ment. Several possibilities of hydroxyl bonding arise in this structure. O III has two near oxygen neighbours other than those attached to the same Si atom: O V at a distance of 2·5 Å and O I[1] at 2·7 Å. The bond O III–O V, being the shorter, is perhaps more likely to be the hydroxyl bond, although the difference is hardly significant at this stage. O V has three close oxygen neighbours other than O III. From a consideration of the valency angles of O V, a hydroxyl bond between O II[8] and O V can be excluded. Hydroxyl bonding of O V to either O I or O IV[6] is, however, equally probable.

The structural formula of dicalcium silicate α-hydrate is $Ca_2(SiO_3OH)OH$.

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Aluminium Monochromator with Double Curvature for High-Intensity X-ray Powder Photographs.

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The construction and performance of an X-ray monochromator using an aluminium single crystal with double curvature is described. The point-focus obtained in this way is used for taking powder photographs of the Guinier type. The gain in intensity is about 16 times as compared with a monochromator using a quartz crystal with single curvature and polished surface, all other conditions being equal.

1. Introduction

As far as we know, point-focusing monochromators using crystals with double curvature have not yet been used in practical X-ray work, although the principle and advantage of such monochromators have been recognized for many years. Shenfil, Danielson & DuMond (1951) have constructed a point-focusing monochromator using two cylindrically curved quartz crystals with their axes at right angles (the geometry of this design was described by DuMond (1950)). This monochromator was intended for the study of low-angle scattering, but its intensity is rather low for several reasons (among others, the losses at the two reflexions).

The bending of a crystal plate to a surface with double curvature can probably be effected only by means of plastic deformation, which greatly reduces the number of possible substances. It may be quite possible to use sodium chloride, but we have chosen aluminium for our trials. We hoped to take advantage of the high intensities reported by Cauchois, Tiedema & Burgers (1950) for spectra from aluminium single crystals. For the case of transmission these authors found the intensities of spectra from aluminium single crystal lamellae to be 5–10 times as great as from quartz lamellae. For reflexion we have found that about 4 times as long an exposure is required with a quartz monochromator (plane $10\bar{1}1$ and with polished surface*) as with an aluminium monochromator (planes 100, second order, or 111) in order to obtain the same integrated intensity (estimated visually) in powder photographs, all other essential conditions being equal.

* It is known that the integrated intensity of reflexions from quartz is lower if the surface has been polished than if it has only been ground.

2. Principle

Let Fig. 1 first illustrate the case of a cylindrically

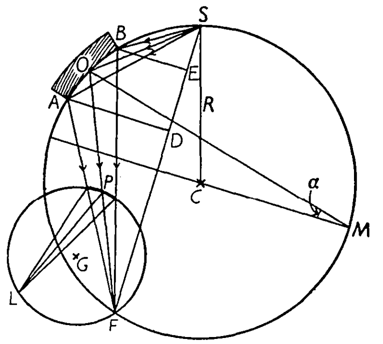

Fig. 1. The plane of the focusing circle. C, centre of the focusing circle of radius R. S, X-ray source. O, centre of monochromator crystal. F, focus. G, centre of Guinier camera with sample P and powder line at L. For the arrangement actually used (Cu $K\alpha$ radiation and the reflexion 200 from aluminium) R = 170 mm, α = 5°. AD = 48·5 mm., BE = 41·5 mm, corresponding to a length of the monochromator arc AOB = 40·0 mm. The resulting maximum aperture ASB = 6·7° is never used in practice but is reduced by slits to about 4·5°.

ground and bent monochromator in an asymmetric position. M is then the centre of curvature of the cylindrically curved, reflecting lattice planes. The angle OMC $(=\alpha)$ determines the distances $SO = 2R \sin(\theta-\alpha)$ and $OF = 2R\sin(\theta+\alpha)$.

If the reflecting lattice planes of the monochromator crystal form a surface with double curvature generated by the focusing circle rotating around the chord SF, the focus F will be a point focus.

It can easily be shown that, in order to obtain maximum intensity in this case, the crystal must be brought as close to the source as possible. In the plane of Fig. 1 and for constant R the crystal always subtends the same angle at S. At right angles to this plane, however, the subtended angle increases as the distance SO decreases, i.e. as the angle α increases. The intensity factor due to absorption in the monochromator crystal in this case has the form $\sin(\theta+\alpha)/[\sin(\theta+\alpha)+\sin(\theta-\alpha)]$ and, consequently, also increases with α. We have, therefore, used an asymmetric position with a value for α which brings the crystal as close to the exit slit system of the X-ray tube as is convenient for the adjustments. This also has the advantage of allowing good space between crystal and focus for the camera with its specimen holder and slit systems.

The following approximate calculation gives a rough value of the increase in intensity when a monochromator with double curvature is substituted for a cylindrical monochromator. The source S is assumed to be a point and all conditions except the form of the monochromator are assumed to be unchanged.

If the height (i.e. the dimension in the axial direction) of the illuminated area of the sample P is h, the effective height of the cylindrical crystal at O is $h.SO/(SO+OP)$.

For the case of the crystal with double curvature it is assumed that the focus F is a point. Then the effective height of the crystal in this case is $h.OF/PF$. The ratio of the effective heights of the crystals for the two cases can be assumed to be equal to the intensity ratio. One then obtains a value for the gain factor of $OF(SO+OP)/PF.SO$. In our case the distances are $OF = 156$, $SO = 101$, $OP = 86$, and $PF = 70$ mm. The approximate gain factor would then be 4·1.

It was stated in § 1 that an aluminium monochromator gives about 4 times as intense reflexions as a quartz monochromator of the same form and with polished surface. As a consequence, an increase in intensity of about 16 times might be expected, if, under the above-mentioned conditions, a point-focusing aluminium monochromator is substituted for a cylindrically curved quartz monochromator. As will be seen from § 4, this figure has actually been attained under experimental conditions.

3. Preparation of the monochromator crystals

The preparation of the aluminium single crystals was carried out in essentially the same way as described by Tiedema (1949) and by Cauchois, Tiedema & Burgers (1950).

The starting material was cold-rolled aluminium sheet of 1 mm. thickness and of 99·5% purity. It is essential that the reduction in thickness by cold-rolling is high, in the range 85–95%. Strips 20 mm. wide and about 300 mm. long were annealed by passing them through a tube furnace at 550° C. at a rate of 6–10 cm./min. The strip was then stretched with an elongation of 2–2·5%. The next stage was recrystallization of one end of the strip at 630° C., passage through the furnace being at a rate of 3 cm./hr. This procedure aims at forming a single crystal of some centimetres length within the first 5 cm. of the strip. This single crystal must border on the remaining polycrystalline portion of the strip. After removal of oxide with 3% hydrofluoric acid, the size of the crystals can be seen after etching with a dilute mixture of nitric and hydrochloric acid.

The orientation of the single crystal was determined by means of a Laue photograph. The other end of the strip (containing polycrystalline material) was then bent into a cylinder of radius $2R$ by means of a mould of heat-resistant steel made in two cylindrically shaped halves. This bending must be effected very carefully. We steadied the end of the strip by surrounding it with two lead plates and fed the resulting sandwich in between the two halves of the mould. After thus obtaining the approximate radius the lead plates were removed and the halves of the mould were pressed together. The whole strip, together with the mould, was then placed in a sort of goniometer in which the

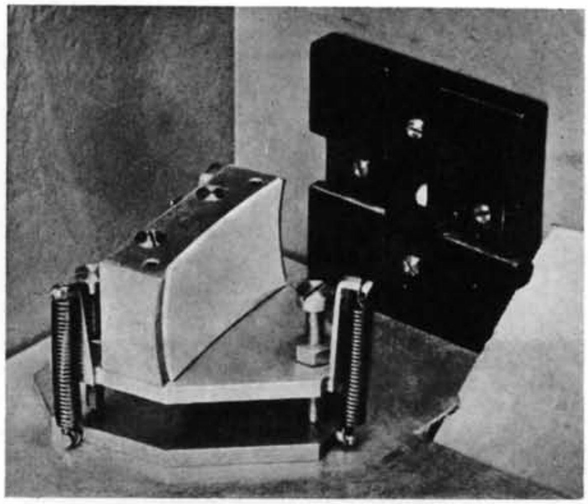

Fig. 2. Aluminium single crystal with double curvature in holder.

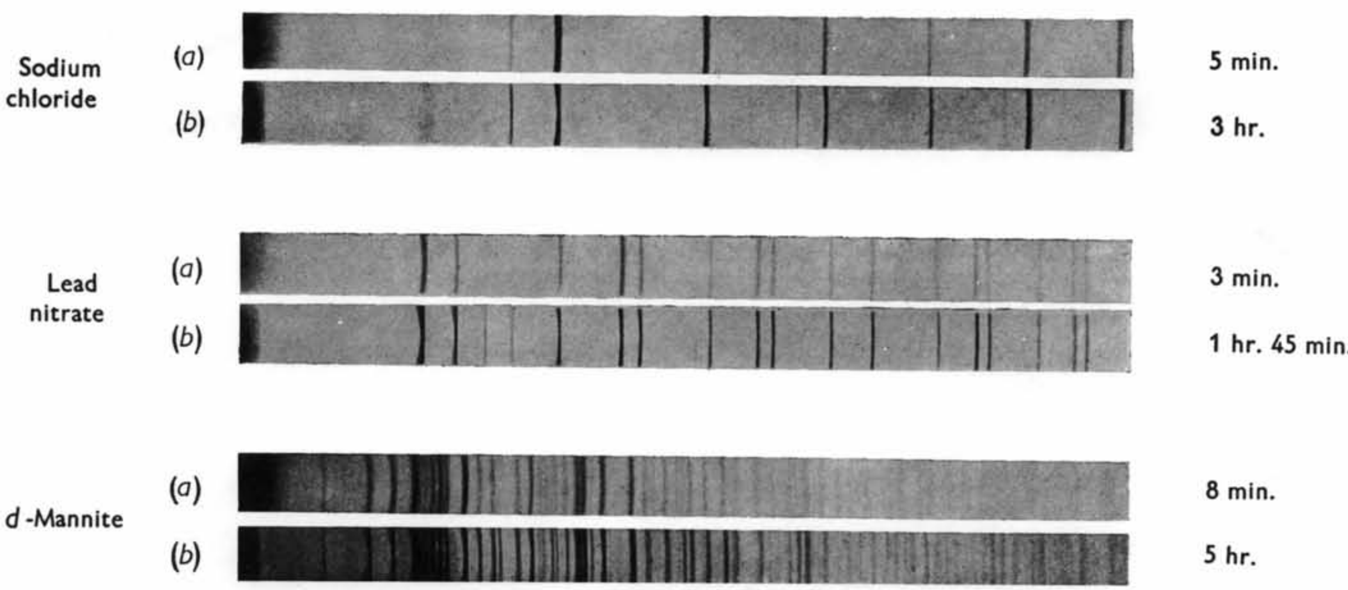

Fig. 3. Comparison of powder photographs (Guinier method, Cu $K\alpha$ radiation, 45 kV., 25 mA.) from the same sample with (a) aluminium monochromator with double curvature and the geometrical data given in Fig. 1, (b) cylindrical quartz monochromator (reflecting plane $10\bar{1}1$, $R=250$ mm., $\alpha=3°$). The exposure time for series (a) is about $\frac{1}{35}$ of that for series (b), which gives approximately equal integrated intensities for a given sample. The illuminated area of the sample is 2·2 times as large in series (a) as in series (b), which corresponds to an intensity ratio of about 16 if all essential conditions other than the monochromators are identical.

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mould could be turned around two perpendicular axes while the other end of the strip, containing the single crystal, was fixed. The mould was then turned (resulting in bending and twisting of the middle part of the strip) until the cylindrical part of the strip came into the desired position with reference to the lattice of the single crystal. Either 100 or 111 can be used as the reflecting plane, but we have found that with a strip of the above mentioned length the bend must not exceed about 15°. A twist of 45° is permissible and, in view of the cubic symmetry, this does not imply any limitation. If the monochromator is to be used in an asymmetric position it is necessary at this stage to take into account the angle α (see Fig. 1).

The strip was then disengaged from the mould and the goniometer and passed, with the end containing the single crystal leading, through the furnace at 630° C. and at a rate of 2 cm./hr. The single crystal then grew into the polycrystalline part of the strip and finally into the cylindrically curved end. After that, the cylindrical part was sawn off and bent in a new mould with a radius of curvature equal to R. Annealing (in the mould) at 630° C. for 24 hr. and removal of the oxide film completed the preparation of the cylindrical monochromator crystal.

The double curvature was achieved by pressing the cylindrical crystal in a mould with the final shape. The convex half of this mould (forming part of the convex surface of revolution generated by the arc AOB in Fig. 1 rotating around the axis SF) was made of brass in the lathe. The main axis of the lathe corresponded to SF and the tool was mounted on a circular feed device with its centre in a position corresponding to C. The concave half of the mould was made by casting type metal on the convex half.

After the formation of the double curvature the aluminium crystal was annealed at 630° C. for 24 hr. and after removal of oxide was cemented to the concave half of the mould. This was used as a support and mounted in a holder with facilities for adjustment (Fig. 2).

4. Performance

As already stated in § 1, a cylindrical aluminium monochromator gives about four times the intensity of a cylindrical quartz monochromator with polished surface, all other essential conditions being equal. The focusing properties, however, were not quite as good as for the quartz monochromator. As a result the lines of powder photographs were somewhat broader with the aluminium monochromator than with the quartz monochromator.

It is rather natural that the sharpness of powder lines obtained with the monochromator with double curvature should be less satisfactory. This is due to inferior focusing in the equatorial plane (the plane of Fig. 1). The width of the focus in this plane is about 0·5 mm. (its height in the axial direction is about 2·5 mm.), while the corresponding width of the focal line from the cylindrical quartz monochromator is about 0·25 mm. For a great many purposes, however, where a very short exposure time is essential, these powder photographs ought to be quite useful.

Fig. 3 enables a comparison to be made between the performances of the new monochromator and a cylindrical quartz monochromator. After reduction to as far as possible identical conditions, the intensity ratio is about 16, i.e. the figure that was anticipated in § 2.

Efforts to improve the focusing properties of the new monochromator type are continuing along various lines. It must be mentioned that the final result has been found to vary considerably even if the same procedure of preparation is followed. It may well be, therefore, that better monochromators than the best we have made could be obtained if a larger number could be tested.

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