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Effect of Surface Polishing on the Thermoelectric Power of InSb Crystals

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Abstract. The effects of surface polishing on the thermoelectric properties of InSb crystals have been studied. For this purpose, we grew high - quality InSb crystals. This crystal has the real surface and we call it "as prepared". Also, a cleaned surface was obtained by polishing processes. A detailed work for comparing the measured thermoelectric in the pre-mentioned two cases was done. This included the study of the crystals by means of X- ray diffraction and the scanning electron microscopy images (SEM). The goal is to find out the influence of the surface treatment on the values of thermoelectric power (α). This was done over a wide range of temperatures (200-500K). Unique- results were obtained for the first time first time, and were represented using useful histograms.

***Keywords*:** InSb, Crystal Quality, Semiconductors, Physical Parameters, Thermoelectric Properties, X- ray Diffraction, Surface Polishing.

1. Introduction

Among III–V compound semiconductors, InSb has the smallest band gap, measuring 0.17 eV at 300 K that corresponds to IR wavelength (7.3 μm) and the highest carrier mobility. Because of these properties, InSb is suitable for fabricating high- speed electronic components, magnetic sensors and infrared photodetectors [1-4]. As for the importance of this compound and attempts done to improve its quality, it attracts many investigators to grow this unique semiconductor. T. Duffar et al. [5] used the Bridgman growth without crucible contact using the de-wetting phenomenon for the growth of antimonide semiconductors. Growth and characterization of indium antimonide crystals were made by N.K. Udayashankar and H.L. Bhat [6]. InSb quantum dot LEDs were grown by P.J. Carrington et al. [7]. The InSb crystal was grown by Se-Hwan Park et al by using the Bridgman method at various crystal growth speeds [8].The quantum Hall effect at cleaved surfaces of InSb was studied by R. Masutomi et al. [9]. Electrical conduction mechanism and photon-generated carrier recombination process in amorphous InSb films were studied by Yanping Yao et al [10].) Band structures of narrow gap semiconductors in terms of the coulombic polaron effect were studied by J.D. Fan et al [11]. H. Simchi et al. [12] studied passivation of the InSb surface for manufacturing infrared devices. The InSb wafers were cleaned with CP4A etchant (HNO_3:CH_3COOH: HF: H2O at ratios of 2:1:1:10). Mesa etching characterization of InSb for high - density image array applications was studied by Kow-Ming Chang et al. [13]. The main concern in our crystal growth lab is growing semiconductor crystals and investigating their physical properties. The most important problem in such kind of crystal is the crystal defects, which defects limit the crystal quality. This is why we are interested in the improvement of the crystal quality. Since literature survey proved that the present work is presented for the first time, we undertook this research to shed light on the influence of the polishing process on the thermoelectric power and hence find out the influence of the surface conditions on some estimated physical parameters (required for applications).

2. Experimental Procedure

We can sum up the Experimental Procedures in the following points:

2.1 Crystal Growth

The phase diagram of InSb [14] which has already been published was considered for the growth of InSb crystals. It is also an established fact that high-quality crystals can also be obtained by the Bridgman technique [15]. This is the reason why the crystal growth was done by a modification of the Bridgman technique which is close to the known traveling solvent method (TSM) technique [16]. The growth ampoules for the experiments were quartz (12 mm diameter and 20 cm length). One end of quartz tube was sealed. The quartz tube was cleaned by the known procedures prior to loading the charge. The ampoule was designed with a

proper conical tip at its bottom to facilitate nucleation process needed for crystal growth. Then the filled ampoule was sealed under a dynamic vacuum of~10^{-5} Torr. After that, the sealed ampoule was loaded into the furnace. The temperature profile was adjusted to melt the feed materials. The temperature gradient was measured with accurate K-type thermocouples during the growth. The ampoule was heated up to for 24 h to guarantee a homogeneous distribution of the materials in the melt. Lowering rates selected were in the range 2 mm h^{-1} and the temperature gradient was near 12 K cm^{-1}. Even after solidification, the lowering rate was maintained at the same rate till the entire ampoule was out of the furnace. Details about the applied technique have already been published [17, 18].The principal difficulty experienced in these methods is the extremely required slow growth rate.

2-2 X-ray Diffraction

Localized variations in the intensity within any individual diffracted spot arise from structural effects or uniformity in the lattice planes causing the spot, and this forms the basis for the x-ray topographic technique. This topographic contrast arises from differences in the intensity of the diffracted beam as a function of position inside the crystal. The difference between the intensities diffracted from one region of the crystal which diffracts kinematically to another which diffracts dynamically is one of the ways that dislocations can be rendered visible in topography.

After weeding out the product crystals, they were examined by two different investigations. The first analysis is the x-ray diffraction which was measured stepwise with angle, second value of 2Θ at ambient temperature which is measured with a model D 5000 Siemens diffractometer (Germany) in central metallurgical institute Cairo Egypt. This identification is important because it revealed that the grown crystal has a cubic structure with lattice parameter equal to 6.477 ˚A without any secondary phases. This is in a good agreement with ASTM and JCPDS-ICDD No. 6–208 [19].

2-3 Scanning Electron microscopy

The grown crystals were analyzed in central laboratory of in (Egypt-Qena) using a JEOL JSM-5500 LV scanning electron microscope (SEM), (JEOL, Japan). The operating working voltage for the Scanning electron microscope was 5 kV.

2-4 Mechanical Treatment (polishing)

For mechanical polishing the obtained crystal was cut into samples, using a Discoplan-TS Struers (Struers, Denmark), for cutting and grinding of thin sections. The sample was prepared for polishing with carborundum of three different grain sizes (coarse, medium, and fine to glass finish). After this process, the crystals were cleaned with distilled water and allowed to dry without heat treatment. The objective of the mechanical polishing was to remove the saw damage, obtain flat surface and improve the surface quality.

2-5 Thermoelectric power Measurements (TEP)

The cylindrical shape of the produced crystal was suitable for the purpose of TEP measurements. Only the length was adjusted to be 0.3 cm by polishing. The crystal diameter was 1 cm. A two-part calorimeter was used. The inner part acted as a holder on which the crystal was mounted on a flat end of a copper cylinder. It was heated electrically (the flat end was insulated from the crystal by thin sheets of mica). The second part of the calorimeter acted as a jacket to keep the measurements under a vacuum of about 10^{-3} torr with the aid of Edward rotary pump (England). The outer part was also used as the main source of heating. The applied vacuum was very important because it prevented crystal oxidation in the case of high-temperature measurements and eliminated water vapor condensation in the case of low- temperature measurements (which were done using liquid nitrogen). By using two temperature control elements (type Elcont and Eliwell- Italy) we could control the temperature of the two heaters, and keep a temperature difference along the crystal fairly stable. The temperature difference was kept less than 5 K. Two calibrated thermocouples (made of copper-constantan) served as a temperature-sensing element between the two ends of the crystal. Measurements of temperature and thermo-voltage were carried out simultaneously. During measurements we considered the following:

1) The temperature of the crystal was the average of those at the two ends of the sample.

2) To establish the temperature and equilibrium state around the crystal a relatively long time was needed.

3) The used method for measuring α is the differential method (not the integral one).

4) Silver past was used as Ohmic contacts.

3. Results and Discussion

3-1. X-Ray Diffraction Results

The X - ray diffraction pattern of InSb crystals, for as prepared is shown in Fig. (1). Samples of indium antimonide powder, taken from a single crystal sample, prepared in our laboratory (Qena) were identified by X - ray diffraction for comparison. Also another chart for X - ray diffraction pattern, in the case of treatment of the grown crystal by polishing, was obtained. It is not appeared to avoid figures crowd. It is shown that the two charts indicate a single crystalline phase structure without any secondary phases.

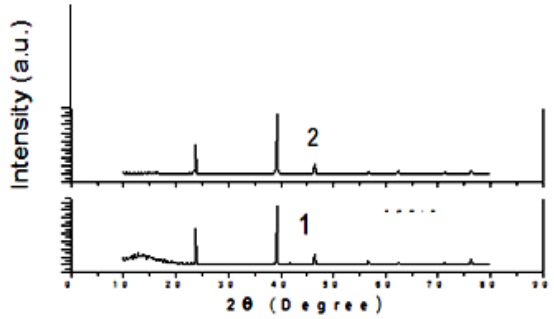

Fig. (1). X-ray diffraction pattern for the case of the as prepared crystal (1) and the polished sample (2).

From the X - ray diffraction data we obtained the following:-

-The crystal structure are stills the same because peak positions are the same.

-The reflecting planes (111), (220), (311), (400), (331), (422) and (511) are observed for both the as prepared and the polished crystals. Also, the main peak (100) is noticed and was used to compare the crystal qualities.

-Since the difference between the intensities diffracted from one region of the crystal which diffracts kinematically to another which diffracts dynamically is one of the ways that dislocations can be rendered visible in topography so, careful look to the peak intestines in the X - ray diffraction patterns gives evidence about the formation of dislocations when the crystal was etched.

- Diffraction line broadening is also observed in the case of polishing which can have many physical sources: size, strain, coherent precipitates, and misfit between phases with different chemical compositions, etc. Hence more work is recommended strongly to reveal what happened definitely to the crystal structure of the compound.

- It is clear from the last two points that as prepared crystal quality is better than the quality of the polished one.

3-2. SEM of the understudy samples

The morphology of the two samples (as prepared and polished) as observed under SEM is presented in figure (2). It can be observed that defects are formed on the surface of the polished sample while the as prepared one looks with less abnormality. This is may due to the deviation from the stoichiometry to include the whole sample surface. These results confirm the results obtained from XRD.

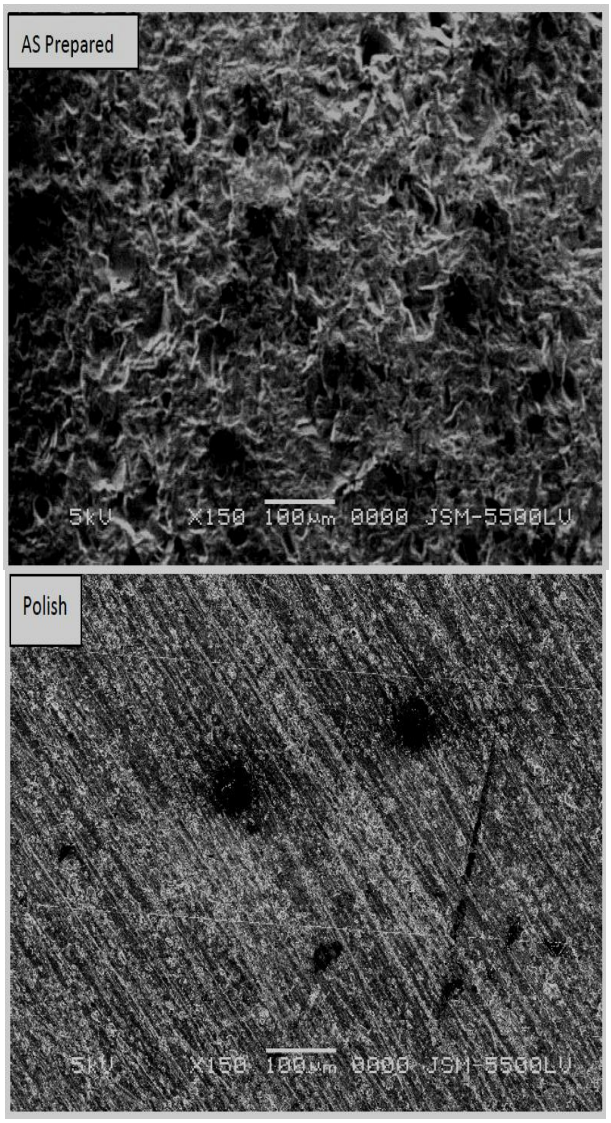

Fig. (2). SEM of the understudy samples.

It must be mentioned that the two images were taken under the same conditions. Accordingly one can't exclude the saw effect which usually causes surface damage.

3-2. Results of the thermoelectric power (TEP)

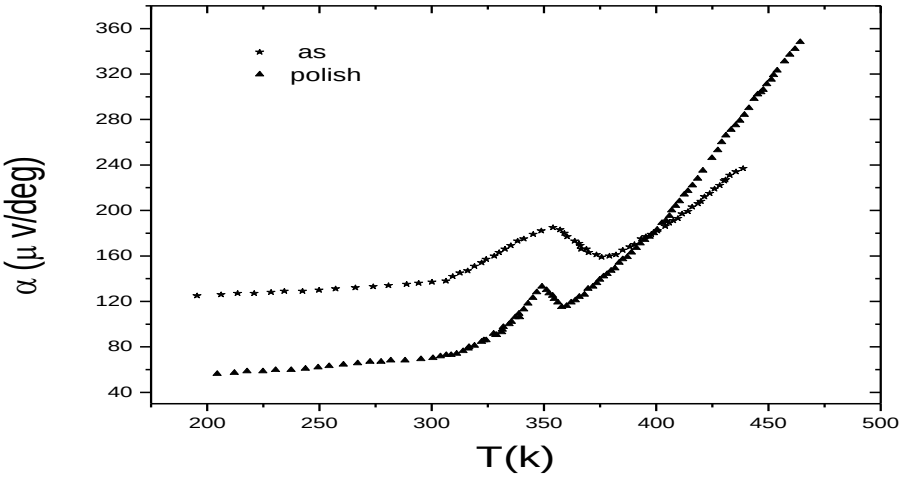

Fig. (3). Variation of the thermoelectric power α against temperature

The thermoelectric power or TEP (α value) was studied in a wide range of temperatures (200-500K) for both the as prepared and polished crystals. The obtained results and the above figure (which contains two figures) indicate the following:-

The general behavior of the curves is the same and it contains:-

- α starts to be fairly constant till reaches room temperature.

- TEP gradually increases with the temperature rise till about 350 K.

- At 350 K, α possesses a peak where this is the transition temperature as we believe.

The monotonic rise of α in this range is due to the fact that in the extrinsic conductivity range and the expected affect of temperature or the thermal activation on the minority carriers starts.

- Finally above 350 K, the behavior is characterized by the diminution in α values till about 375 K after which it increases very rapidly.

- The decrease of α above 350 K is argued to the influence of mixed condition as the intrinsic range is approached.

The first impression about the behavior of α against T in both cases yields that nothing change in the behavior except for making upward or downward shift to values of α.

Comparison between the as prepared crystal and the mechanically treated by polishing indicates that α is shifted downward and hence its value is decreased by polishing, as shown in Fig. (3).

Now we come to the estimation of many important physical parameters the case of the as grown and polished crystals. In order to do this we must mention the following:-

1-The present research is a complementary part of the electrical conductivity and Hall effect work we already have done

1- Figure (3) is redrawn to justify the relation between (α -1/T) to apply an important formula in the intrinsic range. [20]

$$\alpha = -\frac{k}{e}\left(\frac{\mu_n-\mu_p}{\mu_n+\mu_p}\left(\frac{\Delta E_g}{2KT}+2\right)+\frac{3}{4}\ln\frac{m_n^*}{m_P^*}\right) \quad \ldots\ldots\ldots\ldots\ldots\ldots (1)$$

Whereμ_n, μ_p, m_n^* and m_P^* are the electron mobility, the hole mobility, the electron effective mass, and the hole effective mass, respectively. The above expression predicts the (α -1/T) plot should be a straight line. We computed the values(μ_n/μ_p) from the slope. Also, the ratios m_P^*/m_n^* were found from the value m_P^* intercept of the curves. The value ΔE_g (the width of the energy gap) equals 0.17 eV at room temperature was used from the previous Hall effect and electrical conductivity data to deduced the above values. This enables us to find the values μ_n also.

2- Fig. (3) is redrawn again as a plot of α against In T to justify another formula that was suggested by Wilson for utilizing the data of α in the extrinsic region [19]:

$$\alpha = \frac{K}{e}\left(2-\ln\frac{ph^3}{2(2\pi m_p^*\ KT)^{3/2}}\right) \quad \ldots\ldots\ldots\ldots\ldots\ldots (2)$$

This can be written in the form:

$$|\alpha| = \frac{3}{2}\frac{K}{e}\ln T + \text{constant} \quad \ldots\ldots\ldots\ldots\ldots\ldots (3)$$

Thus a plot of α against lnT, for a particular sample, should be a straight line of slope $\frac{3}{2}\frac{K}{e}$.

From the intercepts of the lines (in the impurity region) with the axis, we got m_p^* see in tables (1). Using the ratio m_p^* / m_n^*= see in tables (1) previously obtained from the last relation we could evaluate m_n^* see in tables (1). The slopes of the straight line should be 3K/2e which have the numerical value 129 μV- deg^{-1}. The slope of the plots of α against ln T is seen in tables (1). This discrepancy may be due to the possibility that the effective masses of the charge carriers are temperature dependent. Thus a slow variation of the effective masses with temperature would have a large effect on the mobility variation.

The value of the relaxation time for holes, as deduced, see in tables (1), while it for electrons see in tables(1). Also, the diffusion constants for holes and electrons are calculated. They are D_p and D_n see in tables (1). Among the important parameters computed in this investigation is the diffusion length for both carriers. The diffusion length L_p (for holes as referred by the sub - index P) and the diffusion length L_n = see in tables (1).

Table (1). Comparison between the Physical Parameters in cases as prepared and Polished

Physical Parameters	Sample	
	As prepared	**Polished**
α (μV . deg $^{-1}$) at ٣٥٠K	130	70.1
μ_p	140.7797	584.7226
μ_n	295.405	856.051
m^*_p kg	1.1925E-34	1.72817E-34
m^*_n kg	1.23E-36	3.69E-37
τ_p S	2.0985E-13	1.26313E-12
τ_n S	4.53408E-15	3.94588E-15
D_p $cm^2\ s^{-1}$	3.632116	15.08584
D_n $cm^2\ s^{-1}$	7.62145	22.0861
L_p cm	8.73E-07	4.37E-06
L_n cm	1.86E-07	2.95E-07
α (μV . deg $^{-1}$) at ٣٥٠K	185.35	134.28
m^*_p / m^*_n	97.11768	468.655
The slope of (α - Ln T)(μV . deg $^{-1}$)	122	114.6
μ_n / μ_p	2.09835	1.464029

4. Conclusions

From this work we concluded the following remarks:

- InSb is a very sensitive semiconductor to the surface conditions.
- With mechanical treatment (polishing), we produced a smoother surface (because we removed the saw effect) but a high density of scratches was produced.
- Variation of the charge carriers and the mobility results in a change in TEP.
- Surface treatment is a good tool to improve the crystal properties and the thermoelectric coefficient values. Hence we recommend opening up future work for the effect of another treatments (etching for example).

• The crystal physical parameters are strongly influenced by the effect of surface treatment.

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تأثير تلميع سطح مركب الخواص أنتمونيد الأنديوم البللوري علي الظاهرة الكهروحرارية

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مخلص البحث. للمكانة المتميز لمركب أنتمونيد الأنديوم البللوري في علوم المواد و خاصة في مجال توليد الكهرباء من التدرج الحراري المعروفة باسم الظاهرة الكهروحرارية ؛ فإن البحوث الجارية حالياً –علي النحو الوارد في مقدمة البحث – تهتم بتطوير ذلك المركب الفريد. ومن المعلوم أن التلميع / السنفرة (Polishing) هي إحدي المعالجات السطحية للمواد والتي تؤثر علي خواصها ؛ لذا كان هذا البحث. و قد سار البحث وفقاً للخطوات التالية–:

- تحضير المركب في صورته البللورية باستخدام تقنية جديده تعتبر تطويراً لطريقة بريجمان المعروفة.
- استخدام طريقة حيود الأشعة السينية X – ray diffraction للتأكد من سلامة الطور البللوري للمركب قيد الدراسة.
- دراسة الظاهرة مرتين للمركب قبل ويعد المعالجة.
- اشتقاق العديد من الثوابت الفيزيائية للمركب قبل ويعد المعالجة لبيان تأثير المعالجة السطحية عليها.
- عرض النتائج في جدول واضح في ختام البحث لسهولة المقارنة ومن ثم نوقشت النتائج السابقة تفصيلاً علي النحو الوارد في متن البحث.