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High temperature thermal properties for metals used in LWR vessels

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Abstract

Because of the impact that melt relocation and vessel failure has on subsequent progression and associated consequences of a light water reactor (LWR) accident, it is important to accurately predict the heatup and relocation of materials within the reactor vessel and heat transfer to and from the reactor vessel. Accurate predictions of such heat transfer phenomena require high temperature thermal properties. However, a review of vessel and structural steel material properties in severe accident analysis codes reveals that the required high temperature material properties are extrapolated with little, if any, data above 700 °C. To reduce uncertainties in predictions relying upon this extrapolated high temperature data, INL obtained data using laser-flash thermal diffusivity techniques for two metals used in LWR vessels: SA 533 Grade B, Class 1 (SA533B1) low alloy steel, which is used to fabricate most US LWR reactor vessels; and Type 304 Stainless Steel SS304, which is used in LWR vessel piping, penetration tubes, and internal structures. This paper summarizes the new data, compares it to existing data in the literature, and provides recommended correlations for thermal properties based on this data.

1. Introduction

Melt relocation and vessel failure impacts the subsequent progression and associated consequences of a light water reactor (LWR) accident. Hence, it is important to accurately predict the heatup and relocation of materials within the reactor vessel and heat transfer to and from the reactor vessel. However, a review of vessel and structural steel material properties used to predict such phenomena in severe accident analysis codes, such as SCDAP/RELAP5-3D© [1], MELCOR [2], and MAAP [3], reveals that the required high temperature material properties are extrapolated with little, if any, data above 700 °C. To reduce uncertainties in predictions relying upon this extrapolated high temperature data, INL obtained data using laser-flash thermal diffusivity techniques for two metals used in LWR vessels: SA533 Grade B Class 1 (SA533B1) low alloy steel, which is used to fabricate most US LWR reactor vessels; and Type 304 Stainless Steel (SS304), which is used in LWR vessel piping, penetration tubes, and internal structures.

Prior to obtaining new high temperature data, existing thermal property data for these materials were reviewed, so that new data could be compared with data available in the literature. Results from this review are found in Section 2 of this paper. New high temperature data obtained in this effort and comparisons of the new data with available data in the literature are reported in Section 3. Section 4 summarizes insights from this study.

1.1. Method

The effort reported in this paper relied upon pulsed thermal diffusivity methods (TDMs) to obtain high temperature material thermal properties. TDMs are based on the material's thermal diffusivity, α; the thermophysical property that best determines the speed of heat propagation by conduction during changes of temperature with time. The thermal diffusivity, α, is defined by

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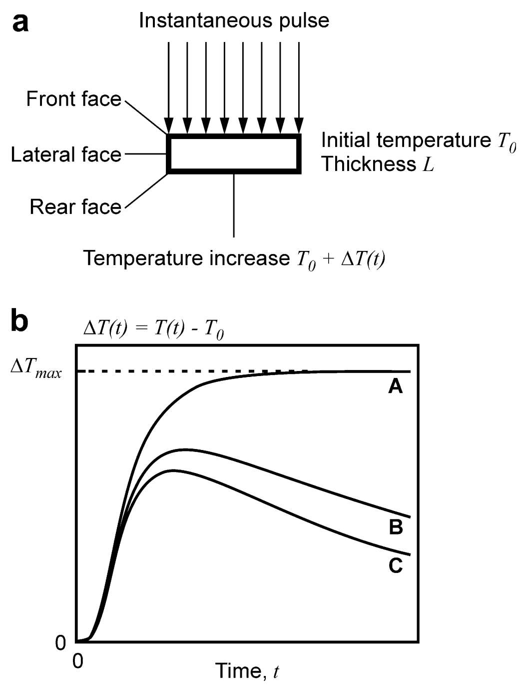

Fig. 1. Flash method (a) geometry and (b) temperature increases for various experimental conditions.

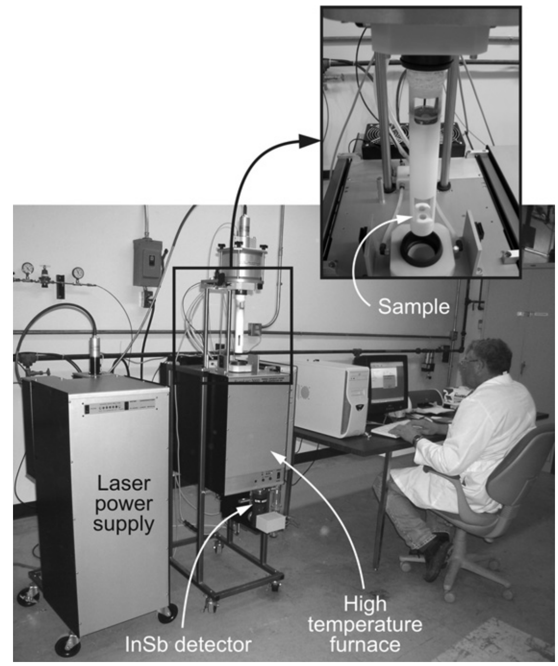

Fig. 2. Laser-flash thermal property analyzer (Anter FL5000) installed at HTTL.

$$\alpha = k/\rho c_p, \quad (1)$$

where k represents the thermal conductivity, c_p represents the specific heat capacity, and ρ represents the density. As indicated in this definition, the thermal diffusivity affects any conductive transient heat transfer process within the medium. The higher the thermal diffusivity, the faster the heat propagation.

Since its introduction by Parker et al. [4] the flash method has become popular among thermal diffusivity measuring methods because of its speed (commercial systems may go from room temperature to 1600 °C in less than a day), accuracy (commercial systems are reporting accuracies of ±3% or better), and reproducibility (commercial systems are reporting reproducibilities of ±2% or better). After the sample is stabilized at a desired temperature, T_0, a nearly instantaneous pulse of energy (usually from a laser or other discharge source) is deposited on its front face, and the temperature increase, $T(t)$, on the rear face of the sample is recorded as a function of time (see Fig. 1(a)). The thermal diffusivity is then determined from the resulting thermogram (see Fig. 1(b)). If no heat loss is involved, the rear face temperature will rise to a maximum and remain at that level indefinitely (curve A). However, with increasing heat losses, the temperature on the rear face decreases after reaching a maximum value (curves B and C).

The original method proposed by Parker assumes an isotropic and adiabatic sample (e.g., no heat loss). Several analytical methods, such as that proposed by Clark and Taylor [5], have been introduced over the years to enhance the Parker method by considering heat losses, finite pulse duration, and non-uniform pulse heating. In a study performed by Thermitus,[1] it was concluded that the accuracy of these various methods is similar, provided that certain test criteria are met, such as the sampling frequency, the signal to noise ratio, etc.

The thermal conductivity may be calculated from thermal diffusivity measurements using Eq. (1). However, the density and specific heat capacity must be determined by other methods before the thermal conductivity can be calculated. Commercial software, which can determine the heat capacity by a comparative method with a reference sample that has similar properties, is often provided with TDM systems.

1.2. Approach

In this project, TDM data were obtained using an Anter thermal properties analyzer (FL 5000) system (see Fig. 2) installed at INL's high temperature test laboratory (HTTL). This system uniformly heats a small disk-shaped sample (typically, 12 mm in diameter and 2–4 mm thick) over its front face with a very short pulse of energy from a laser in a temperature-controlled furnace. The time-temperature history of the rear face of the sample is recorded through high-speed data acquisition from a solid-state optical sensor with very fast thermal response. Thermal diffusivity is determined from the time interval after the flash for the rear face to increase in temperature (using the Clark and Taylor method). Specific heat capacity and thermal

[1] Thermitus, Marc–Antoine, *Thermal Diffusivity and the Flash Method*, personal communication.

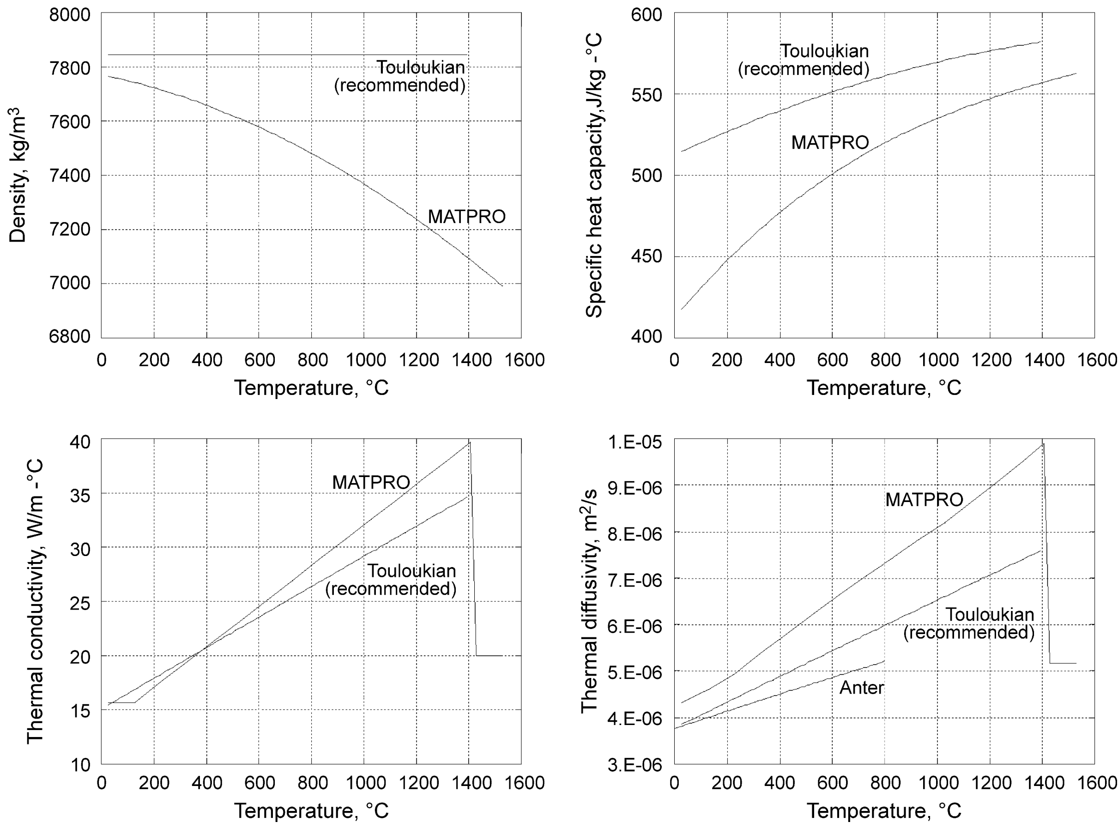

Fig. 3. Comparison of selected reference information for stainless steel material properties.

conductivity data were estimated using comparative techniques with software provided by Anter.

2. Existing data

As part of this effort, test data are compared to available data in the literature. Additionally, reference sample material properties are required to estimate test sample thermal conductivity and specific heat capacity using comparative techniques associated with pulse diffusivity methods. In this section, selected literature data are compared; and the 'preferred' or 'recommended' values for the density, specific heat capacity, thermal conductivity, and thermal diffusivity are presented for test and reference materials considered in this study. As documented in this section, all data for these metals are extrapolated for temperatures above 1000 °C (and in some references, are actually extrapolated at temperatures above 700 °C).

2.1. Stainless steel

Fig. 3 compares curves based on stainless steel material property data (diffusivity, thermal conductivity, specific heat capacity and density) found in Touloukian [6], the MATPRO library used in the SCDAP/RELAP5-3D© [1] and MELCOR [2] state-of-the-art severe accident analysis codes, and the thermal diffusivity information provided by Anter for the reference stainless steel sample [7]. Note that Touloukian and MATPRO curves are extrapolated for temperatures above 1000 °C. It is also interesting to note that large differences exist in values from these references. Thermal diffusivity values provided by Touloukian and Anter are as much as 60% lower than values recommended in the MATPRO material data library. MATPRO stainless steel 304 data are based on information in Peckner and Berstein [8]. Information from the analyst who developed this MATPRO correlation indicates that the data were very limited and the highest temperature data point was below 730 °C.[2] These discussions and new thermal diffusivity data obtained with the FL5000 led to the recommendation that information provided by Touloukian be adopted for this study.

2.2. Iron

The reference iron sample provided by Anter is often used as a comparison material because its thermal diffusivity is similar to carbon steel. Fig. 4 compares curves based on iron material property data (diffusivity, thermal conductivity, density, and diffusivity) found in Touloukian [7] and the reference sample thermal diffusivity information provided by Anter [8]. Because information for all the material properties of interest is available in Touloukian (and because the Touloukian data are consistent with informa-

[2] D.L. Hagrman, Consultant, email to J. Rempe, INL, July 6, 2005.

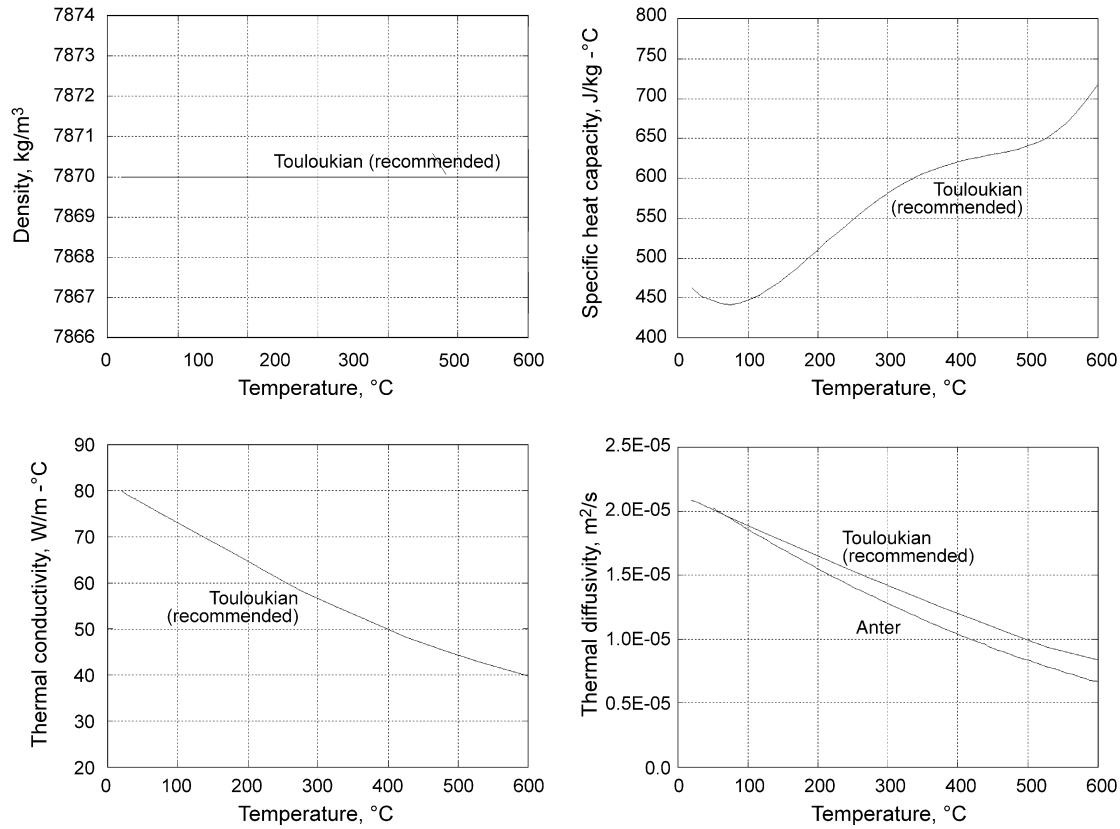

Fig. 4. Comparison of selected reference information for iron material properties.

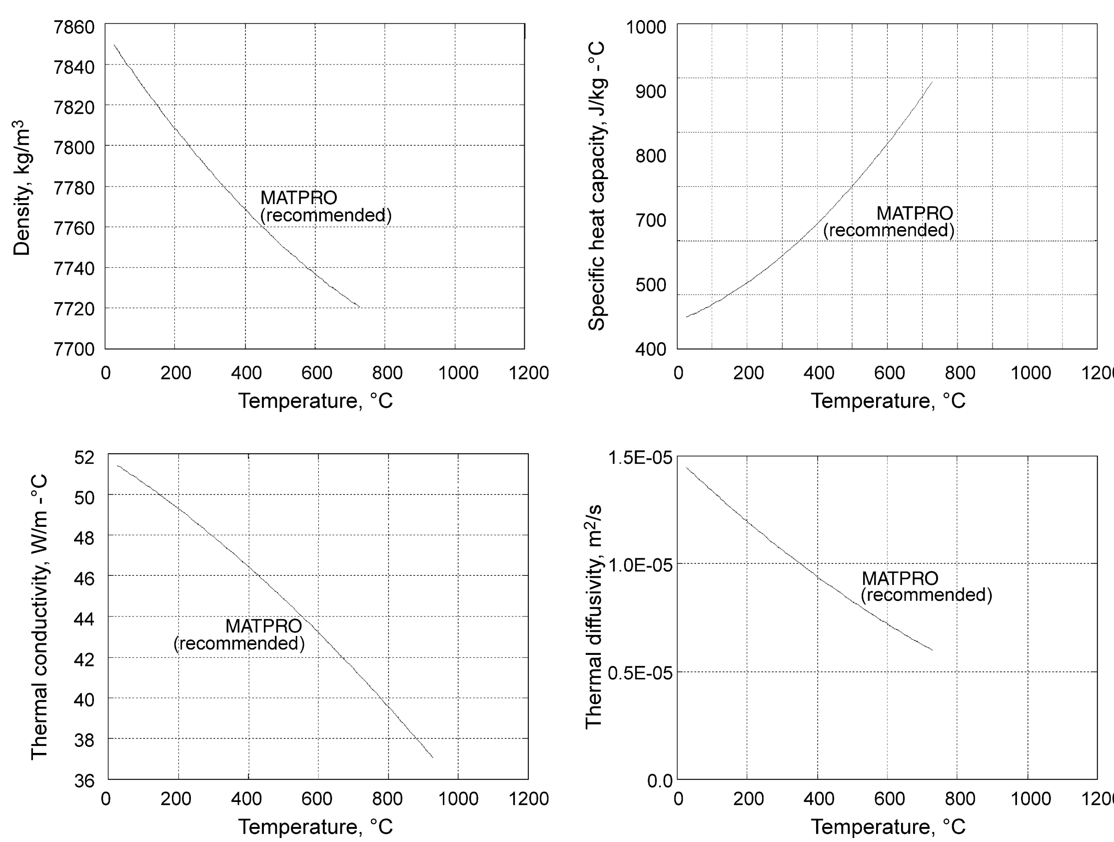

Fig. 5. Recommended SA533B1 material properties.

tion provided by Anter data), the Touloukian data are recommended for use in this effort.

2.3. SA533 B1

Fig. 5 contains curves based on material property data found in MATPRO for SA533B1 vessel steel. MATPRO SA533B1 data are based on information from Spanner et al. [9]. Because no additional data for this material were found, the MATPRO data are recommended.

3. Results

As noted in Section 1, TDM data were obtained in this project using an Anter FL5000 system installed at INL's HTTL. Although this system is capable of obtaining data up to 1600 °C, tests were limited to 1400 °C or lower in order to preclude sample melting. Test samples were approximately 12 mm in diameter and ranged from 2 to 4 mm thick. Test samples were designated by a test number (e.g., 400–430), their approximate thickness (e.g., 2–4), a numeric letter (e.g., A–C to denote the first through third sample of a particular thickness), their coating (e.g, 'C' for graphite sprayed, 'BN' for boron nitride sprayed, or 'GB' for grit blasted), and the voltage at which samples were tested (e.g., 1200–1500 V).

Thermal diffusivity data were calculated using the Clark and Taylor method [5], which is encoded into software provided by Anter for the FL5000 system. Specific heat capacity and thermal conductivity data were estimated using comparative techniques with software and reference samples provided by Anter. However, peak temperatures for which specific heat capacity and thermal conductivity data could be estimated were constrained to peak test temperatures for these reference samples (due to concerns about variations in data if samples were subjected to temperatures at or above where they experience a phase transition).

3.1. Stainless steel

Thermal diffusivity data for stainless steel samples (SS304) are plotted in Fig. 6. As indicated by the legend in this figure, SS304 data were obtained from testing 13 samples with thicknesses varying from 2 to 4 mm, laser powers varying from 1000 to 1500 V, and various types of sample coatings (graphite, boron nitride, and grit blasted). Data in this figure suggest that variations in test parameters did not produce any discernible trend in the test data. Fig. 7 compares this new SS304 data with data obtained from the Anter reference stainless steel sample, Anter reference sample thermal diffusivity data, and the recommended diffusivity (based on Touloukian data) from Fig. 3. As noted in Section 2, Fig. 3 recommended values for stainless steel are extrapolated above 1000 °C. As shown in Fig. 7, the SS304 data agree fairly well with reference sample data. Values for the SS304 and reference samples also agree fairly well with Fig. 3 recommended values at lower temperatures. More scatter occurs in data obtained at higher temperatures. However, high temperature data appear lower than the recommended Touloukian values at temperatures above 600 °C.

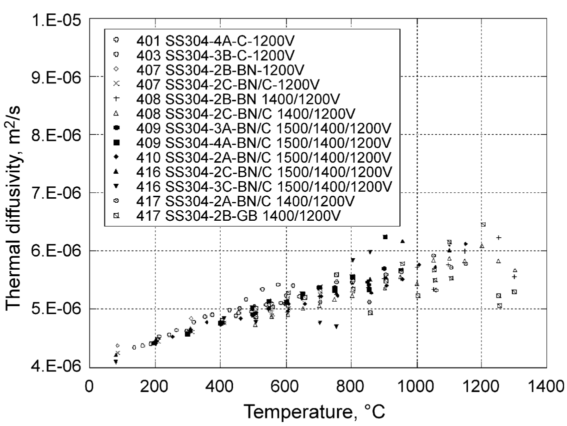

Fig. 6. SS304 thermal diffusivity data.

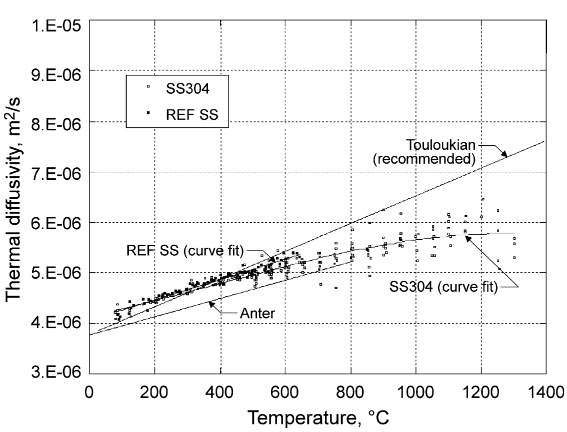

Fig. 7. Comparison of SS304 and reference stainless steel thermal diffusivity data with values provided in the literature (literature curves extrapolated above 1000 °C).

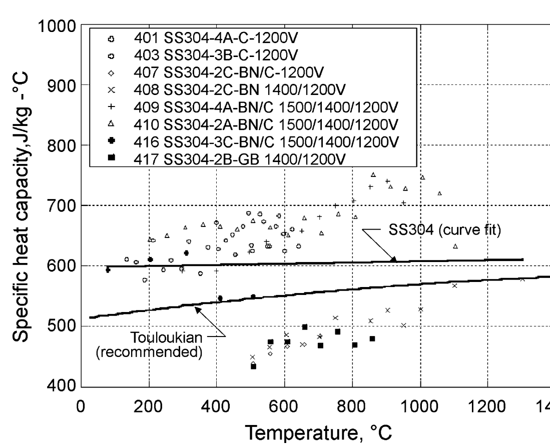

Fig. 8. Comparison of SS304 specific heat capacity data derived from FL5000 with data published in Touloukian.

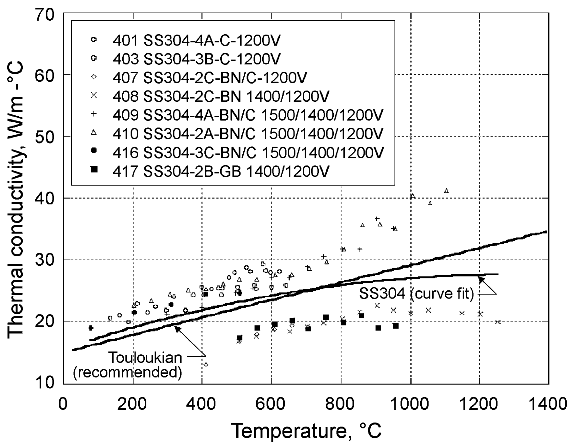

Fig. 9. Comparison of SS304 thermal conductivity data derived from FL5000 with data published in Touloukian.

Using software provided by Anter, the specific heat capacity and thermal conductivity of the SS304 samples were estimated using comparative techniques and data for a similar sample (in this case, the reference stainless steel sample provided by Anter). Figs. 8 and 9 illustrate results obtained using these techniques. Recommended curves (the curves from Fig. 3, which are based on Touloukian data) are also plotted in these figures.

Curve fits from data estimated with the Anter comparative software are similar to the recommended values from Fig. 3. However, there is considerable scatter in the data. More testing is recommended to reduce the uncertainty in the specific heat capacity and the thermal conductivity of this material (especially at higher temperatures).

3.2. SA533 B1 data

Thermal diffusivity data for SA533B1 samples tested at INL are plotted in Fig. 10. The SA533B1 data were obtained from testing nine samples with thicknesses varying from 2 to 4 mm, laser powers varying from 1000 to 1500 V, and various types of coatings (graphite, boron nitride, and grit blasted). Data suggest that variations in test parameters did not significantly affect test data. As in

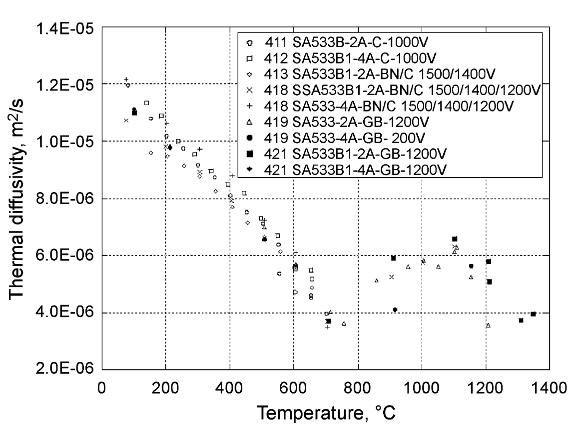

Fig. 10. INL SA533B1 thermal diffusivity data.

SS304 tests, there is more scatter in higher temperatures SA533B1 data. However, a change in the behavior of the SA533B1 diffusivity occurs at temperature above 730 °C, which is the temperature where this material starts to experience a transformation (from ferritic to austenitic steel)

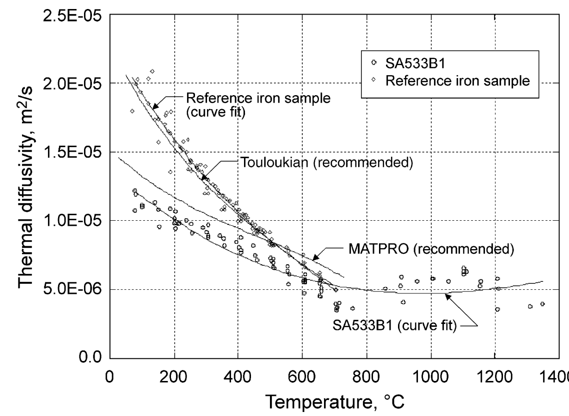

Fig. 11. Comparison of INL SA533B1 thermal diffusivity data, reference iron sample data, and values provided in the literature.

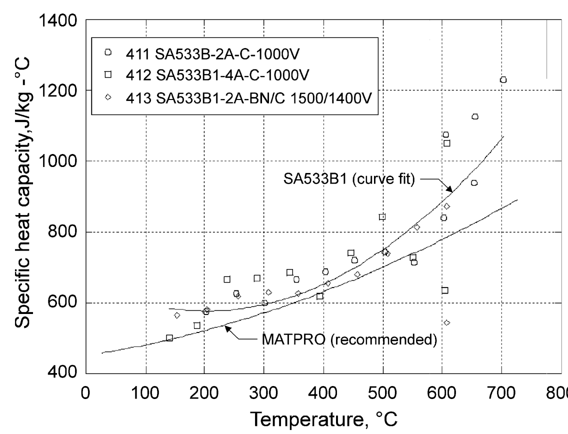

Fig. 12. Comparison of INL SA533 specific heat capacity data and MATPRO data.

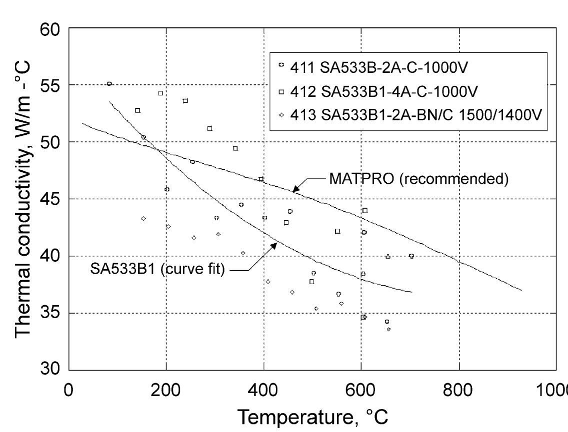

Fig. 13. Comparison of INL SA533B1 thermal conductivity data and MATPRO data.

Table 1
Comparison of FL5000 TDM results with available literature data

Material	Data source	Thermal diffusivity (m^2/s)				Thermal conductivity (W/m^2 °C)			
		20 °C	600 °C	1000 °C	1200 °C	20 °C	600 °C	1000 °C	1200 °C
SS304	Reference	3.7E−6	5.5E−6	6.5E−6	7.2E−6	15	24	29	33
	INL	3.7E−6	5.2E−6	5.6E−6	5.8E−6	20	25	27	28
SA533B1	Reference	1.5E−5	5.5E−6	NA	NA	52	44	NA	NA
	INL	1.2E−5	5.2E−6	5.2E−6	5.2E−6	57	38	NA	NA

[10]. Data also indicate that the general behavior of thermal diffusivity for carbon steel is more similar to that of iron than the stainless steel reference data. Fig. 11 compares the SA533B1 data with data obtained for an Anter reference iron sample, Touloukian iron thermal diffusivity data (from Fig. 4), and the recommended SA533B1 diffusivity data (based on MATPRO data from Fig. 5). As shown in Fig. 11, the newly obtained iron data are consistent with data found in Touloukian. The new SA533B1 data are similar (but somewhat lower) than data recommended by MATPRO. More scatter occurs in data obtained at higher temperatures.

Using software provided by Anter, the specific heat capacity and thermal conductivity of the SA533B1 samples were estimated using comparative techniques and recommended material property data for reference iron. Figs. 12 and 13 illustrate results obtained using these techniques. Recommended curves (see Fig. 5 from MATPRO information) are also plotted in these figures.

Curvefits from data estimated with the Anter comparative software yield similar values to the recommended values in Fig. 5. However, there is considerable scatter in the data. More testing is needed to reduce the uncertainty in the specific heat capacity and the thermal conductivity of this material (especially at higher temperatures). Results from this effort suggest higher specific heat capacities and lower thermal conductivities for SA533B1 at temperatures above 500 °C.

4. Summary

Laser-flash thermal diffusivity techniques were applied to obtain thermal property data up to 1400 °C for metals used in LWR vessels. Low temperature results (less than 1000 °C), as summarized in Table 1, indicate that the measured diffusivity data behavior is similar to data available in the literature (at least for those materials where data were available in the literature). High temperature diffusivity data obtained for stainless steel differed from values reported in the literature (although it should be noted that literature values were extrapolated for temperatures above 1000 °C).

Figs. 14 and 15 compare curvefits for the newly obtained thermal diffusivity and thermal conductivity data obtained for these materials. As shown in Fig. 14, diffusivity values for the SA533B1 experienced a minimum at temperatures greater than 700 °C, which corresponds to where a phase transition occurs in this material. Values for the thermal diffusivity for these materials at high temperatures (e.g., greater than 1000 °C) are similar (ranging from 0.05 to 0.065 cm^2/s).

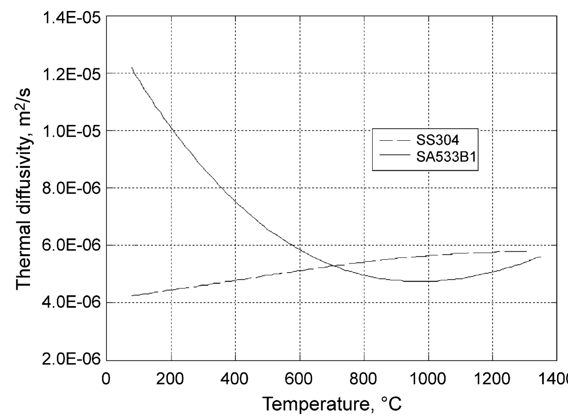

Fig. 14. Comparison of thermal diffusivity data for materials tested.

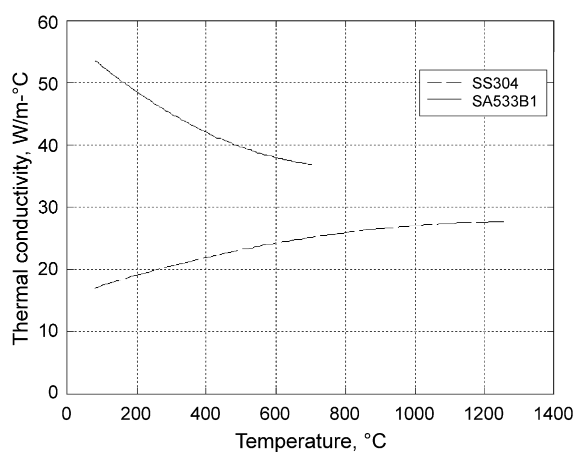

Fig. 15. Comparison of thermal conductivity data for materials tested.

As suggested by lower temperature data in the literature, curves in Fig. 15 show that the temperature-dependent behavior of these two materials differs. As temperature increases, the thermal conductivity for SS304 increases but the thermal conductivity of SA533B1 decreases.

5. Product disclaimer

References herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the US Government, any agency thereof, or any company affiliated with the Idaho National Laboratory.

Acknowledgements

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References

[1] Idaho National Engineering and Environmental Laboratory, SCDAP/RELAP5-3D© Code Manuals, INEEL/EXT-02-00589, Idaho National Engineering and Environmental Laboratory, May 2002.
[2] Sandia National Laboratory, MELCOR Computer Code, Version 1.8.6, <www.melcor.sandia.gov>, accessed December 8, 2006.
[3] Fauske Associates, Incorporated, Modular Accident Analysis Program, Version 4, <www.maap.com>, accessed December 8, 2006.
[4] W.J. Parker, R.J. Jenkins, C.P. Bulter, G.L. Abbot, J. Appl. Phys. 32 (9) (1969) 1679.
[5] L.M. Clark III, R.E. Taylor, J. Appl. Phys. 34 (7) (1979) 1909.
[6] Y.S. Touloukian et al., Thermophysical Properties of Matter, IFI/Plenum Publishing, New York, 1973.
[7] Anter Laboratories, Inc., Declarations of Reference Material Conformity, Pittsburgh, PA.
[8] D. Peckner, I.M. Berstein (Eds.), Handbook of Stainless Steel, McGraw-Hill Book Company, New York, 1977, p. 19–36.
[9] J.C. Spanner, et al., Nuclear Systems Material Handbook, TID-26666, April 1976.
[10] G.E. Korth, Metallographic and Hardness Examinations of TMI-2 Lower Pressure Vessel Head Samples, NUREG/CR-6194, March 1994.