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A thermal conductivity and electromotive force measurement system for nuclear fuels and materials

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ABSTRACT

The development of advanced nuclear fuels requires detailed understanding of their transmutation and micro-structural evolution. Alloy fuels have the advantage of high thermal conductivity and improved characteristics in fuel-cladding chemical reaction. However, information on thermodynamic and thermophysical properties is limited. The objective of this work was to develop an experimental system, integrated with thermal conductivity measurement capability to measure the thermodynamic properties of solid materials, from which an understanding of their phase change(s) can be determined. With the coupled system, both thermal conductivity and electromotive force (EMF) may be measured. In order to validate the system, the apparatus was employed to measure the EMF of several materials. As an initial calibration test, the EMF of Chromel was measured from 100 °C to 800 °C and compared with theoretical values. Subsequent EMF measurements were made for pure iron, iron-nickel alloy, and ANSI 1018 carbon steel rods. The measured phase transition temperatures were compared with the corresponding alloy equilibrium phase diagrams. The results indicate that the system is able to determine material phase change based on EMF measurement. In the future, this prototype system is to be adapted for hot-cell use on irradiated samples.

1. Introduction

Advanced nuclear fuels are essential to transmute highly radioactive and long lived transuranic isotopes in order to close the nuclear fuel cycle. An important aspect of fuel development is characterization of the performance and behavior of transmuted nuclear fuels, which is closely related to the temperature-dependent material structure of the alloy fuels. The methods for revealing material phase diagrams may be categorized as either computational or experimental. While the computational method (CALPHAD) [1] predicts/extrapolates the phase equilibria by applying Gibbs energy minimization, the experimental techniques are the foundation for the computation and provide inputs for the optimization of the phase diagram.

Many experimental techniques can be used to determine the thermodynamic properties of a material such as calorimetric measurement [2], gas phase equilibria techniques [3] and electromotive force measurement [4]. Among these methods, the EMF measurement technique has unique advantages. When a thermal cell is built, it involves a direct measurement of system EMF and Seebeck coefficient (thermopower or thermoelectric power) of a sample material when it is contacted with a reference conductor whose thermoelectric properties are known. When a sample material undergoes a pressure induced or temperature led phase transformation, both its structural and electronic properties change correspondingly. The Seebeck coefficient generally presents an anomalous behavior in the form of an abrupt variation corresponding to the change of material structure [5–11].

Another advantage for the EMF measurement adopted in this research is the integrated measurements with thermal conductivity. An advanced comparative axial heat flow technique for thermal conductivity measurement has been developed in this laboratory [12–15]. With the available system, the thermal EMF can be measured directly without incurring any additional cost and equipment. Compared to other measurement systems, the overall system design is unique to provide possible measurement of both thermal conductivity and specimen EMF up to high temperatures (~1000 °C). The results provided in this paper demonstrate the utility of the unique design approach where control of the temperature gradient is provided by the natural gradient in the tube furnace, a radiative heat sink, and small heaters placed in the assembly.

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The objective of this study is to describe a laboratory-based experimental system for measurement of material thermodynamic properties. The system is to be combined with thermal conductivity measurements and used in a controlled environment such as a glove box or radioactive hot cell. Calibration of the system using N type and S type thermocouple (TC) elements as reference materials was performed with respect to several samples. A calibration test on a material with no phase transition was done using a Chromel wire, one leg of the K type TC. Tests performed on materials with phase change(s) in their solid state include: (i) pure iron, a pure element; (ii) iron-nickel alloy, a binary alloy; and (iii) ANSI 1018 carbon steel, a multiple component alloy. At a later stage, the measurement system will be adapted for the measurement of material structure-dependent thermal properties of alloy fuels.

2. Measurement principle and system

An electric potential (EMF) is generated in a metal or semiconductor when there is a temperature difference. The Seebeck coefficient [$V\,K^{-1}$], a measure of the voltage produced with a unit temperature difference is expressed as $S = \Delta V/\Delta T$ where ΔV is the measured EMF [V] and ΔT is the temperature difference [K]. The EMF depends only on material and temperature difference but not on the dimension of the conductor.

The thermopower determination is usually conducted in comparison to a reference because the leads to the voltmeter generate another EMF. Fig. 1 presents an illustration of the configuration for thermopower determination [16], with the help of TCs. Two TCs are injunction with the unknown material X, with a temperature difference ΔT in between. The circuit EMF, ΔV_{XR} measured from either one of the two TC leads, is attributed to the combined potential difference of two materials (reference and unknown). The thermopower of the unknown material, S_X, can be expressed as

$$S_X = S_R - \Delta V_{XR}/\Delta T \quad (1)$$

where S_R is the thermopower of the reference material whose value is taken from Ref. [17] with the addition of the absolute thermopower of platinum [18,19]. The measured sample thermopower is usually considered as the material property at the mean sample temperature $T + \Delta T/2$ [20]. The magnitude of temperature difference, ΔT, is limited by the desired accuracy of thermopower in Eq. (1) and the resolution and precision of the equipment used in determining the temperature and electric potential. The applied temperature difference is typically in the range of 2–5 K [20,21].

The measurement system, which was used for thermal conductivity measurement by comparative axial heat flow method, is employed in the EMF measurement (Fig. 2). Detailed description of the system and thermal conductivity measurements of unknown samples are described elsewhere [12–15]. This paper presents only an introduction of the system and results of the EMF measurement side.

The sample and reference bars are surrounded by insulation material to minimize lateral heat loss, both then being enclosed by the guard. Heaters on one side increase the temperature of the test stack and guard and heat sink on the other side cools the unit. An axial temperature gradient is created allowing the thermal conductivity determination by the TC temperature measurements. The system is placed in a MTI OTF-1200X tube furnace to get the required ambient temperature, and the desired temperature differences in the sample and reference bars are built up and maintained by the heaters, driven by a dual-loop Eurotherm 3504 controller. Inert gas is backfilled into the tube furnace to protect the measurement system after the air is purged by a vacuum pump.

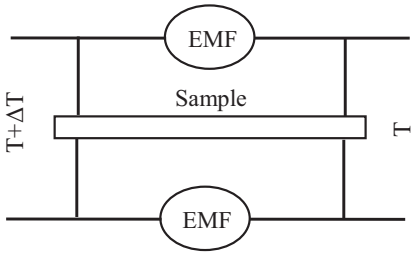

Fig. 1. Schematic illustration for thermopower measurement.

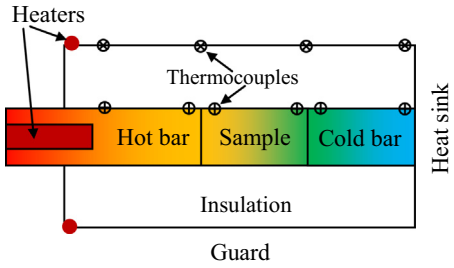

Fig. 2. Illustration of measurement system for thermal conductivity and thermopower.

Thermal conductivity is measured at intermittent temperatures because of the steady state requirement. The EMF measurement, however, is conducted continuously in a quasi-static way. With a controlled temperature difference on the sample set by the controller, the furnace temperature is gradually ramped up or down to change the environment. When passing known phase transition zones, a slower heating rate was employed. TCs are connected to an Agilent 34970 data acquisition unit for simultaneous temperature and EMF measurement as shown in Fig. 1.

To apply the computation of Eq. (1) and quantify the measurement error, a piece of Chromel wire was first measured in the furnace. This Chromel wire, taken from a TC element, is not representative to the fuel pellet but has well-defined thermopower value for comparison. In this first measurement, the temperature difference between the two ends was not controlled, thus it varied from 5 °C to 30 °C and changed sign. This represents extreme conditions that the later on measurements were not reached. N type TCs, in conjunction with the Chromel wire were used as reference material for the EMF measurement. The TC elements are all purchased from Omega.

Three types of rod samples, having similar dimension to the fuel pellets, were measured for EMF. The first specimen was a 99.99% pure iron-nickel alloy that has a low transition temperature. Having the same dimensions, the second and third specimens were, respectively: 99.95% pure iron and a piece of commercial ANSI 1018 carbon steel. The high purity metal and alloys were purchased from ESPI Metals. The carbon steel was purchased from McMaster-Carr.

3. Results and discussion

Fig. 3a presents the measured thermopower variation of Chromel with respect to change of temperature. For comparison, the values from literature are superimposed on the same figure. It indicates that the measured values closely follow the literature ones even with relatively large and variable ΔT from both the positive and negative legs of the N type TC (NP or NN) measurements. The two large spikes in measured thermopower near 200 °C are caused by the change of sign of temperature difference, i.e. the hot and cold ends of the specimen were reversed. With a temperature difference approaching zero, Eq. (1) is affected by the mea-