CHAPTER 2

EXPERIMENTAL TECHNIQUES

2.1 Introduction.

The various experimental techniques employed during the course of this work are briefly described here. For the quantitative and qualitative measurements of features on the crystal surfaces, the techniques of light profile and multiple beam interferometry have been used alongwith the Vickers Projection Microscope. Uniform silvering of a crystal surface is essential for better resolution and contrast. For this, silvering technique is used. It will also be described in this chapter. As the optical techniques mentioned above are well discussed in several standard text books. Only a brief description together with their salient features will be given here.

2.2. Vickers Projection Microscope (V.P.M.):

The microtopographical and interferometric studies were made with the help of Vickers Projection Microscope (inverted metallurgical type) (Fig. 2.1). The specimen to be observed is kept on a freshly movable stage above the objective and both the transmisson and reflecting systems can be used by simply moving the collimating system, up or down. This microscope consists of a universal illuminator, housing, a semi-reflecting plate, iris lens and the objective lenses. The collimating system comprises of powerful mercury or point-o-lite lamp, a condenser and an aperature controlled by an iris diaphragm. Translational movement in two mutually perpendicular directions of the lamp permits centering of the diaphragm. An image of the source can be formed on the field iris. Movement of the lamp and condenser perpendicular to the axis of the microscope, makes it possible to vary the convergence of light incident on the specimen.

For visual observations, an eyepiece with a reflector is pushed into the tube below the objective. For photomicrography, a projection eyepiece is used and the final image is formed after reflection in the projection mirror, in the plane of the screen. Very slight refocussing is necessary when the visual system is changed over to the projection system.

It should be noted that spherical aberration for high power objectives or lenses should be zero for clear image. Different kinds of lenses such as achromatic, fluorite and apochromatic were used to minimize spherical aberration for different wavelengths. Besides magnifications, resolving power of the lens should be high to detect two nearest points. Resolving power of a lens is proportional to its numerical aperature which can be increased by using liquid of high refractive index between objective and specimen. The fluid should be non-drying. Working distance (the distance between objective lens and specimen at the time of focus) should be less when oil is used to increase resolution. Specimens observed in the above method are usually spoiled due to oily contamination of the surface. Maximum limit of useful magnification is about 1000 X N.A. (numerical aperture). Useful magnification can be increased with oil immersion objectives whose focal lengths are generally less than or equal to 2 mm. The depth of focus also decreases with the focul length of the objective used.

V.P.M. used in the present study, has advantages and disadvantages over many other microscopes. The microscope, as such is easy to handle and has no restricted working conditions. A large number of accessories can be easily fitted with Vickers projection microscope, e.g. polarizing equipment, microhardness tester, phase contrast equipment, light profile, Fizean and Feco fringes. All these help in carrying out the study over a large varied field with equal ease and high sensitivity. In addition, both transmisson and reflecting systems can be utilized for transparent and opaque crystals without difficulty. Macro-and micro-examination of the specimens can be done with ease.

Of course, the magnification and the resolution obtained with an electron microscope is quite high, but this is only the lateral magnification. It does not give adequate normal resolution. The Vickers microscope with its light profile equipment or with the multiple beam interference technique, can be used to determine heights and depths of various features on crystal surfaces. The order of accuracy by the light profile is 0.1 micron, while with the interference method it is about $\lambda/2$ and under favourable conditions it can determine heights and depths of a few angstrom units. Depending upon the nature of the surface features, observed in the present work a judicious choice of objective, eyepiece and bellow distance was made for obtaining desired magnification and resolution.

2.3 Light Profile Microscopy:

The light profile technique of Tolansky (1952) is an improvement of Schmaltz's (1936) "light cut method". Here a parallel line graticule is fitted into the housing of field iris of the V.P.M. The images of the lines are formed on the surface of the specimen. However, if the metal tongue internal reflector which is a metallized sector of about 30° angle is used, it sends an off-centered beam of rays which is incident on the specimen at an angle of about 30°, when high power lens is used. When some irregularities are present on the specimen surface the images of the slit will no longer be straight, they will show some shift. This shift can be measured by a filar micrometer eyepiece fitted in the visual eyepiece tube. From the shift in the profile, the actual depth or height of the surface features can be easily calculated, if we know the values of the constants for different objectives used with the microscope. This constant is due to difference in magnification of profile and linear magnification of microscope. The constant quantity for a particular objective should be multiplied by linear magnification for obtaining magnification of profile. The values of the constant for different objectives used with the microscope in this work are as follows:

Objective	Oil immersion		
	2 mm	4 mm	8 mm
Constant	1	1.38	0.56

The present "light profile technique" has following advantages over the original "Schmaltz's light cut method".

1) The magnification was restricted to 400 diameters for light cut method, while in light profile it was up to 2000 diameters.

2) In light cut method, the surface having the irregularities was not visible, while in light profile it is clearly observed, hence interpretation of the results becomes easier.

3) For light cut technique, a special apparatus is required to be prepared with two microscopes at right angles to each other, while in the light profile method single lens is used for both illumination and viewing.

2.4 Multiple Beam-interferometry:

When a monochromatic light from an extended source, is directed to the interferometer, modified transmission Haidenger fringes are formed. If a lens is placed in the path of such a system of parallel rays, Airy summation at the focus of the lens is automatically obtained. The intensity distribution is no longer Cosine^2 type as in the case of two beam fringes, but is modified by the multiple beam combination. The whole fringe shape is quite independent of the absorption. The principal factors affecting the usefulness of the multiple beam fringes are the contrast and sharpness which in turn depend upon the reflectivity of the surfaces [Tolansky,(1948)]. However, an increase in the reflectivity, increases the absorption, thereby reducing the contrast of the reflecting system. Hence a compromise between reflectivity, transmission and absorption is required and it is found that the value of reflectivity of about 80% gives quite sharp fringes with a good contrast (absorption 3.5%).

The following are the various conditions for the production of highly sharpened multiple-beam intereference fringes having high resolution.

1) The surface must be coated with highly reflecting films having minimum absorption.

2) The film must contour the surface exactly and be highly uniform in thickness.

3) Monochromatic light or at the most a few widely spaced monochromatic wave-lengths should be used.

4) The interfering surfaces must be seperated by a few wavelengths of light.

5) A parallel beam (within 1^{o}-3^{o} tolerance) should be used.

Two beam interference technique lacks the precision usually found in the multiple reflections of the multiple-beam interference technique.

For the present interferometric study of etched and the indented cleavage surfaces, a metallic Jig was prepared. The jig consists of two circular plates (either of brass or aluminium), having a circular hole in the center. The two plates are separated by three springs placed at the corners of an equilateral triangle, and can be brought close to one another by

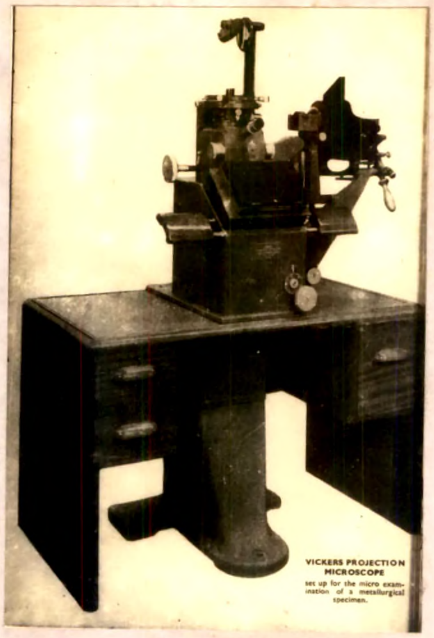
VICKERS PROJECTION
MICROSCOPE
set up for the micro examination of a metallurgical specimen.

Fig. 2.1

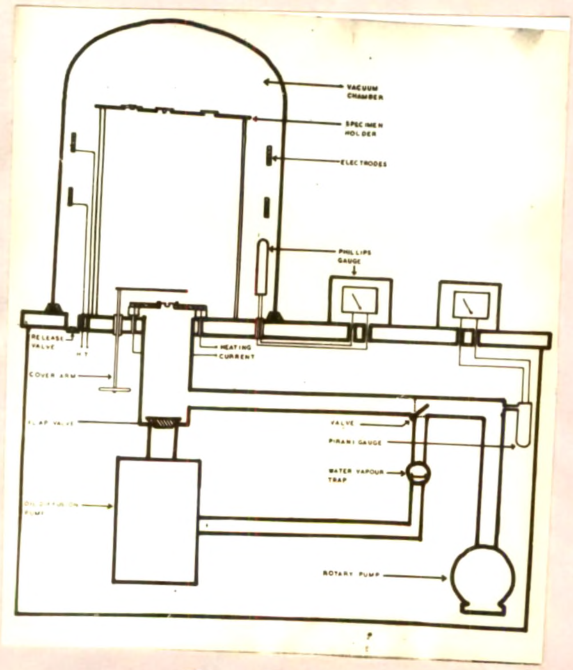
VACUUM
CHAMBER
SPECIMEN
HOLDER
ELECTRODES
PHILLIPS
GAUGE
RELEASE
VALVE
H.T.
HEATING
CURRENT
COVER ARM
VALVE
PIRANI GAUGE
WATER VAPOUR
TRAP
ROTARY PUMP

Fig 2.2

adjusting the three nuts provided, which press against the three springs. The minimum distance between the two reflecting surfaces is obtained by adjusting all the screws and should be of the order of a few wavelengths of light. Both the flat and the crystal surface were silvered, having the appropriate film thickness (a few hundred angstroms) by the silvering technique described below.

2.5 <u>Silvering Technique:</u>

For multiple beam interferometric and better microtopographical studies at higher magnification and resolution, the enhancement of the reflectivity of a surface is found to be necessary. This is done by coating a thin uniform film of silver on the surface by evaporating it under very low pressure (10^{-5} cm of Hg). For evaporating silver films, a commercial evaporating unit of the type 12EA manufactured by Edwards and Co., London was used (fig. 2.2). Here the samples which are to be coated are kept on a stage, in the vaccum chamber, which is in the form of a large pyrex bell jar, 35.6 cm in height and 30.5 cm in diameter. This is connected through the tubing to an evacuating system consisting of a three-stage silicon oil diffusion pump, backed by a rotatory pump which produces the rough vacuum of the order of 0.2 mm of Hg. The diffusion pump as such, can be used only after rough vacuum is created in the system. The backing (rough) vacuum and the final vacuum can be read directly by the Pirani gauge and Philips ionization gauge incorporated in the unit.

Silver is kept in a molybdenum filament (boat). When the pressure in the chamber falls to below 10^{-5} cm of Hg by diffusion pump, mean free path of silver is more than few diameters of chamber. When this pressure (10^{-5} cm of Hg) is reached, the boat is heated by a current of about 50 amps; the silver melts and starts evaporating. The filament is covered with an adjustable shutter in order to protect the substrate from receiving the vapour of the burnt impurities which can have a serious influence in increasing absorption. It is important to note that when the pressure falls below 10^{-5} cm of Hg, silver should be evaporated otherwise, there may be some spurious results due to small mean free path and moisture which directly affects the reflectivity and absorption of the thin film coated. It is known that water vapour and hydrocarbons present in the chamber seriously affect the quality of the film. Hence it is desirable that they should be removed from the chamber. Usually a petri dish containing fresh phosphorous pentoxide is kept in the chamber to absorb moisture, at the time of evaporation. The thickness of the silver film and hence the reflectivity is judged either by viewing filament from the top of the bell jar, or by calibrating the filament in terms of current and time of evaporation of silver.

The uniformity of the silver deposited is of paramount importance. The amount of condensation, in perfect vacuum from a point source, at any point on a plane surface is inversely proportional to the square of the distance of the point from the source and to the cosine of the angle between the normal to the plane and the line joining the point in question to the source. The distance between the source and the condensing plate= is about 20 cms in the present case. It is found under the above mentioned conditions that the silver is deposited uniformly over the whole surface. It has been observed that the most stringent interference tests fail to reveal any serious variations in average silver thickness over such a surface, while no local variations can be detected at magnifications of about 400. To obtain high reflectivities of about 90%, the thickness of the film should be nearly 500 A^{o}. Oil and impurities on the surface to be silvered reduce the reflectivity coefficient somewhat, so a condition of cleanliness of surface is of vital importance. The cleaning agents depend upon the type of surface to be silvered.

2.6 Etch Method:

The crystal surfaces were etched in etchants in the usual manner, i.e. by dipping the crystal in an etchant of desired concentration for definite time period at a constant temperature and then washing it in running distilled water. It is known that rinsing may deform the crystal if there is an appreciable change of temperature occurred at the time of rinsing. Hence adequate care in the present work was taken during the rinsing of the crystal.

The etching was carried out in two different ways, depending upon the temperature of etching viz. etching (i) at room temperature and (ii) at high temperatures.

(i) Etching at room temperature:-

Since the rate of stirring of etching solution noticeably affects the etch pattern, the room temperature etching was carried out under the following conditions: (1) Static (no stirring) (2) Dynamic (stirring at regular speed). In static etching, the motion of etchant relative to the crystal being etched is absent, while in dynamic etching the stirring of the etchant at a controlled speed by a rotor is made with respect to the crystal. Alternatively a crystal can be fixed at the end of a rotor, which can be rotated in a steady etchant under controlled conditions. It is advantageous to keep motion of etchant uniform, so that turbulant flow of etchant is avoided.

(ii) Etching at high temperatures:

Chemical etching can be done at high temperatures to get quick reaction between etchant and a crystal surface. High temperature etching alters the normal rate of etching at room temperature. In this method etchant is therefore kept in an incubator or a muffle furnace (depending upon the etching temperature) at the time of etching.

Thermal etching usually occurs at high temperatures (near the melting point or decomposition temperature of the crystal). The crystal is kept in a crucible in the muffle furnace at desired temperature for thermal etching. Details of muffle furnace and incubator will be given in the relevent chapters. Similarly dehydration of the crystal surface is also one type of etching which usually takes place at high temperature. In the present work dehydration of cleaved surfaces of gypsum crystals is studied at temperatures (round about 100°C). The observations are reported in the concerned chapters.

2.7 Determination of Surface Traces on a Crystal Surface:

A number of natural phenomena are capable of producing visible traces of important crystallographic planes on exterior crystal surfaces. Because these traces are intimately associated with the crystal structure, they may be very useful in several significant ways. Thus for example, traces may serve as a basis for determining crystal orientation. On the other hand they may also give significant information about the plastic deformation or phase transformation phenomena that created the traces. Slip is an important source of crystallographically related surface traces. In its simplest form, a crystal is sheared on a plane of low Miller indices, known as the slip plane. As a result, steps, called slip lines are produced on the crystal surface. Such a slip line is, therefore, the intersection of a major crystallographic plane with the crystal surface. Surface traces of twins are also of importance. These may be either due to deformation twins, annealing twins or growth twins.

The determination of planes, producing traces on the surface becomes easier when the surface under observation is known. Such is indeed the case in the present microtopographical studies of cleavage surfaces of single crystals of calcite. Under this condition, it is easier to use stereographic method. Excellent accounts of the method are available in several books. (see e.g., Tutton, 1922; Barret, 1953 and Bunshaw, 1968). Briefly, it consists of first drawing, in a standard stereographic projection of the crystal, great circles corresponding to the surface of measurement. Then it is required to determine, on each of these surfaces, the angle between the traces and a given reference direction on the crystal surface. A set of points corresponding to the observed traces, are next plotted on the stereographic projection at the proper angles along great circle of each surface. Additional great circles are then drawn through corresponding pairs of these points, i.e. through one point on each circle, representing a crystal face.

These define, the planes associated with the observed traces. The poles of these latter traces should then be plotted in the standard projection with the aim of identifying their miller indices. Then it is possible to determine the directions of observed surface traces.

In the present work, microtopographical studies of etch and indentation features on cleavage faces of calcite are reported. During this study several surface traces are observed. Since the crystal face on which the traces are observed are known it became easier to determine the directions of these surface traces and corresponding planes by following the stereographic method described above. Instead of drawing a stereogram it is also possible to determine traces by measuring angles of this trace with some standard reference line such as slip line or edge of a crystal. From the known morphological data of natural crystal of calcite it is possible to indentify these traces.