Seebeck coefficient measurements on Li, Sn, Ta, Mo, and W

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A B S T R A C T

The thermopower of W, Mo, Ta, Li and Sn has been measured relative to stainless steel, and the Seebeck coefficient of each of these materials has then been calculated. These are materials that are currently relevant to fusion research and form the backbone for different possible liquid limiter concepts including TEMHD concepts such as LiMIT. For molybdenum the Seebeck coefficient has a linear rise with temperature from $S_{Mo} = 3.9\ \mu V\ K^{-1}$ at 30 °C to 7.5 $\mu V\ K^{-1}$ at 275 °C, while tungsten has a linear rise from $S_W = 1.0\ \mu V\ K^{-1}$ at 30 °C to 6.4 $\mu V\ K^{-1}$ at 275 °C, and tantalum has the lowest Seebeck coefficient of the solid metals studied with $S_{Ta} = -2.4\ \mu V\ K^{-1}$ at 30 °C to $-3.3\ \mu V\ K^{-1}$ at 275 °C. The two liquid metals, Li and Sn have also been measured. The Seebeck coefficient for Li has been re-measured and agrees with past measurements. As seen with Li there are two distinct phases in Sn also corresponding to the solid and liquid phases of the metal. In its solid phase the $S_{Sn-solid} = -1.5\ \mu V\ K^{-1}$ at 30 °C and $-2.5\ \mu V\ K^{-1}$ near the melting temperature of 231 °C. There is a distinct increase in the Seebeck coefficient around the melting temperature as the Sn melts and stays relatively constant over the rest of the measured temperatures, $S_{Sn-melt} = -1.4\ \mu V\ K^{-1}$ from 235 °C to 275 °C.

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1. Introduction

In order to address many of the concerns associated with solid divertor materials, research is ongoing into flowing liquid metal divertor concepts [1]. These would be able to replenish and repair themselves by virtue of their liquid nature. However, flowing molten metals in high magnetic fields suffer from MHD drag effects, potentially requiring prohibitively high pumping power to drive the flow. The thermoelectric magnetohydrodynamic, or TEMHD, effect, particularly as implemented in the LiMIT device under development at the University of Illinois at Urbana-Champaign (UIUC), may enable flowing liquid metal films to be implemented in high magnetic field devices by providing a driving force within the liquid metal itself [2,3]. Crucial to the operation of any device employing TEMHD is the thermoelectric current generated by the temperature gradient between the top and bottom of the device and required knowledge of the Seebeck coefficient. In order to aid in the evaluation of the suitability of different materials for TEMHD devices, thermopower of Li, Sn, Ta, Mo, and W was measured relative to 316 stainless steel and the results are presented below.

The Seebeck effect refers to the phenomena that the voltage across the junction of two metals is a function of temperature. This is the mechanism by which thermocouples measure temperature, because if the relationship between temperature and voltage is known, the temperature may be ascertained by reading the voltage across such a junction. Mathematically, the Seebeck effect is described by:

$$\Delta V = -S\Delta T \tag{1}$$

$$E = S\nabla T \tag{2}$$

where the proportionality S is the relative Seebeck coefficient, or thermopower, between two materials. Fig. 1 shows an example thermocouple circuit. If we denote the absolute Seebeck coefficient, of materials A and B as S_A and S_B respectively. The relative thermopower, or relative Seebeck coefficient is given by:

$$S_{B-A} = S_B - S_A \tag{3}$$

Since S is a function of temperature for virtually all junctions, (3) may then be substituted into (1) directly only for small temperature differences. For larger temperature differences, an integral over T is required. As mentioned previously, the Seebeck coefficients of a variety of materials are important to the operation of thermoelectric magnetohydrodynamic devices. Thermoelectric magnetohydrodynamic devices rely on the interaction of a thermoelectric current with a magnetic field to drive flow of a liquid metal. Research into the use of these devices as potential divertor concepts in magnetic confinement fusion devices is ongoing, and it is crucial to the understanding of these devices to have knowledge of the Seebeck coefficients of the materials used. It was the intention of the authors, therefore, to assay many potential materials in order to expand the choices for the constituent materials of such a device.

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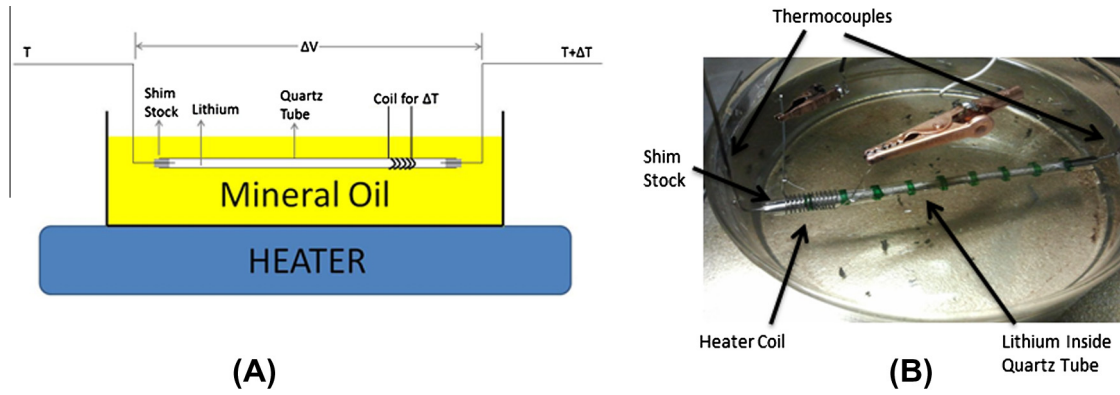E-mail address: Fiflis1@illinois.edu (P. Fiflis).

Fig. 1. (a) Schematic of the apparatus. (b) Photo of the apparatus.

2. Experimental setup

The Seebeck coefficient measurements performed at UIUC were performed in an apparatus designed and built at UIUC. Several important concerns were to be addressed with the design of the apparatus. First, it is important that a temperature gradient be established across the material to be investigated. It is also important that the bulk temperature of the material be adjustable as well to investigate the dependence of the Seebeck coefficient on temperature. Another concern is proper measurement of the small voltage established across the sample. The experiments performed were conducted in an oil bath placed on top of a ceramic hot plate. The setup consisted of a wire of diameter of approximately 2 mm of the material to be analyzed suspended in the mineral oil (Multitherm PG-1). A quartz tube with inner diameter of 3 mm and length 75 mm is placed over the wire to be analyzed. This quartz tube serves two purposed, one, to contained materials with low melting points that will liquefy during a portion of the test, and two, to insulate the material from a small heater coil. This small heater coil is used to induce a temperature gradient along the wire. The heater coil is a nichrome wire wrapped into a 5 turn solenoid. About 1A of current is run through the wire to establish the

temperature gradient, however, this current is adjustable via a variable resistor that is adjusted to maintain a temperature difference of about 8 K across the wire. The average temperature of the wire is adjusted via the ceramic hot plate which heated the entire oil bath. The temperature difference and voltage across the sample were then measured via a pair of stainless steel coated thermocouples. Wires attached to the stainless steel coats measured the voltage while the thermocouple measured the temperature. The temperature difference and voltage were tracked using a LabJack U6 data acquisition device and a pair of LabJack Tick-InAmps. The voltage signal and thermocouple voltages were both amplified. The data was then compiled and analyzed with the aid of LabView program written specifically for the task. Since the Seebeck coefficient is a function of temperature, it was recorded vs. the average wire temperature. This process is the differential thermopower measurement described in [5]. A photo of the apparatus and a schematic are shown in Fig. 1.

3. Results

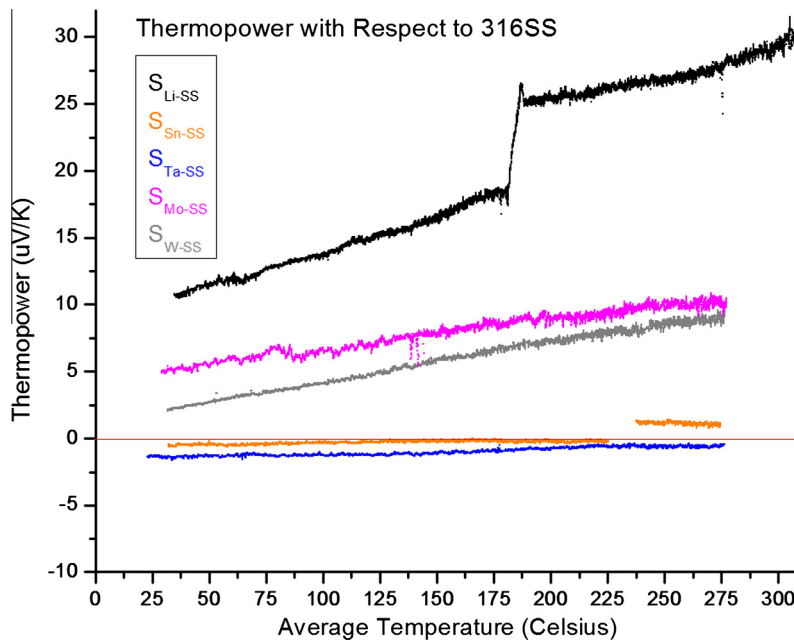Measurement of the Seebeck coefficient of any material requires that there be a junction between the material to be

Fig. 2. Relative thermopower of various materials vs. *T* referenced to 316 stainless steel.
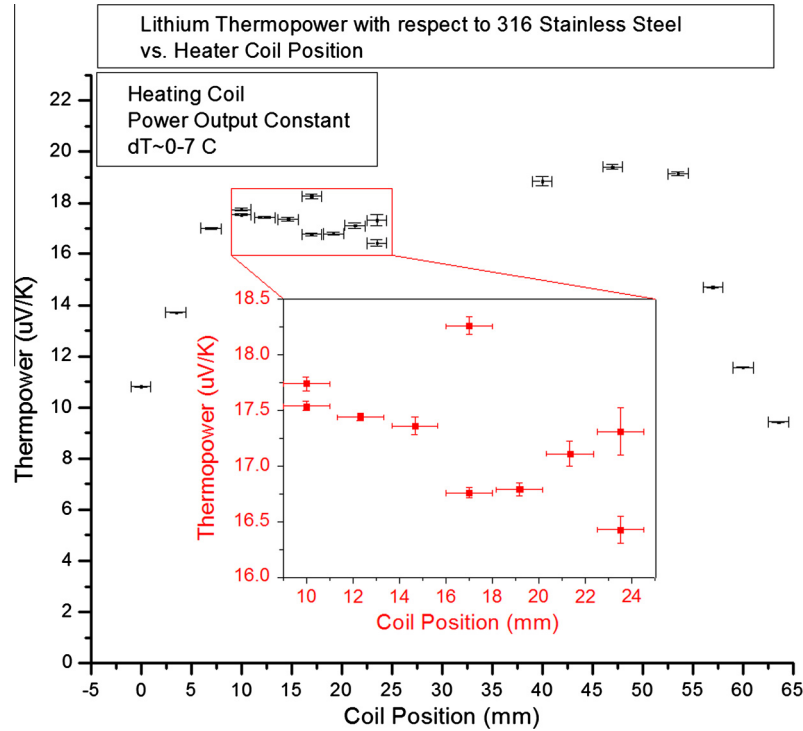
Fig. 3. Lithium–stainless steel thermopower vs. coil position.

investigated and some reference material. The Seebeck coefficient is then measured relative to this reference and then the Seebeck coefficient of the reference is subtracted out, yielding the absolute thermopower of the material. All of the materials measured herein where measured relative to stainless steel, and are presented as S_{x-SS}, to signify the relative thermopower between material x and stainless steel. To find the relative thermopower between two arbitrary materials then, one need only subtract the two Seebeck coefficients, i.e. the relative thermopower between materials x and y is given by:

$$S_{x-y} = S_{x-SS} - S_{y-SS} \qquad (4)$$

An equation for the absolute Seebeck coefficient of stainless steel is given by [6]:

$$S_{SS} = 2.5269 - .0143T + 8E - 6T^2 \qquad (5)$$

where T is in Kelvin, and which may then be used to determine the absolute thermopower of any of the materials presented here by proper implementation of (1). The measured Seebeck coefficients are shown in Fig. 2.

Results show that for the solid metals the trend is almost linear with temperature. For molybdenum the Seebeck coefficient rises with from $S_{Mo} = 5.0 \ \mu V \ K^{-1}$ at 30 °C to 10.0 $\mu V \ K^{-1}$ at 275 °C. Tungsten from $S_W = 2.0 \ \mu V \ K^{-1}$ at 30 °C to 8.8 $\mu V \ K^{-1}$ at 275 °C, and tantalum has the lowest Seebeck coefficient of the solid metals studied with $S_{Ta} = -1.5 \ \mu V \ K^{-1}$ at 30 °C to $-0.5 \ \mu V \ K^{-1}$ at 275 °C. The Seebeck coefficient for Li has been re-measured and agrees with past measurements. The Seebeck coefficient for Sn has two distinct phases, the $S_{Sn-solid} = -0.5 \ \mu V \ K^{-1}$ at 30 °C and 0 $\mu V \ K^{-1}$ near the melting temperature of 231 °C. There is a distinct jump in the Seebeck coefficient around the melting temperature as the Sn melts, $S_{Sn-melt} = 1.0 \ \mu V \ K^{-1}$ from 235 °C to 275 °C.

Second order polynomial fits of the above data yield the following approximations for the absolute Seebeck coefficient of each material, with T in K, and S in $\mu V \ K^{-1}$:

Lithium:

$$S_{Li} = \begin{cases} 7.5156 - .0169T + 8.1E - 5T^2 & \text{for } 298 < T < 448 \\ -4243.173 + 17.614T - 0.018092^2 & \text{for } 448 < T < 453 \\ 67.6979 - .1985T + 2.21E - 4T^2 & \text{for } 453 < T < 580 \end{cases}$$

$$(6)$$

Molybdenum:

$$S_{Mo} - 2.2718 + .024T - 1.13E - 5T^2 \quad \text{for } 298 < T < 550 \qquad (7)$$

Tantalum:

$$S_{Ta} = 1.2904 - .0169T + 1.56E - 5T^2 \quad \text{for } 298 < T < 550 \qquad (8)$$

Tungsten:

$$S_W = -6.2834 + .0252T - 3.9E - 6T^2 \quad \text{for } 298 < T < 550 \qquad (9)$$

Tin:

$$S_{Sn} = \begin{cases} -.7812 - 5E - 4T - 7E - 6T^2 & \text{for } 298 < T < 502 \\ -55.561 - .00466T + 2.18E - 4T^2 & \text{for } 502 < T < 510 \\ -85.226 + .330T - 3.24e - 4T^2 & \text{for } 510 < T < 550 \end{cases}$$

$$(10)$$

Most show a positive trend of S vs. T, however, one feature in particular stands out. Both the curves for lithium and tin show a jump in the Seebeck coefficient. This is indicative of the melting point of each material, as these jumps occur at 180 °C and 231 °C which are respectively the melting points of lithium and tin. Previous experiments by Bidwell [7], Kendall [8], and Surla [4] also investigated the Seebeck coefficient of lithium. The measurements conducted by Surla [4] were conducted with an experimental setup nearly identical to ours, the apparatus in both cases being the same device. It was found by Surla [4] that the measured Seebeck coefficient was heavily dependent on the position of the heater coil. This dependence was further investigated yielding the graph shown in Fig. 3. This graph shows that when the coil was placed

near the edge of the sample, the measured Seebeck coefficient was lower than when the heater coil was placed nearer the middle. It was found that a large region existed away from the edges where the Seebeck coefficient was relatively stable with regard to heater coil position, as shown by the zoomed in portion of Fig. 3. Data for the Seebeck coefficient with the heater coil near the middle of the sample was hard to obtain due to the difficulty in establishing a temperature difference between the two ends of the sample. However, there appears to be a strong edge effect near the ends of the sample. Therefore, the Seebeck coefficients reported here are using a coil position that reproduces the results of Surla [4] which in turn match the results given by Kendall [8]. This position was originally chosen to maximize the temperature gradient across the sample obtained for a given current through the heater coil and is located 5 mm from the end of the sample for the duration of each test.

Previously, measurements of the Seebeck coefficients of both lithium and tungsten were compiled by Shercliff [9]. However, due to the disagreement between the measurements by Bidwell [7] and Kendall [8], the Seebeck coefficient of both lithium and tungsten were re-measured in order to confirm their accuracy. The values measured agree with those reported by Shercliff to within a couple $\mu V\,K^{-1}$. The measurements taken here confirm the results of Surla [4] and Kendall [8] over those of Bidwell [7].

4. Discussion

Since the motivation for these experiments was in support of a TEMHD device, the most relevant results are the relative thermopowers between each of the liquids and each of the solids. For example, the relative thermopower between lithium and tungsten is proportional to the driving TEMHD force in a device whose base was made of tungsten and whose working fluid was lithium. Comparing the thermopower of Li–W and Li–SS, we can see that using tungsten would provide about 2/3 of the driving force that stainless steel would provide given the same temperature gradient, $S_{Li-W} = 19.0\ \mu V\,K^{-1}$, $S_{Li-SS} = 28.4\ \mu V\,K^{-1}$ at 250 °C. It can also be seen that tin would also work as a working fluid, but would almost certainly need to be used with tungsten or molybdenum. $S_{Sn-W} = -8.3\ \mu V\,K^{-1}$ and $S_{Sn-SS} = 1.1\ \mu V\,K^{-1}$ at 250 °C. Note that the relative thermopower for the Sn–W system is negative. This means that in a TEMHD system, the direction of the tin flow in tungsten trenches would be opposite that for lithium in the same trenches. Lithium has shown promise as a potential plasma facing component material in fusion devices [1], and a TEMHD system involving lithium has been demonstrated to self-pump lithium through a series of trenches [2]. This system uses stainless steel as the trench material, however, the results above show that such a device would also work with tungsten, molybdenum, or tantalum. Tantalum and stainless steel offer significant thermopowers, however, there are issues with their ability to survive in a fusion environment. Tungsten and molybdenum offer more durable options. And even though the thermopower would be reduced, it may be necessary to use these materials.

5. Conclusions

The Seebeck coefficient of a variety of materials that could potentially be used in a TEMHD liquid metal divertor concept was measured. Two potential candidates for the liquid metal were investigated, lithium and tin. Four potential substrate candidates, molybdenum, tungsten, stainless steel, and tantalum were also investigated. For molybdenum from $S_{Mo} = 3.9\ \mu V\,K^{-1}$ at 30 °C to 7.5 $\mu V\,K^{-1}$ at 275 °C. Tungsten $S_W = 1.0\ \mu V\,K^{-1}$ at 30 °C to 6.4 $\mu V\,K^{-1}$ at 275 °C, and tantalum $S_{Ta} = -2.4\ \mu V\,K^{-1}$ at 30 °C to $-3.3\ \mu V\,K^{-1}$ at 275 °C. For Sn, $S_{Sn-solid} = -1.5\ \mu V\,K^{-1}$ at 30 °C and $-2.5\ \mu V\,K^{-1}$ near the melting temperature of 231 °C and $S_{Sn-melt} = -1.4\ \mu V\,K^{-1}$ from 235 °C to 275 °C above the melting temperature. Of all of these, lithium had the highest Seebeck coefficient, Stainless steel and tantalum provide the highest thermopower for a flowing lithium TEMHD device, $S_{Li-SS} = 28.4\ \mu V\,K^{-1}$ and $S_{Li-Ta} = 28.8\ \mu V\,K^{-1}$ at 250 °C, however concerns about the durability of these materials, particularly tantalum, in a fusion environment, may prohibit their use in such a device. Molybdenum and tungsten would provide a lesser thermopower, but the durability of these materials might necessitate their use. Tin may also work as a flowing liquid with these materials, Sn–Mo, and Sn–W would have significant thermopowers, and the flow would be in the opposite direction of lithium.

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