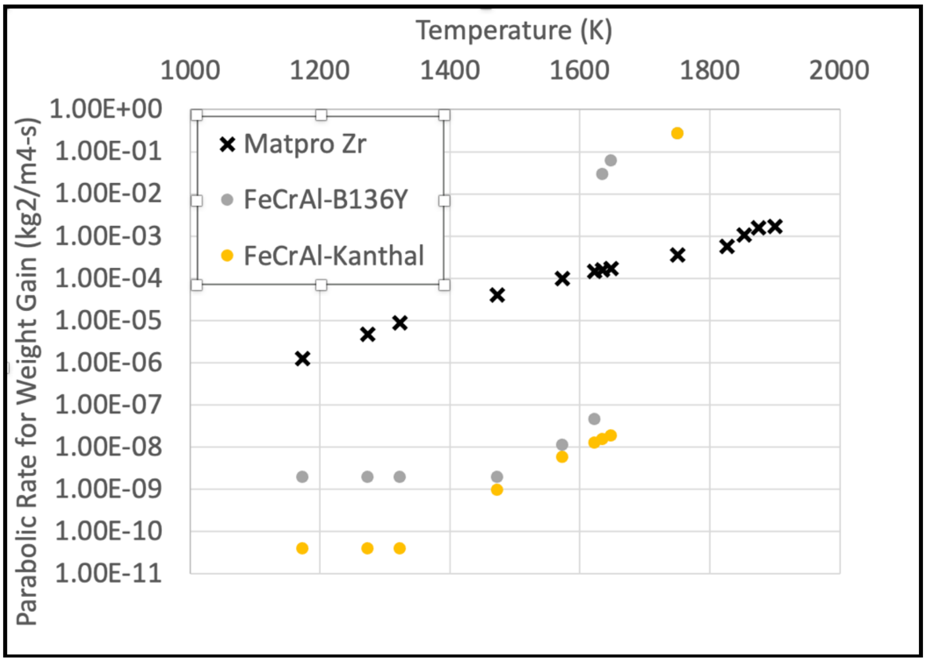


Figure 1: Oxygen weight parabolic rate coefficient for Zr, B1367, and KANTHAL APM

The proposal to include a task in the IAEA CRP to create a open, peer reviewed ATF material property data base was based on two factors. First, although the MATPRO library and reference handbook was originally developed by the US NRC for their DBA fuel behavior codes and then expanded for SCDAP/RELAP5 for severe accidents, it has become widely used and referenced in the international nuclear community. It's value, in large measure, is that it allowed international researchers, model developers, and analysts to share a fundamental, common set of peer reviewed models and correlations that could then be independently assessed and then used in a wide range of applications for more than 4 decades. Thus, it seemed that the collaborative development and assessment of an ATF specific material property data base that could be used and improved by the CRP participants, and then made available to the international nuclear R&D community upon completion of the CRP would be appropriate under the IAEA's auspices. Second, unlike MATPRO which was developed for a relative limited number of materials with a very extensive experimental data base for both DBA and severe accidents, there are many possible ATF materials that might ultimately be selected for use, but with a very limited, and recent experimental data base, particularly for BDBA conditions. Thus, working within the framework of the IAEA CRP that has an international community supporting the activities with experiments, model development, and assessment seemed to be the only possible way to create a material property data base that could have the same international impact as MATPRO.

Like the development of MATPRO, the development and assessment of a ATF material property data base is a very time and labor intensive process. There are five collaborative activities being led by ISS and our collaborators.

1. Develop a reference library of “peer reviewed” open publications on ATF material property models and experiments.
2. Develop and document an open source, peer reviewed set of ATF material property models and correlations.
3. Implement ATF correlations (and associated models) into the MATSIM library for assessment by CRP and ISS collaborators.
4. Support the design and assessment of CRP integral experiments including the development of RELAP/SCDAPSIM [7] and ASYST [8-10] input models.
5. Support the assessment of the ATF models and correlations using reference RELAP/SCDAPSIM and ASYST reference plant models.

3 APPROACH USED TO DEVELOP AND DOCUMENT MATERIAL PROPERTY CORRELATIONS

Although the information is still limited relative to UO_2 and Zr-based alloys, more than 100 publicly available documents/papers currently been added to the reference library for ATF materials that are being reviewed and characterized by materials experts that are collaborating on this task. Many of these documents or references to these documents have been provided by participants in the CRP as well as the ISS collaborators. One of the authors for this paper, S. Khahil from Alexandria University has the technical lead on coordinating the review of the documents, extracting any proposed correlations or relevant data for specific materials and properties, and comparing the data/correlations, and where appropriate, developing a combined correlation and uncertainty assessment for those specific properties. She is supported by her graduate students and another author, R. Mustafa from the Jordan Atomic Energy Commission. Other participants in the CRP and those interested in supporting the development and assessment can also participate in the review and documentation process.

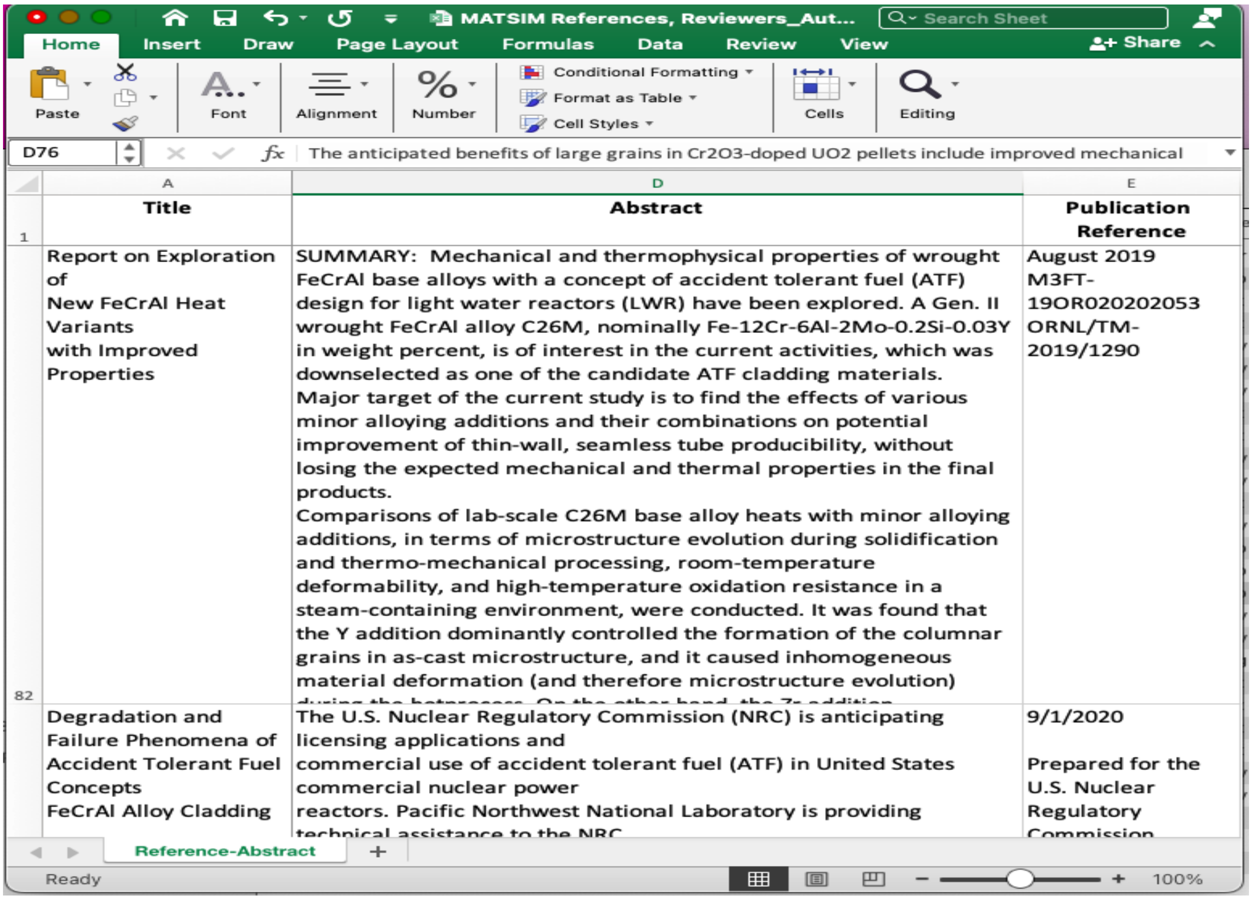
Spreadsheet templates have established to document the review of the literature and documentation of the correlations. Figure 2 shows an example of the spreadsheet template that has been used to document the review of the literature. Figure 3 shows and example of the spreadsheet template used to document the references and correlations used for each material property. Although the publication of periodic review papers in the open literature is encouraged, such publications are up to the team leading the review as well as the individual reviewers.

Participants in the IAEA CRP and other groups working on the development and assessment of ATF materials, implementing ATF material property models and correlations into their models or codes, or would like to evaluate the influence of the new materials have been encouraged to participate in the review and assessment process as outlined in Figure 4. *Only experimental data or correlations previously published in publicly available, peer review papers or documents will be* *included in the data base and reference material.* Once the data base and associated documentation has been provided to the IAEA at the completion of the CRP, the IAEA has indicated that they will continue to maintain and support the data base. The procedures providing updated information and access to the data base are expected to be included in relevant IAEA technical documents and publicly available information. ISS, and our collaborators, will continue to maintain the updated MATSIM library options for use by licensed software users.

4 IMPLEMENTATION IN MATSIM

Two different approaches were taken in implementing the different FeCrAl material properties. One approach, as represented in Figure 5, assigns a unique numeric identifier for each different material so the property and material are then be identified by a common name with numeric identifier. In the example shown in Figure 5, the density of one FeCrAl alloy is coded as a function with an identifier of "cden28". The name "cden" is used in the MATPRO library for cladding density with the density of Zircaloy used. Thus, the user of this function can either simply replace "cden28" in the function with "cden" and replace the existing Zircaloy density with FeCrAl. Another more general approach is to expand the existing MATPRO routine to include a material identifier so the appropriate correlation is used for the specified material. This approach, although more general, requires changes in the calling routines and input to incorporate a unique material identifier. An example of this approach is shown in Figure 6, where the oxygen weight gain parabolic rate coefficients for Zircaloy, Zr-Nb (VVER), and two FeCrAl alloys are included in the material property routine.

The implementation of the new MATSIM ATF models and correlations into ASYST VER 3.5 is well underway. When verification testing is completed, ASYST VER 3.5 will be released for independent testing and assessment.



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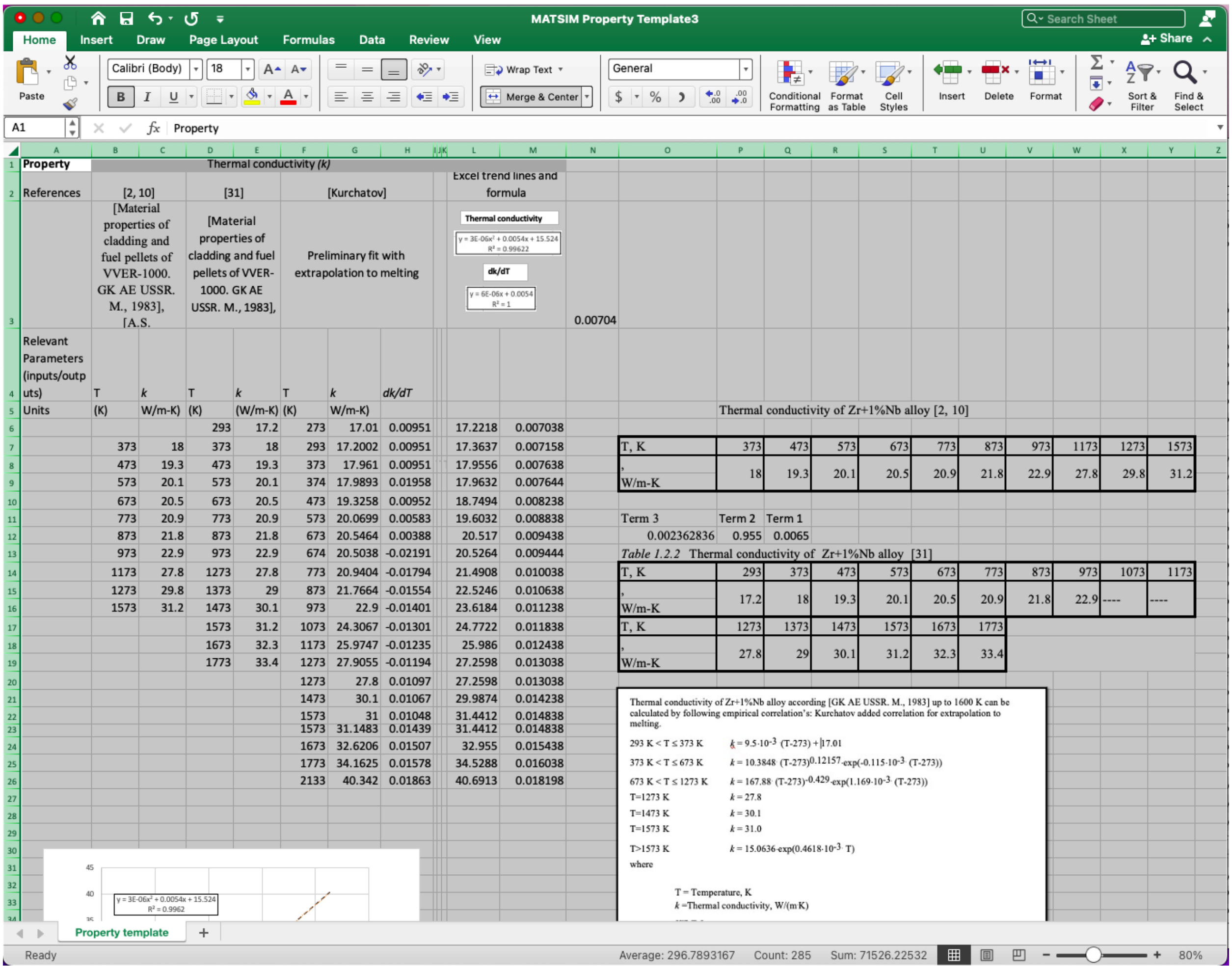
The anticipated benefits of large grains in Cr2O3-doped UO2 pellets include improved mechanical

	A	D	E
1	**Title**	**Abstract**	**Publication Reference**
82	Report on Exploration of New FeCrAl Heat Variants with Improved Properties	SUMMARY: Mechanical and thermophysical properties of wrought FeCrAl base alloys with a concept of accident tolerant fuel (ATF) design for light water reactors (LWR) have been explored. A Gen. II wrought FeCrAl alloy C26M, nominally Fe-12Cr-6Al-2Mo-0.2Si-0.03Y in weight percent, is of interest in the current activities, which was downselected as one of the candidate ATF cladding materials. Major target of the current study is to find the effects of various minor alloying additions and their combinations on potential improvement of thin-wall, seamless tube producibility, without losing the expected mechanical and thermal properties in the final products. Comparisons of lab-scale C26M base alloy heats with minor alloying additions, in terms of microstructure evolution during solidification and thermo-mechanical processing, room-temperature deformability, and high-temperature oxidation resistance in a steam-containing environment, were conducted. It was found that the Y addition dominantly controlled the formation of the columnar grains in as-cast microstructure, and it caused inhomogeneous material deformation (and therefore microstructure evolution)	August 2019 M3FT-19OR020202053 ORNL/TM-2019/1290
	Degradation and Failure Phenomena of Accident Tolerant Fuel Concepts FeCrAl Alloy Cladding	The U.S. Nuclear Regulatory Commission (NRC) is anticipating licensing applications and commercial use of accident tolerant fuel (ATF) in United States commercial nuclear power reactors. Pacific Northwest National Laboratory is providing technical assistance to the NRC	9/1/2020 Prepared for the U.S. Nuclear Regulatory Commission

Figure 2: Spreadsheet Template for the Review of the Literature



	A	B	C	D	E	F	G	H	L	M	N
1	**Property**	Thermal conductivity *(k)*									
2	References	[2, 10]		[31]		[Kurchatov]			Excel trend lines and formula		
3		[Material properties of cladding and fuel pellets of VVER-1000. GK AE USSR. M., 1983], [A.S.		[Material properties of cladding and fuel pellets of VVER-1000. GK AE USSR. M., 1983],		Preliminary fit with extrapolation to melting			Thermal conductivity y = 3E-06x^2 + 0.0054x + 15.524 R^2 = 0.99622 dk/dT y = 6E-06x + 0.0054 R^2 = 1		0.00704
4	Relevant Parameters (inputs/outputs)	T	*k*	T	*k*	T	*k*	*dk/dT*			
5	Units	(K)	W/m-K)	(K)	(W/m-K)	(K)	W/m-K)				
6				293	17.2	273	17.01	0.00951	17.2218	0.007038	
7		373	18	373	18	293	17.2002	0.00951	17.3637	0.007158	
8		473	19.3	473	19.3	373	17.961	0.00951	17.9556	0.007638	
9		573	20.1	573	20.1	374	17.9893	0.01958	17.9632	0.007644	
10		673	20.5	673	20.5	473	19.3258	0.00952	18.7494	0.008238	
11		773	20.9	773	20.9	573	20.0699	0.00583	19.6032	0.008838	
12		873	21.8	873	21.8	673	20.5464	0.00388	20.517	0.009438	
13		973	22.9	973	22.9	674	20.5038	-0.02191	20.5264	0.009444	
14		1173	27.8	1273	27.8	773	20.9404	-0.01794	21.4908	0.010038	
15		1273	29.8	1373	29	873	21.7664	-0.01554	22.5246	0.010638	
16		1573	31.2	1473	30.1	973	22.9	-0.01401	23.6184	0.011238	
17				1573	31.2	1073	24.3067	-0.01301	24.7722	0.011838	
18				1673	32.3	1173	25.9747	-0.01235	25.986	0.012438	
19				1773	33.4	1273	27.9055	-0.01194	27.2598	0.013038	
20						1273	27.8	0.01097	27.2598	0.013038	
21						1473	30.1	0.01067	29.9874	0.014238	
22						1573	31	0.01048	31.4412	0.014838	
23						1573	31.1483	0.01439	31.4412	0.014838	
24						1673	32.6206	0.01507	32.955	0.015438	
25						1773	34.1625	0.01578	34.5288	0.016038	
26						2133	40.342	0.01863	40.6913	0.018198	

Thermal conductivity of Zr+1%Nb alloy [2, 10]

T, K	373	473	573	673	773	873	973	1173	1273	1573
, W/m-K	18	19.3	20.1	20.5	20.9	21.8	22.9	27.8	29.8	31.2

Term 3	Term 2	Term 1
0.002362836	0.955	0.0065

Table 1.2.2 Thermal conductivity of Zr+1%Nb alloy [31]

T, K	293	373	473	573	673	773	873	973	1073	1173
, W/m-K	17.2	18	19.3	20.1	20.5	20.9	21.8	22.9	----	----
T, K	1273	1373	1473	1573	1673	1773				
, W/m-K	27.8	29	30.1	31.2	32.3	33.4				

Thermal conductivity of Zr+1%Nb alloy according [GK AE USSR. M., 1983] up to 1600 K can be calculated by following empirical correlation's: Kurchatov added correlation for extrapolation to melting.

293 K < T ≤ 373 K $k = 9.5\cdot10^{-3}\cdot(T-273) + 17.01$

373 K < T ≤ 673 K $k = 10.3848\cdot(T-273)^{0.12157}\cdot\exp(-0.115\cdot10^{-3}\cdot(T-273))$

673 K < T ≤ 1273 K $k = 167.88\cdot(T-273)^{-0.429}\cdot\exp(1.169\cdot10^{-3}\cdot(T-273))$

T=1273 K $k = 27.8$

T=1473 K $k = 30.1$

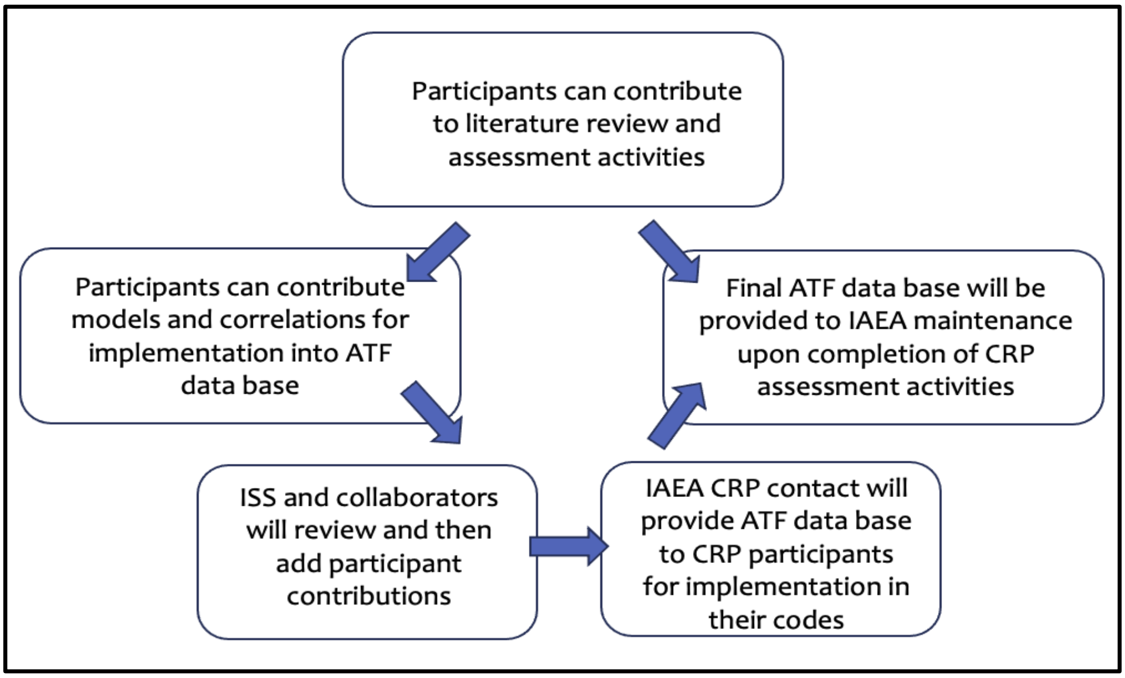
T=1573 K $k = 31.0$

T>1573 K $k = 15.0636\cdot\exp(0.4618\cdot10^{-3}\cdot T)$

where

T = Temperature, K
k =Thermal conductivity, W/(m·K)

Figure 3: Spreadsheet Template for the Review and Analysis of Data and Correlations for a Specific Material Property



Participants can contribute to literature review and assessment activities

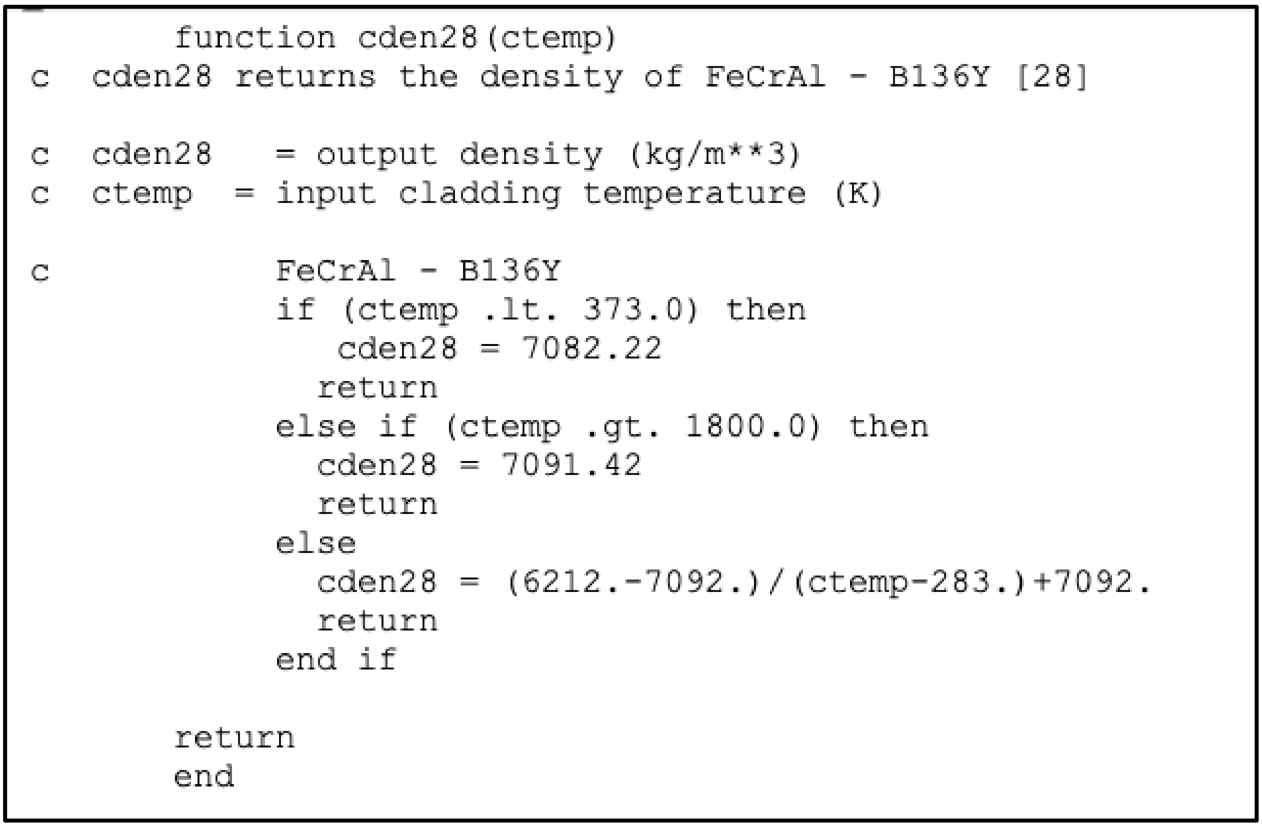
Participants can contribute models and correlations for implementation into ATF data base

Final ATF data base will be provided to IAEA maintenance upon completion of CRP assessment activities

ISS and collaborators will review and then add participant contributions

IAEA CRP contact will provide ATF data base to CRP participants for implementation in their codes

Figure 4: Current Process to Contribute to the Development of an Open ATF-specific Models and Correlations Data Base

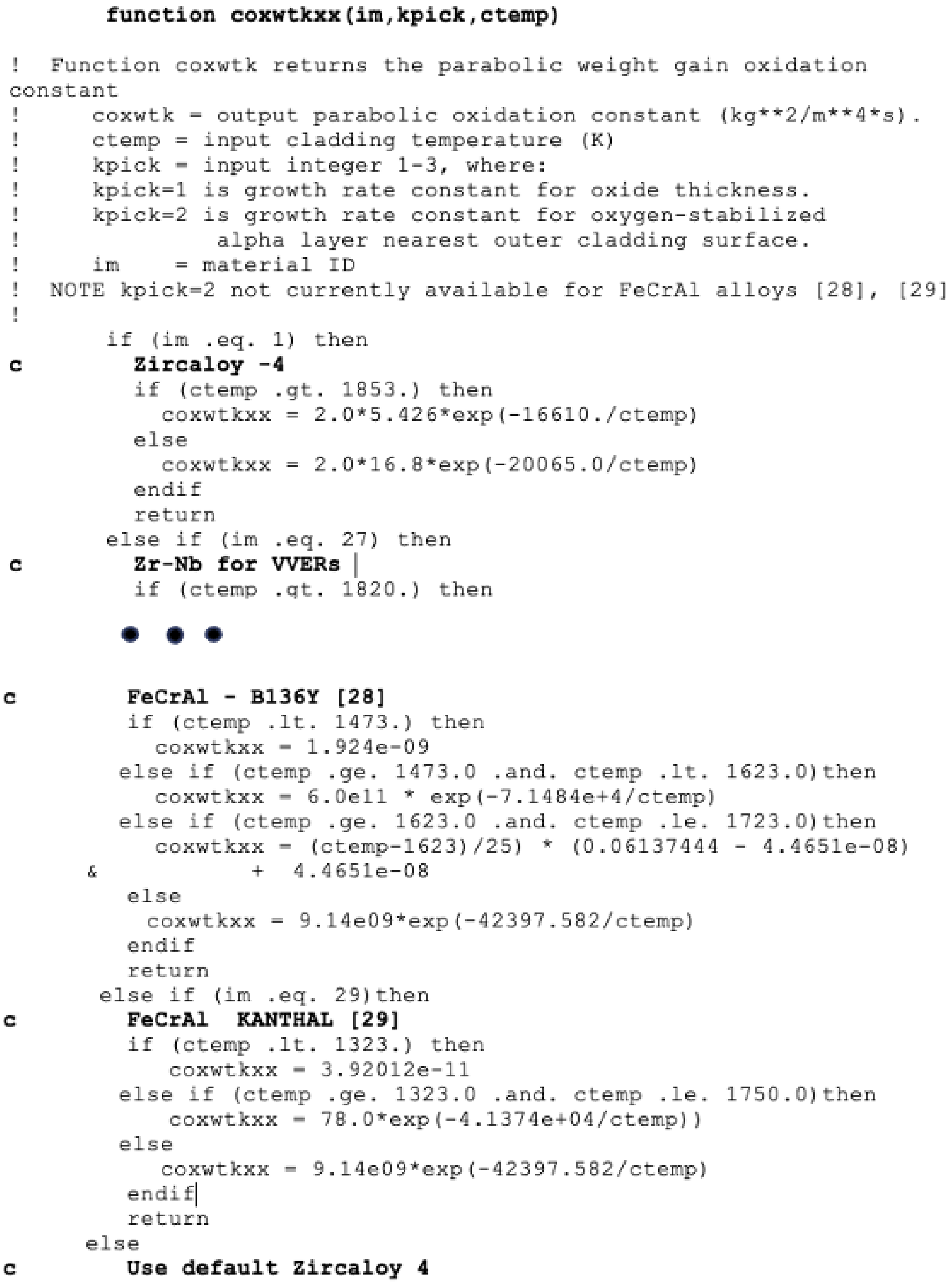


```
       function cden28(ctemp)
c  cden28 returns the density of FeCrAl - B136Y [28]

c  cden28   = output density (kg/m**3)
c  ctemp  = input cladding temperature (K)

c           FeCrAl - B136Y
            if (ctemp .lt. 373.0) then
               cden28 = 7082.22
              return
            else if (ctemp .gt. 1800.0) then
              cden28 = 7091.42
              return
            else
              cden28 = (6212.-7092.)/(ctemp-283.)+7092.
              return
            end if

       return
       end
```

Figure 5: Implementation Using a Unique Material Property Names for Each Property and Material

5 STATUS OF THE ASSESSMENT AND APPLICATION OF ATF-SPECIFIC MODELS AND CORRELATIONS

The independent testing and assessment of the new ATF-specific models and correlation will continue for an extended period of time as IAEA CRP activities are completed over the next year or so. Additional materials will be added to the ATF data base as proposed new SE and IE experiments are designed and performed. Under the current time line, it is anticipated that new models and correlations for coated Zr cladding will be added as a first priority to support the proposed integral experiments to be performed in Japan. The independent testing of the FeCrAl models and correlations, as implemented in ASYST VER 3.5, will begin in the near future with the publication of initial results in the fall of 2022 or spring of 2023. ISS, and their collaborators, will initially start with the analysis of the integral QUENCH experiments that have been completed by KIT. The expected time sequence for this work is to start with the re-analysis of the QUENCH-06 experiment using comparative studies using Zircaloy as used in the original experiment and then replacing the Zircaloy with FeCrAl to repeat the analysis. QUENCH-06 was performed in the 1990s as an International Standard Problem so the input models and uncertainty assessments for the experiment have been well established [11]. Some of the CRP and ISS collaborators are also planned to develop input models for the QUENCH-19 experiment and assess the influence of some of the variations in the thermal hydraulic boundary and expected results as was noted in section 3. Other ISS collaborators are planning to use some of the detailed RELAP/SCDAPSIM input models that have been developed for training, for use in the ongoing Fukushima Daiichi decommissioning R&D activities [12], and for the more recent VVER-1000 studies described in a paper also being presented at this meeting [13]. These detailed input models include PWR, BWR, and VVER designs and have been used for both DBA and BDBA transient analysis.



```
        function coxwtkxx(im,kpick,ctemp)

!  Function coxwtk returns the parabolic weight gain oxidation
constant
!     coxwtk = output parabolic oxidation constant (kg**2/m**4*s).
!     ctemp = input cladding temperature (K)
!     kpick = input integer 1-3, where:
!     kpick=1 is growth rate constant for oxide thickness.
!     kpick=2 is growth rate constant for oxygen-stabilized
!              alpha layer nearest outer cladding surface.
!     im    = material ID
!  NOTE kpick=2 not currently available for FeCrAl alloys [28], [29]
!
       if (im .eq. 1) then
c        Zircaloy -4
         if (ctemp .gt. 1853.) then
           coxwtkxx = 2.0*5.426*exp(-16610./ctemp)
         else
           coxwtkxx = 2.0*16.8*exp(-20065.0/ctemp)
         endif
         return
       else if (im .eq. 27) then
c        Zr-Nb for VVERs
         if (ctemp .gt. 1820.) then

         ● ● ●

c        FeCrAl - B136Y [28]
         if (ctemp .lt. 1473.) then
           coxwtkxx = 1.924e-09
        else if (ctemp .ge. 1473.0 .and. ctemp .lt. 1623.0)then
           coxwtkxx = 6.0e11 * exp(-7.1484e+4/ctemp)
        else if (ctemp .ge. 1623.0 .and. ctemp .le. 1723.0)then
           coxwtkxx = (ctemp-1623)/25) * (0.06137444 - 4.4651e-08)
     &           +  4.4651e-08
         else
          coxwtkxx = 9.14e09*exp(-42397.582/ctemp)
         endif
         return
       else if (im .eq. 29)then
c        FeCrAl  KANTHAL [29]
         if (ctemp .lt. 1323.) then
            coxwtkxx = 3.92012e-11
        else if (ctemp .ge. 1323.0 .and. ctemp .le. 1750.0)then
            coxwtkxx = 78.0*exp(-4.1374e+04/ctemp))
        else
           coxwtkxx = 9.14e09*exp(-42397.582/ctemp)
         endif
         return
      else
c        Use default Zircaloy 4
```

Figure 6: Implementation Using a Unique Material Identifier with Routines Including All Materials for a Specific Property

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