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## Thermoelectric Power Measurements on Some Chevrel Phase Superconductors

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The absolute thermoelectric power measurements on  $\text{AgMo}_6\text{S}_8$ ,  $\text{Mo}_6\text{Se}_8$ ,  $\text{PbMo}_6\text{S}_8$ ,  $\text{SnMo}_6\text{S}_8$ ,  $\text{SnMo}_6\text{Se}_8$  were made from 4.2 to 300 K. All the compounds have positive thermoelectric power near room temperature.  $\text{Mo}_6\text{Se}_8$  and the compounds containing small metal ion such as Cu and Ag show a negative peak and the compounds containing large metal ion such as Pb, Sn show a positive peak in the thermopower vs. temperature graph. There seems to be a correlation between low temperature peak in the thermopower and delocalisation of the metal ion in the compound. Two mechanisms may account for the peaks in the thermopower viz. phonon drag effect and the enhancement of thermopower caused by the renormalisation of energy, velocity and relaxation time due to electron-phonon interaction. In the present case the mechanism mentioned later does not seem to provide an explanation for the negative peaks in the thermopower.

### 1. INTRODUCTION

Thermoelectric power measurements on some Chevrel phase compounds such as solid solutions of  $\text{Cu}_{1.8}\text{Mo}_6\text{S}_{8-y}(\text{Se/Te})_y$  [1],  $\text{PbMo}_6\text{S}_8$  and  $\text{EuMo}_6\text{S}_8$  [2] have already been reported. In the case of the solid solution and  $\text{PbMo}_6\text{S}_8$ , the data on the temperature variation of thermopower has been analysed as the sum of an electron diffusion term  $S_e$  and a phonon drag term  $S_{ph}$ . From the contribution due to the electron diffusion term, the energy derivative of the logarithm of the density of states at the Fermi level has been found out using Mott-Wilson formula [3].

In this paper we have reported the temperature variation of thermopower for the compounds  $\text{SnMo}_6\text{S}_8$ ,  $\text{SnMo}_6\text{Se}_8$ ,  $\text{Mo}_6\text{Se}_8$ ,  $\text{AgMo}_6\text{S}_8$ . In all the cases except  $\text{AgMo}_6\text{S}_8$  the measurements were carried out in the temperature range 4.2 to 300 K.

### 2. EXPERIMENTAL RESULTS

The preparation and characterisation of the compounds have been reported by Umarji [4]. The temperature variation of thermoelectric power has been found out for the compounds under study using the experimental set-up described in [5]. Figs. 1 and 2 show the results obtained. The thermopower data on  $\text{PbMo}_6\text{S}_8$  [2] and  $\text{Cu}_{1.8}\text{Mo}_6\text{S}_8$  [1] have also been included in Fig. 1 and Fig. 2 respectively for the purpose of comparison.

To investigate whether pelletisation produces any irreproducible effect on the thermopower data, measurements were carried out on the same sample of  $\text{Mo}_6\text{Se}_8$  pelletised twice. From the Fig. 2 it is evident that the agreement between the two runs is good.

### 3. DISCUSSION

From Figs. 1 and 2 it is clear that in all these Chevrel phase compounds under study the thermoelectric power is positive at the high temperature end of the measurements. In the

case of all large cation compounds the value of thermoelectric power always remains positive and also exhibits a hump in the low temperature range. For  $\text{PbMo}_6\text{S}_8$  the peak occurs around 35 K; for  $\text{SnMo}_6\text{S}_8$  between 40 and 65 K and for  $\text{SnMo}_6\text{Se}_8$  between 50 and 60 K. On the other hand in the case of  $\text{Mo}_6\text{Se}_8$  and small cation compounds, the thermoelectric power changes sign when the sample is cooled down from room temperature. Besides this, the thermoelectric power also exhibits a minimum in the low temperature range. Preliminary measurements on  $\text{AgMo}_6\text{S}_8$  indicate a behaviour similar to that in  $\text{Cu}_{1.8}\text{Mo}_6\text{S}_8$ .

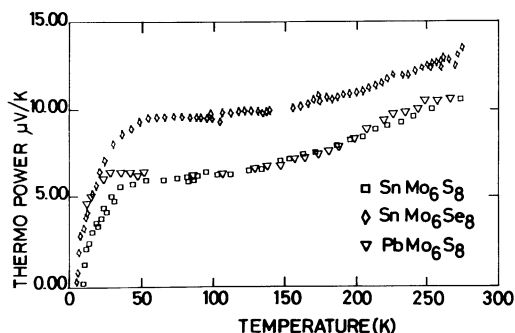


Fig. 1. Temperature variation of thermopower for  $\text{SnMo}_6\text{S}_8$ ,  $\text{SnMo}_6\text{Se}_8$  and  $\text{PbMo}_6\text{S}_8$ .

From Figs. 1 and 2 it is evident that there exists a correlation between the low temperature thermoelectric power and the delocalisation of cation from the inversion centre. In the case of  $\text{Cu}_{1.8}\text{Mo}_6\text{S}_8$  and  $\text{AgMo}_6\text{S}_8$  where the delocalisation is relatively more [5], the low temperature thermoelectric power exhibits a negative peak and in the case of  $\text{PbMo}_6\text{S}_8$  and  $\text{SnMo}_6\text{S}_8$  where the delocalisation is comparatively less the low temperature

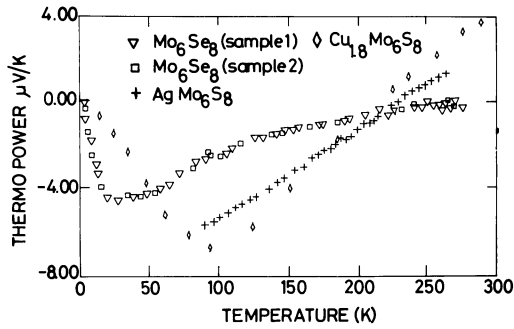


Fig. 2. Temperature variation of thermopower for Mo<sub>6</sub>Se<sub>8</sub>, Cu<sub>1.8</sub>Mo<sub>6</sub>S<sub>8</sub> and AgMo<sub>6</sub>S<sub>8</sub>.

thermoelectric power exhibits a positive peak.

The thermoelectric power can be written as a sum of the electron diffusion term,  $S_e$ , and the phonon drag term,  $S_{ph}$ . At high temperature the electron diffusion term dominates the thermopower and this can be written as,

$$S_e = \alpha T \quad \dots \dots \dots (1)$$

where  $\alpha$  is given by

$$\alpha = \frac{\pi^2 k^2}{3|e|} [(d(\ln N_d)/dE) - (d(\ln A)/dE)]_{E_F} \quad \dots \dots (2)$$

according to Mott [3]. Here  $N_d(E)$  is the number density of states in the d-band and A the area of the Fermi surface. If one assumes the second term in Eq. 2 to be negligible, then  $(d(\ln N_d)/dE)_{E_F}$  can be found out from the thermoelectric power measurements. Because thermoelectric power is positive for all the materials at the high temperature end, neglecting  $d(\ln A)/dE$  term in Eq. 2 will result in positive value for  $d(\ln N_d)/dE$ . The Fermi level will be placed on the rising part of the density of states curve. However, the band structure calculations of Nohl et al. [7] place the Fermi surface on the falling part of the density of states curve for many of these materials. This implies  $d(\ln A)/dE$  at  $E_F$  is negative for these materials.

There are two mechanisms which can explain the peaks observed in the low temperature range: one is phonon drag effect and the other is due to the enhancement of diffusion thermopower due to the renormalisation of energy, velocity and relaxation time caused by electron phonon interaction [8].

Assuming phonon drag effect is operative its contribution can be determined by subtracting the electron diffusion term from the total thermoelectric power. The sign of the phonon drag peak depends on whether the scattering process is N or U process. This will depend on the relation of the Fermi surface to the Brillouin zone.

Kaiser [9] has shown that the renormalisation of electron energy, velocity and relaxation time can explain the positive peak in the thermoelectric power of some of the solid solutions  $Cu_{1.8}Mo_6S_{8-y}(Se/Te)_y$  in which the residual resistance ratio,  $R_0/R_{RT}$ , is high. However, the negative peaks observed in the small cation Chevrel phase compounds can not be explained on the basis of renormalisation effect unless one makes extreme assumptions on the velocity renormalisation factor.

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