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Surface Tension and Contact Angles of Molten Cadmium Telluride[1]

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The surface tension and contact angle of molten cadmium telluride (CdTe) were measured as a function of temperature by the sessile drop technique. A FORTRAN code was developed to calculate the surface tension of sessile drops, with the contact angle ranging from 0 to 180°. The wetting of cadmium telluride melt was studied on different surfaces. The surface tension of cadmium telluride was about 160 ± 5 dynes $\cdot$ cm^{-1}[1.6N $\cdot$ m^{-1}.] at the melting point of 1093°C. The contact angle of CdTe melt was about 65° on a quartz optical flat, 75° on commercial fused quartz, and 125° on boron nitride coated quartz.

KEY WORDS: cadmium telluride; contact angle; melting point; surface tension; vapor pressure.

1. INTRODUCTION

Cadmium telluride (CdTe) crystals have important applications in radiation detectors [1–8], infrared sensors [1, 3, 4], laser windows [1, 3], and solar cells [8–10]. The fabrication of these devices requires large, single crystals with low defect densities. However, current technology in the growth of these crystals on earth is limited to small crystals with a high concentration of defects such as dislocations, twins, low-angle grain boundaries, and precipitates.

Terrestrial crystal growth techniques are affected by gravity-related effects, notably buoyancy-driven convection and possibly self-deformation, which can result in undesirable qualities in crystals. In this context, crystal

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growth in space has many potential advantages. However, while adverse effects of gravity are eliminated in a microgravity environment, surface tension effects, notably Marangoni[3] effects, assume prominence [11, 12]. The prediction of the extent of the Marangoni effect necessitates a thorough knowledge of the surface tension data of molten CdTe at different temperatures. Surface tension data are also required in order to calculate the maximum stable length of a liquid column.[4] Furthermore, a complete study of the wetting properties of CdTe melts should help in understanding the "rib-like" [13] and wavy structures [14] observed on the surface of crystals grown in space. The extent of wetting also indicates the probability of impurity incorporation from the walls of the container to the melt and the free energy of heterogeneous nucleation.

Surface tension and wetting data of CdTe melt are essential in the design of flight hardware for the growth of these crystals in space. To our knowledge no attempt, with the exception of the effort led by Offisterov [15], has been made to fill the need for surface properties of CdTe melts. In this work, we measured the surface tension of molten CdTe by the sessile drop method with provision for high-temperature experimentation. The temperature range between which the measurements were made was 1093 to 1150°C. The contact angle also was measured on fused quartz optical flats, commercial fused quartz flats, and boron nitride-coated flats. Other surfaces are currently under study.[5] The surfaces were chosen on the basis of the current usage in semiconductor materials processing.

2. SESSILE DROP TECHNIQUE

The sessile drop technique is one of the most widely used methods for the measurement of surface tension [16]. This is due to the accuracy that can be obtained with this method, plus the advantage of simultaneously obtaining the contact angle of the liquid on that substrate. It is also easily adaptable for a high-temperature [17], high-pressure closed environment. Although the technique of sessile drop method is well documented, we give a brief description of the technique to maintain continuity of the text. In this technique, a drop of liquid whose surface tension is to be measured is placed on a clean, smooth horizontal surface (Fig. 1). In the case of molten solids a few pieces of the solid are taken instead and then melted to form the drop, as was done in this work. The shape of the drop bears a definite relationship to the surface tension of the liquid forming the drop.

[3] Surface tension-driven convection.

[4] In the float-zone technique of crystal growth, it is necessary to know the length of a stable liquid column in order to design an apparatus.

[5] Pyrolytic boron nitride, hydrogen fluoride (HF)-etched surfaces, etc.

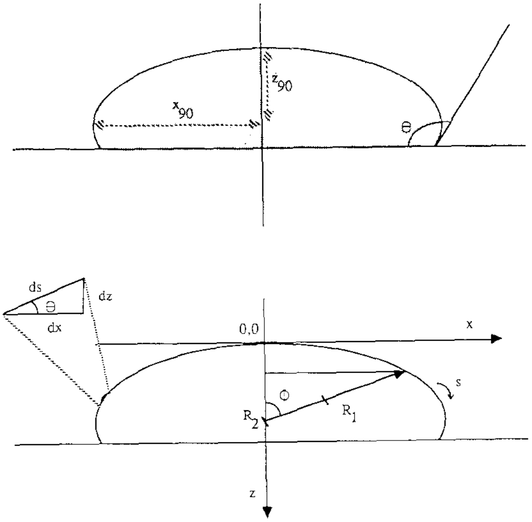

Fig. 1. A typical sessile drop.

The pressure across a curved fluid–fluid interface is related to the surface tension by the Laplace equation, upon which the mathematics of surface tension calculations rests:

$$\gamma\left(\frac{1}{R_1}+\frac{1}{R_2}\right)=\Delta P \tag{1}$$

where R_1 and R_2 are the principal radii of curvature (m), γ is the surface tension ($\mathrm{N\cdot m^{-1}}$), and ΔP is the pressure difference across the interface ($\mathrm{N\cdot m^{-2}}$). Bashforth and Adams applied the Laplace equation to the calculation of the shape of a sessile drop of liquid on a level solid plate and arrived at

$$\frac{1}{\left(\frac{R_1}{b}\right)}+\frac{\sin\phi}{\left(\frac{x}{b}\right)}=2+\beta\frac{z}{b} \tag{2}$$

where ϕ is the angle made of the tangent at any point on the contour of the drop to the horizontal, β is the shape factor, and b is the radius of curvature (m) at the apex of the drop. Details of work on the mathematics of surface tension calculations can be found in the literature (e.g., Refs. 18–25). The surface tension data were obtained by a curve-fitting method to relate the shape of the drop to Eq. (2).

3. DIFFICULTIES

Surface properties are in themselves difficult to obtain due to numerous sources of error. This difficulty is further compounded in the case of a substance like CdTe due to its properties. CdTe melts at 1093°C. At its melting point stoichiometric cadmium telluride exhibits $8 \times 10^4\ \mathrm{N \cdot m^{-2}}$ Cd pressure and $3.8 \times 10^4\ \mathrm{N \cdot m^{-2}}$ Te pressure. The effects of the vapor pressure become pronounced from 300–400°C onward. The high vapor pressure, extreme proclivity to oxidize, and toxic nature of CdTe require the use of a completely sealed enclosure to house the drop of molten CdTe. It was observed in a number of experiments that even traces of oxygen or water vapor drastically changed the results of wetting by CdTe melts. Another problem frequently encountered was vapor transport. Cadmium telluride does not exist in the vapor form. On sublimation it decomposes according to the following equation:

$$2\mathrm{CdTe} \rightarrow 2\mathrm{Cd} + \mathrm{Te_2}$$

Vapor transport to the cooler walls of the container and subsequent condensation was a major problem. Condensation on the walls obscured the drop. This necessitated the use of excess elemental cadmium[6] to create a cadmium overpressure in the ampoule and suppress transport of the vapors to the wall. Furthermore, the high temperatures involved complicated the recording of an analyzable drop on photographic film. Surface tension and contact angle are both highly sensitive to impurities. It was hence necessary to minimize impurities both in the CdTe and on the substrate.

4. EXPERIMENTAL DETAILS

The experimental setup consisted of a cylindrical quartz ampoule with optical flats at both ends and a horizontal substrate on which the CdTe was placed. A sketch of the experimental setup is given in Fig. 2. CdTe pieces were placed inside the ampoule and the ampoule was sealed under a pressure of 10^{-5} Torr ($1.3 \times 10^{-3}\ \mathrm{N \cdot m^{-2}}$) after backfilling a number of times with a mixture of 5% hydrogen–helium gas. The ampoule was then placed on the sample holder, which was a quartz boat inside the furnace in the central zone of a tube furnace. The furnace used for this research was a three-zone, split-tube, horizontal silicon carbide unit (Thermcraft Inc.). Temperature control was achieved by a digital PID controller (Wahl Instruments) driving an SCR (Eurotherm) for each of the three zones. Overtemperature control provision was also made. Photographs were

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[6] Exactly 1.4×10^{-4} kg was used for the 4-in.-long ampoule and 7×10^{-5} kg was used for the shorter ampoule of 2 in.

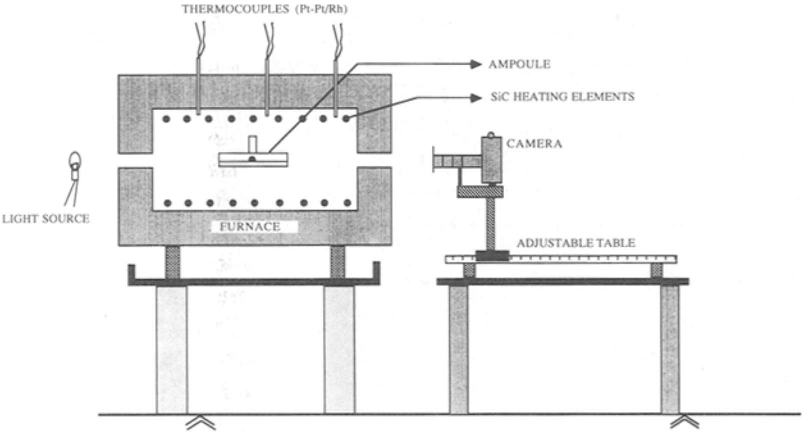

Fig. 2. Sketch of the apparatus.

taken with a Nikon FM2 camera with a Nikon F-200 lens and Nikon TC-301 teleconvertor. A gelatin filter (Kodak 38-A) was used to filter the red light emitted from the furnace.

A typical experimental run consisted of melting the CdTe pieces to form a single drop. The temperature was gradually increased in increments of 5°F (2.8°C). After allowing time for thermal equilibrium at each temperature, the drop profile was recorded photographically. The photographs were digitized using a Sigma Scan digitizer pad. After proper dimensioning the drop profile data were used in the computer program to get the surface tension of the melt at that temperature. Contact angles were obtained directly from the photographs of the drops.

5. RESULTS

5.1. Surface Tension

The results are shown in Figs. 3 and 4. The computed surface tension of CdTe ranged between 160 dynes$\cdot$cm^{-1} (1.6 N$\cdot$m^{-1}) at 1093°C and about 150 dynes$\cdot$cm^{-1} (1.5 N$\cdot$m^{-1}) at 1150°C on the two different types of fused quartz. Anomalous behavior was noticed in the surface tension data for CdTe on boron nitride-coated quartz (discussed later).

5.2. Contact Angle

Contacts angles were obtained on three different substrates. They are shown in Figs. 5, 6, and 7. On a fused quartz optical flat, molten CdTe had

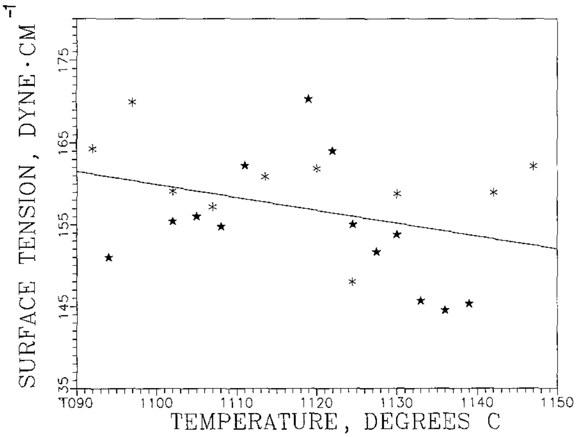

Fig. 3. Surface tension of cadmium telluride on fused quartz. Each symbol represents different experimental runs.

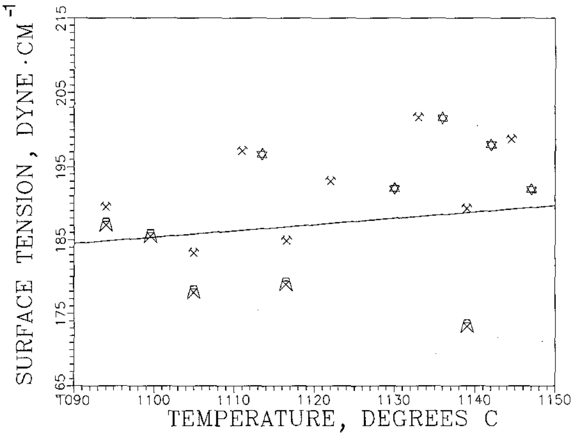

Fig. 4. Surface tension of cadmium telluride on boron nitride-coated quartz. Each symbol represents different runs under identical experimental conditions.

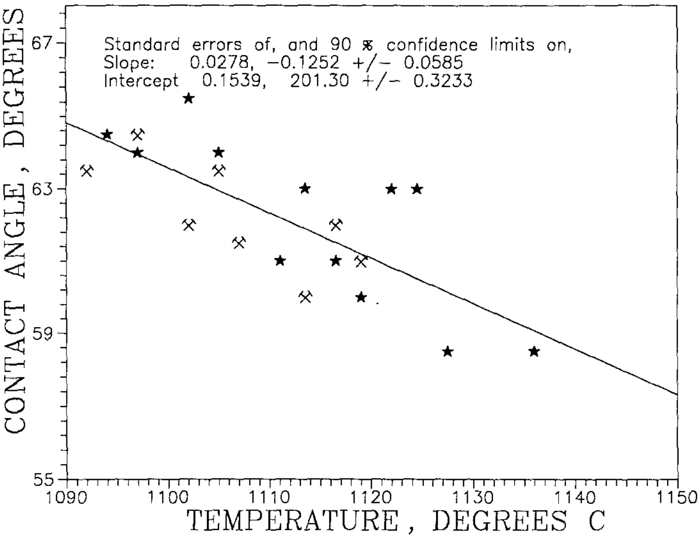

Fig. 5. Contact angle of cadmium telluride on fused quartz (optical flat). Each symbol represents different runs under identical experimental conditions.

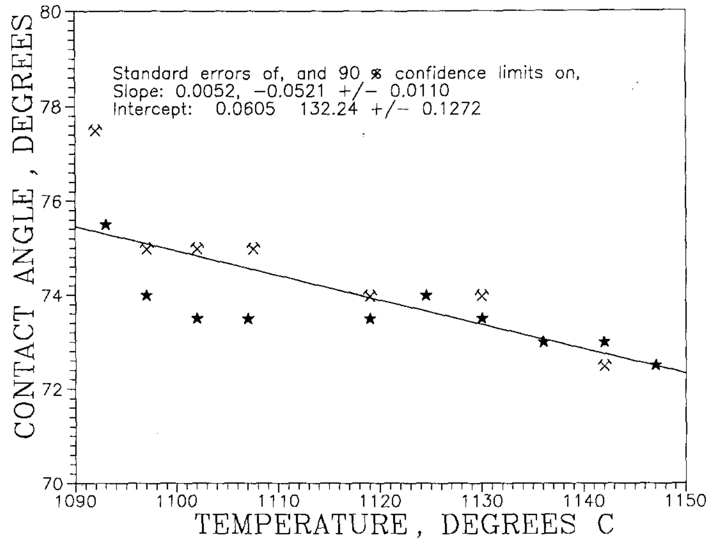

Fig. 6. Contact angle of cadmium telluride on fused quartz (commercial). Each symbol represents different runs under identical experimental conditions.

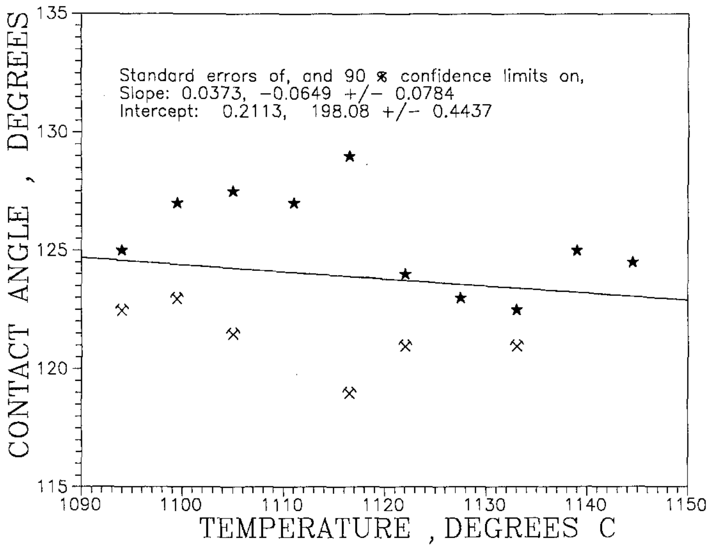

Fig. 7. Contact angle of cadmium telluride on a boron nitride-coated surface. Each symbol represents different runs under identical experimental conditions.

a contact angle of approximately 65° at 1093°C. In contrast, boron nitride was nonwetting, with an observed contact angle ranging from 123 to 127°. On commercial fused quartz the contact angle was 75° at the melting point. Confidence limits are given in the respective figures.

6. DISCUSSION

Surface tension depends on temperature, composition, and surface charge. It is independent of the substrate on which the drop rests unless it is contaminated by the substrate. Hence, there should be no difference in the surface tension data obtained from our experiments on fused quartz and those on boron nitride-coated quartz. While the data in Fig. 3 follow the normal trend of fall of surface tension with temperature, it is difficult to ascertain a trend from the data in Fig. 4. A possible explanation for this anomaly is that one or both substrates contaminated the melt. In addition, the effect of excess Cd (added to control vaporization) has not been determined yet and could prove to be a major factor in explaining the scatter in the data.

Contact angle data establish the wetting of CdTe on quartz. The difference in the wetting of two different samples of quartz can be accounted for by SEM pictures of these surfaces, which were different.The SEM pic-

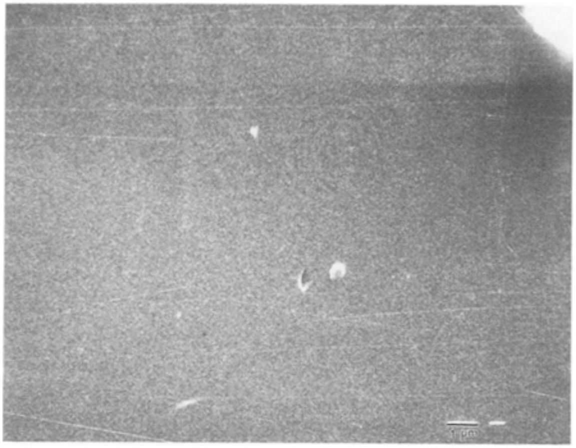

Fig. 8. SEM of fused quartz optical flat.

tures of these substrates are shown in Figs. 8, 9, and 10. From the SEM picture of the surface it is seen that the ground and polished quartz optical flat is extremely smooth in comparison to the commercially obtained quartz. The SEM picture of the boron nitride coated surface shows particles of roughly 5–10 μm. Also from the size of the particles and the accompanying "holes" and crevices, it is possible that the microscopic contact angle might be different from the macroscopically observed values. The macroscopic contact angle is an input to the computer program and

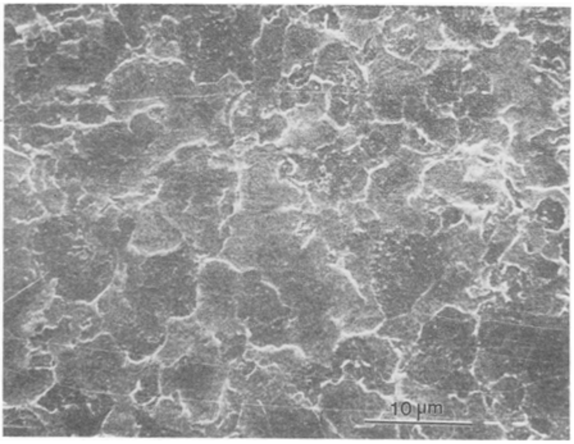

Fig. 9. SEM of commercial fused quartz flat.

Fig. 10. SEM of boron nitride-coated surface.

this could have been one of the reasons for the observed increase in the surface tension values.

Sources of error in this study can be found both in experimental procedure and in digitizing the drop profile. These errors probably explain the large scatter observed in the surface tension and contact angle data obtained in this study. The major sources of error are in the ampoule alignment, contamination of the melt, and digitization of the drop profile. In the calculations of the surface tension it is assumed that the drop is perfectly axisymmetric and the substrate horizontal. But with the present apparatus the substrate could be aligned only directly. This often resulted in drops that were not perfectly axisymmetric. Contamination of the melt by the substrate seems a plausible explanation to the anamoly[7] observed with boron nitride-coated surfaces. The deviation of the contact angle on different boron nitride surfaces can be explained in terms of the very rough surface of the boron nitride-coated substrate. The digitizer used in this study had an accuracy of 0.127 mm (1.27×10^{-4} m) and a resolution of 0.025 mm (2.5×10^{-5} m). However, the procedure of digitizing the drop is entirely dependent on the accuracy of the operator.

In recent experiments[8] with slightly varying Cd overpressure, the surface tension values were slightly higher (≈ 5–15 dynes $\cdot$ cm^{-1}) than the values obtained by the authors, which suggests that the original data did not suffer from any serious errors. The continuation of our work was

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[7] The difference in surface tension on quartz and boron nitride-coated quartz.

[8] These experiments are being conducted by Mr. Rajaram Shetty and Clarkson University.

to reduce the scatter in the data by improvements in the photgraphic technique and the digitization.

We have developed an experimental procedure for surface tension and contact angle of high-melting and moderate vapor pressure substances. From the above experiments we established that molten CdTe wets quartz but not boron nitride. Further studies are being continued on the wetting properties on other substrates, effect of stoichiometry on surface tension, and other semiconductor melts like GaAs, which also has a high vapor pressure and melting point. At the completion of this study a complete picture of the wetting properties of CdTe and GaAs should be available.

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