

NEW METHODS FOR DETECTING CERTAIN CRYSTALLINE DEFECTS IN QUARTZ

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ABSTRACT

Two additional new methods were found for the detection of those crystalline defects in quartz, arising probably from submicroscopic inclusions, which have been detected by the writer's method of etching quartz with hydrofluoric acid under an alternating electric field. The first method consists of subjecting a quartz plate under investigation to the action of an electric discharge at low pressure, and then immersing it in hydrofluoric acid. The second method is simply to etch a plate by exposing it to the vapors of alkaline metals at about 350° C. In each case etching patterns were observed on most of the Z-section plates with the form of a system of polygons or zonal patterns similar in appearance, but not in their elementary constituent figures, to those produced by the method of etching under an electric field.

In the first method, four essential facts have been found: (1) the effect of an electric discharge as manifested by the persistency of the etching patterns is restricted to a certain depth of the superficial layer of the exposed surface, (2) this effect can be removed by heat treatment, (3) no etching patterns were produced on X-section plates, (4) dew patterns similar to etching patterns can be formed by condensing water vapor on the unexposed surface of a plate that has been newly subjected to the action of an electric discharge. Tentative explanations for the first three facts have been proposed and the reason for the fourth one requires further investigation.

INTRODUCTION

It was shown by the writer (1) that the application of a high alternating electric field normal to a quartz plate during etching with hydrofluoric acid results in the formation of two kinds of elementary etch figures. Figures of one kind differ from those of the other both in size and in structure. While the figures of microscopic dimensions are exactly the same as are those ordinarily found in etching under a non-electric field, those of visible size are produced only when there is an active electric field. (The former will subsequently be called the ordinary figures, and the latter the electric figures.) It was also shown that the electric figures (2), produced on plates cut from the so-called high grade crystals, are generally not distributed at random but are more concentrated in some regions than in others, forming a system of concentric polygons together with some special zones of irregular boundaries (in some cases only the zonal pattern is present). Furthermore, there is no such preferential distribution of the etch figures in the case of etching without an electric field. As previously remarked, the appearance of the polygonal

* In previous publications the family name was placed first instead of last in accordance with the Chinese custom.

and zonal patterns indicates the existence of two kinds of crystalline defects arising probably from submicroscopic inclusions. Subsequent work (3) on comparing the etching patterns of plates cut from the same blocks of quartz, proved conclusively that these defects are spatially distributed in a crystal. As a matter of fact, the appearance of the concentric polygonal patterns strongly reminds one of the annual rings of wood and it is fairly certain that this pattern indicates the contours of the crystal during early stages of its growth.

Regarding the polygonal etching pattern, mention should be made of the investigation of C. Frondel (4) on the coloring effect by α -rays on quartz. He showed that smoky bands having directions parallel to the rhombohedral faces of the mother crystal are sometimes developed in quartz plates by the application of α -rays. According to Van Dyke and Clyde P. Glover (5), the polygonal etching patterns produced by the writer's method and the colored patterns of Frondel are both formed in the same region of a given plate.

In view of the frequent occurrence of crystalline defects and also of their possible connection with the well-known variation of certain physical constants of quartz as previously indicated (2), it seemed worth while to see under what other conditions these defects could be detected. As a result of such an undertaking, two additional new methods were found; the one employing the combined action of an electric discharge and hydrofluoric acid, and the other to the corrosive action of the vapors of alkaline metals.

EXPERIMENTAL

The Z-section and X-section plates of a thickness varying from one to two millimeters were used throughout the work. The quartz plates used were either polished or smoothed with M303 emery as the case might require. As the techniques employed in the study of the effects on the crystalline defects of quartz by the electric discharge method and the action of the vapors of alkaline metals are different, they are described in the following under separate headings:

Method 1, By Means of the Electric Discharge

The discharge apparatus used resembled the ordinary sputtering type, of which the essential parts are diagrammatically shown in Fig. 1. A bell-jar about 20 cm. in diameter and 30 cm. in height was used as the discharge chamber. It rested on a thick iron platform, *D*, with its well-ground mouth sealed thereto with apiezon-sealing compound to make it air tight. The top opening of the jar was fitted with a brass stopper,

through which an aluminum rod, *A*, was passed, the joints being made air-tight also with apiezon-sealing compound. A circular aluminium plate, *P*, used as the upper electrode of the discharge system, was attached to the lower end of *A*. A flanged aluminum disk, *F*, attached to a massive brass cylinder served as the lower electrode. The quartz plate, *Q*, to be investigated was placed on the disk. For a reason to be pointed out later, *Q* was not to rest directly on the disk, but separated by about 0.2 mm. by means of three small supporting slats, *R*. The

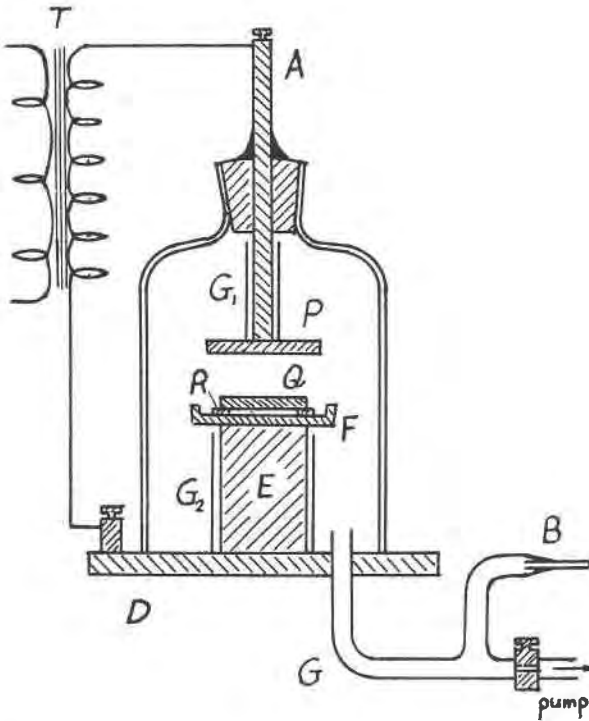


FIG. 1. Discharge apparatus. *P*, upper electrode; *F*, lower electrode; *Q*, quartz plate; *R*, supporting slats of *Q*; *E*, massive brass cylinder; *D*, iron platform; *B*, palladium tube; *T*, transformer.

massive lower electrode possessed large heat capacity which prevented the temperature of the plate, *Q*, from rising dangerously high during discharge. To prevent stray discharge, the brass cylinder, *E*, and the part of the rod, *A*, inside the jar were sheathed with glass tubings *G*₂ and *G*₁, respectively. In order to render the electric field over the quartz plate as uniform as possible, the distance between the two electrodes

was constantly kept smaller than either of the diameters of the two electrodes. Through the glass tubing *G*, the discharge chamber was connected to an evacuating system (not shown), which consisted of an oil pump and a mercury diffusion pump. The pressure of discharge was measured by means of a McLeod gauge. A palladium tube, *B*, attached to *G*, permitted the introduction of pure hydrogen into the discharge chamber in case of necessity. High tension used in the discharge was supplied by a 10,000-volt transformer. The alternating voltage across the electrodes during discharge was about 700 volts and the current passed through was about 3 milliamperes. The duration of discharge varied from half an hour to a score of hours, as required by the tests.

When the discharge was over, the quartz plate, *Q*, was taken out of the jar and ready for any one of the following investigations:

- (1) To study the effect of discharge on the etching of quartz by plunging the plate into hydrofluoric acid for a specified time.
- (2) To see how the temperature would influence the effect of discharge by heating the plate to about 300° C. in an electric furnace for about half an hour before plunging it into hydrofluoric acid.
- (3) To study the effect of discharge on the condensation of water vapor on the plate surface.

In performing the second investigation, the quartz plate was cut into two pieces; one of them being then introduced into a furnace and the other kept at room temperature. When the heating operation was over, the two pieces were simultaneously plunged into hydrofluoric acid. Then a comparison was made of their etched surfaces. As for the third investigation, the water film might be conveniently produced by condensing the vapor in the moist air from the mouth.

Method 2, By Means of the Vapors of Alkaline Metals

The vapors of sodium, potassium, and rubidium were used as the etching agents for quartz. A small capsule of an alkaline metal, together with one or several pieces of quartz plates to be investigated, were introduced into a large Pyrex tubing one end of which had been sealed. The tubing was then connected to an evacuating system. After evacuation and sealing the tubing was placed in a cylindrical electric oven for heating. The alkaline metals begin to vaporize appreciably at 150° C., as could be judged from the coloration of the tubing. But in order to obtain good etched plates it was found necessary to raise the temperature to about 350° C. and to maintain it there for three or four hours. When the etching process was over, the plates were taken out and rinsed with fresh water to remove a layer of colored coating formed on them during etching.

RESULTS

Part 1, Effect of an Electric Discharge

Quartz plates having been subjected to the action of an electric discharge for several or even a score of hours, undergo no apparent changes other than the addition of an almost invisible metallic film on their exposed surfaces. (By exposed surface is meant the surface facing the upper electrode, *P*.) However, when they are immersed in hydrofluoric acid, the etching patterns take the form of a system of concentric polygons and special zones with irregular boundaries are developed on the exposed surfaces of most of the *Z*-section plates but never on their unexposed surfaces. These patterns are identical in form with, but different in their elementary constituent figures from those produced by etching under an a.c. electric field. In fact, the former are composed of the ordinary pyramidal figures which are the usual characteristic etch figures of the *Z*-section, while the latter are composed of star-shaped electric figures. Moreover, in contrast to the figures obtained by electric discharge, the ordinary figures in the polygonal and zonal patterns do not appear to be concentrated in certain regions. Microscopic examination has revealed that in these patterns, the average size of the ordinary figures as well as their number per unit area, are somewhat smaller than those in the other part of the same etched surface. This fact indicates that electric discharge brings about a difference in rate of dissolution of quartz between regions containing submicroscopic inclusions and those free from them. Since the difference both in size and in number of the etch figures in the different regions is small, the visibility of the patterns by electric discharge is generally low, in fact too low to be reproduced photographically. It is to be pointed out that under similar experimental conditions, no etching patterns of a similar nature can be produced on *X*-section plates.

In regard to the pattern-producing effects of electric discharge, there are several related phenomena which deserve mentioning. First, this effect as manifested by the persistency of the polygonal and zonal etching patterns was found to reach in some cases a depth of about 0.03 mm. In other words, the etching patterns continue to exist until the pattern-bearing superficial layer was dissolved by the corrosive acid. The depth of the affected layer does not depend appreciably upon the duration of discharge from some thirty minutes upwards, nor upon the kind of gas (air or hydrogen) used in the discharge. It may be of interest to remark that the pattern depth of 0.03 mm. is about three times the average height of the pyramidal etch figures of which the patterns are composed. The above mentioned fact tends to show that the depth of

the affected layer represents the penetrating range of some sort of moving particles. Second, the visibility of the etching patterns was found to depend markedly upon both the pressure of the discharging gas and the orientation of the quartz plate during discharge. Other things being equal, the optimum pressure for obtaining patterns of high visibility lies between the interval of 0.2 to 0.5 mm. of mercury. For pressures higher than 0.8 mm. or lower than 0.01 mm. the pattern-producing effect vanishes. For unknown reasons it was found necessary to separate the plate and the lower electrode by a distance of some 0.2 mm. Third, the pattern-producing effect acquired by a plate during discharge is removable by heat treatment. This is conveniently shown by heating a plate having been subjected to discharge, in an electric furnace before etching. Such a plate loses the said effect slowly at 150° C. and very quickly at 300° C. or higher. Also, the method of electric discharge furnishes another rather peculiar way for detecting the defects of quartz. When water vapor is made to condense on the unexposed polished surface of a plate freshly stimulated by discharge, polygonal and zonal patterns will be formed on the dew film in case this plate does possess the corresponding defects. The appearance of the dew patterns is similar to the etch patterns which are produced, as pointed out in the foregoing paragraph, on the opposite face, i.e., the exposed surface, when the plate is treated with hydrofluoric acid. Owing to the rapid evaporation of such a dew film, it has not been possible to examine microscopically the size and the manner of distribution of the droplets in the different parts of the film. Patterns of this kind can be reproduced several times after each evaporation; but if the surface is touched with a finger, or a moistened cloth, the peculiar phenomenon disappears. No such patterns have ever been observed on the exposed surface of the same plate. For the observation of dew patterns, a polished surface was found to be indispensable.

Part 2, Effect of Alkaline Metals

The vapors of alkaline metals were found to etch quartz quite perceptibly at about 150° C. The etching becomes more and more vigorous as the temperature rises. However, owing to the formation of a superficial layer of non-volatile material which prevents rather effectively the contact between the surfaces and the metallic vapors, the etching process soon slows down. A well etched plate may be obtained by raising the temperature to about 350° C. and at the same time prolonging the etching duration to several hours. The increase in etching by extending the duration beyond one or two hours appears to be practically negligible.

Figure 2 is a photomicrograph of the etch figures produced by po-

tassium vapor on a Z-section plate. As may be seen the etched face is granulated. Among the granular figures also some curved grooves may be noted. The etching produced by the different metallic vapors are similar. These granular etch figures bear no resemblance to the pyramidal ones produced with hydrofluoric acid.

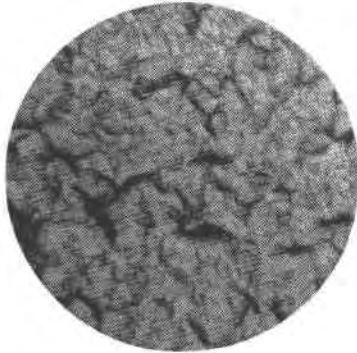


Fig. 2. Etch figures produced by potassium vapor. Magnification 100X.

The main object of this part of the work was to see whether it was possible to detect the defects of quartz by using the etching power of vapors of alkaline metals. The result is affirmative. The appearance of the polygonal and zonal etching patterns caused by the vapors of these metals is similar to those produced by other methods mentioned above. The visibility of these etching patterns is generally low. Considering the three metals used in the experiments, namely sodium, potassium and rubidium, the third is not as effective as the other two in that the colored layer on the plate is insoluble not only in water but also in other cleaning solvents.

DISCUSSION AND CONCLUSION

As is conceivable, when a quartz plate is exposed to electric discharge, it is actually subjected to three types of actions, namely, the irradiation of ultraviolet and visible radiations, the action of an electric field, and the bombardment of speedy particles. Evidently the cause of the pattern-producing effect of an electric discharge must be sought for among these actions. In view of the fact that the form of the ordinary etching figures and the rate of dissolution of quartz in hydrofluoric acid can both be greatly modified by the action of ultraviolet radiations (6), attempts have therefore been carried out to reproduce the etching patterns similar to those by an electric discharge by irradiating the Z-section plates with an intense iron-spark, either before or during etching. However, the result was negative. The fact that an alternating electric field can pro-

duce polygonal and zonal patterns when, and only when, the field is applied to quartz during etching with hydrofluoric acid, discards the possible role of the second action. Hence the responsibility for the specific effect falls exclusively on the bombarding action of speedy particles. Since a speedy particle may penetrate a substance to a certain depth before being stopped, the existence of a superficial layer of the order of 0.03 mm. in thickness and with the polygonal and zonal etching patterns continuing to exist therein, seems to offer a strong support to the third hypothesis. However, a quantitative consideration of the problem will show that the bombardment hypothesis is not as readily acceptable as it may appear to be at first sight. Statements in the subsequent paragraphs will make this point clear.

Since the speedy particles must derive their kinetic energy from the discharging circuit we may first assume these particles to be ionic in nature and then test the validity of the bombardment hypothesis. This can be done by estimating the kinetic energy necessary for an ionic particle to penetrate to a depth of 0.03 mm. into the quartz on the one hand, computing the maximum kinetic energy attainable by it in the discharged circuit on the other hand, and then comparing the magnitude of these two quantities. It is possible to attain this end by resorting to a rather roundabout way. Let us estimate the range of an alpha particle in quartz. The stopping power, S , of a molecule of a given substance is known and can be expressed as

$$S = \frac{R_0 d_0}{R d} \frac{M}{A_0} \quad (1)$$

where R_0 and R represent the ranges of an alpha particle in a standard gas and in the given substance, respectively, d_0 and d , their densities, A_0 the atomic weight of the gas and M the molecular weight of the substance. According to Bragg and Kleeman, the stopping power of an atom is proportional to the square root of its atomic weight and the relative molecular stopping power may be obtained by adding together the stopping powers of the constituent atoms. Hence the molecular stopping power of quartz can be written as

$$S = [(28)^{1/2} + 2(16)^{1/2}]k = 13.3k \quad (2)$$

where k is a constant of proportionality. The value of k is 0.27 as deduced from the known ranges of the alpha particles of RaF both in air (standard gas) and in oxygen by means of eq. (1) and the law of Bragg and Kleeman. Hence the value of S in eq. (2) is equal to 3.59. By taking air at normal conditions as the standard gas and quartz as the given substance and substituting the proper physical constants in eq. (1), the relation

between the range of alpha particles in quartz, R , and that in air, R_0 , becomes:

$$R = 0.0005R_0. \tag{3}$$

The variation of the kinetic energy of a charged particle along an element dx of the trajectory in quartz may be expressed by a formula according to Bethe (7), which is (for a constant mass of the particle):

$$\frac{dW}{dx} = - \frac{4\pi Z^2 e^4 N n}{mv^2} \log_e \frac{2mv^2}{F} \tag{4}$$

in which W represents the kinetic energy of the moving particle: Ze , its charge; v , its velocity; N , the number of electrons per molecule of quartz; n , the number of molecules per cm^3 ; m , the mass of an electron; and F , the geometrical mean of the energies of excitation and ionization of the molecule. Upon substituting W by its equivalent $\frac{1}{2}Mv^2$ where M represents the mass of the moving particle, and integrating the resultant expression, the length of the trajectory, i.e., the range, R of the particle is found to be

$$R = \frac{Mm}{4\pi Z^2 e^4 N n} \int_0^v \frac{v^3}{\log_e \frac{2mv^2}{F}} dv \tag{5}$$

For brevity, let

$$\phi = \int_0^v \frac{v^3}{\log_e \frac{2mv^2}{F}} dv,$$

then

$$\phi = \frac{4\pi Z^2 e^4 N n}{Mm} R. \tag{6}$$

It is to be noted that for a given stopping medium, ϕ is independent of the kind of moving particle but is a function of its velocity alone. Since the value of F for a quartz molecule is not known, the ϕ cannot be evaluated directly from the integral; but it can be deduced from eq. (6) if the various quantities on the right-hand side of this equation are known. Now the ranges in air, R_0 of alpha particles are known for their various kinetic energies, E , so their ranges in quartz, R , and hence the corresponding values of ϕ can be calculated by eq. (3) and (6), respectively. Some of such data on the alpha particles are tabulated in Table 1. Figure 3 is obtained by plotting E taken from the second column of Table 1 as abscissae and ϕ of the fourth column as ordinates. As may be seen from the curve, the E - ϕ relationship is approximately a linear one, and the curve on being extended, passes through the origin. Later

TABLE 1. DATA ON ALPHA PARTICLES. R_0 REPRESENTING THE RANGE OF ALPHA PARTICLES IN AIR; R , THE RANGE IN QUARTZ; E , THE KINETIC ENERGY OF THE PARTICLES EXPRESSED IN ELECTRON-KILOVOLTS; AND ϕ , THE VALUE OF THE CORRESPONDING VELOCITY FUNCTION

| $*R_0$ (cm) | $*E$ (in electron kilo-volts) | $R \times 10^{-4}$ (cm) | $\phi \times 10^{-35}$ |
|-------------|-------------------------------|-------------------------|------------------------|
| 0.55 | 1167 | 2.9 | 1.09 |
| 0.62 | 1328 | 3.3 | 1.23 |
| 0.70 | 1499 | 3.8 | 1.39 |
| 0.80 | 1681 | 4.3 | 1.59 |
| 0.91 | 1873 | 4.9 | 1.80 |
| 1.04 | 2075 | 5.6 | 2.07 |

* Data cited from Madame P. Curie, (Radioactivity) Radioactivité, Hermann & C^{ie}, Paris, 1935, p. 530.

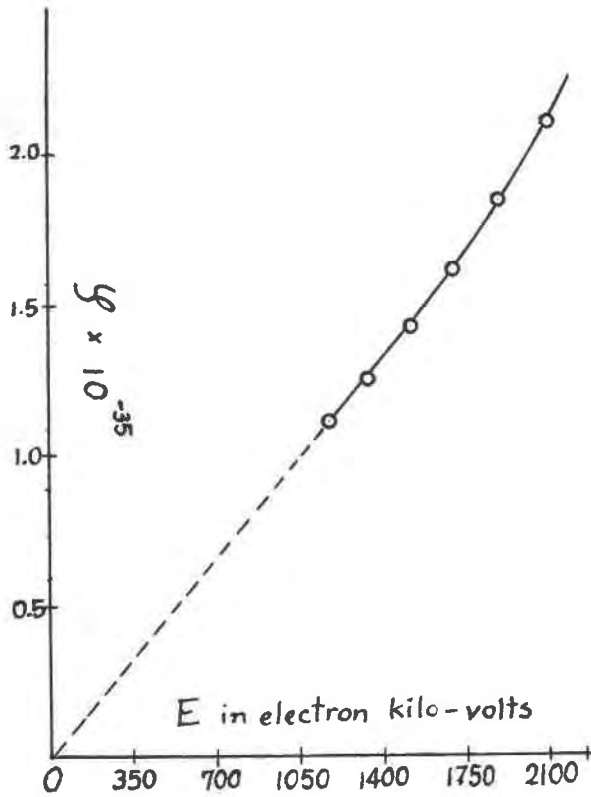


FIG. 3. E representing the kinetic energy of alpha particle, ϕ the corresponding velocity function.

on use will be made of the accompanying figure for estimating the kinetic energies of ions other than the alpha particles.

As is conceivable, molecular ions as well as free electrons will coexist in an electric discharge. These ionic particles will be accelerated by the discharged electric field and may thus attain high speeds. Moreover, as may be inferred from existing spectroscopic data, practically all the charged particles present under the specific discharged conditions are singly ionized. For this reason, Z may always be set to unity in the calculation of ϕ from eq. (6). Consider first, the case of singly ionized molecules of oxygen which should be the heaviest ions in the air under discharge. If the depth of the layer in which the polygonal and zonal etching patterns persists is taken as the penetrating range of a diatomic oxygen ion, the value of ϕ , as calculated from eq. (6) is 0.33×10^{35} . The kinetic energy, E of an alpha particle corresponding to this value of ϕ , is found by extrapolating the curve of Fig. 3 to be about 350 electron-kilovolts. Since charged particles of different kinds, possess the same speed for a given value of ϕ , the ratio of the kinetic energy of one kind of particle to that of the other is the same as the ratio of their masses, i.e.,

$$\frac{E_1}{E_2} = \frac{M_1}{M_2} \quad (7)$$

where E_1 and M_1 represent the kinetic energy and mass, respectively, of one kind of particle and E_2 and M_2 the corresponding quantities of the other kind. Hence the kinetic energy of a singly ionized oxygen molecule having a range of 0.03 mm. in quartz is found to be 2800 electron-kilovolts. On the other hand, if a free electron is taken as the particle which has a penetrating range of 0.03 mm. in quartz, the corresponding value of ϕ will be 1.92×10^{39} . By supposing the linear relationship in Fig. 3 to be still valid for a value of ϕ as large as 1.92×10^{39} and making use of eq. (7), a rough estimate of the kinetic energy of the penetrating electron is possible. The result so obtained is about 2400 electron-kilovolts. For ions with masses intermediate between the mass of an electron and that of a diatomic oxygen molecule, the kinetic energies will be lying within the interval between 2400 and 2800 electron-kilovolts.

The maximum kinetic energy acquirable by a charged particle in an electric discharge may be estimated from the conditions of discharge. Take for example the case of a discharge in air under a pressure of 0.2 mm. Hg, which is the lower limit of the optimum pressure of discharge for obtaining etching patterns of high visibility. At this pressure, the mean free path of an oxygen molecule is about 0.035 cm. and that of an electron, 0.18 cm. The cathode fall of potential for air is known to be

around 340 volts and the anode fall of potential some 20 volts. Hence the maximum kinetic energy of a positive oxygen molecule (O_2^+) will be of the order of 66 electron-volts and that of an electron much less. For any other kind of ions, the mean free path will be somewhat greater or smaller (for example N_2^+) than that of a diatomic oxygen molecule and so will be the kinetic energy. But in every case, the maximum kinetic energy acquirable is more than thirty thousand times smaller than that demanded by the range-velocity relationship. This discrepancy appears to be a serious blow to the bombardment hypothesis under discussion. However, the difficulty is reconcilable by postulating that the effect of discharge on the formation of etching patterns is due to the action of speedy neutral molecules rather than to that of ions; for if Z of eq. (5) which represents the number of the electronic charge of the moving particle, is set at zero, the range R becomes infinite, theoretically. It is very probable that for a neutral particle to be able to penetrate into a solid medium, a minimum kinetic energy is necessary. The energy thus required can be drawn from a speedy ion by the process of collision. Moreover, a speedy ion may also be transformed directly into an energetic neutral one by neutralization.

The statement of the foregoing paragraph indicates that gaseous molecules are liable to be embedded in the superficial layer of a Z -section plate through the process of an electric discharge. On the other hand, it was suggested by the writer that the regions in which the polygonal and zonal etching patterns are found, are the sites of submicroscopic inclusions. According to the quantum mechanical theory of solids the electron energy levels are grouped into quasi-continuous bands separated by gaps of the order of a few electron-volts in width. When foreign molecules are inserted into the lattice they may give rise to localized energy levels within the gaps between energy bands of the base materials. Hence it is reasonable to postulate that for a quartz plate having been subjected to the action of an electric discharge, the localized energy levels created within the gaps of bands will be different between the regions containing submicroscopic inclusions and those free from them. This eventual difference in energy levels might give rise to unequal rates of dissolution of quartz by hydrofluoric acid and thus render the specific regions discernible. That the pattern-producing effect of discharge can be removed by heat treatment is also comprehensible; since vigorous thermal agitation may help the embedded gaseous particles to diffuse away.

The fact that an electric discharge has no pattern-producing effect on X -section plates, might have something to do with the piezoelectric property of quartz. Indeed it seems sound to suppose that for a X -section plate, free charges are liable to be created on the spot of bom-

bardment of a speedy particle and that part of these charges may attach to the bombarding particle, thus rendering the penetrating power of the particle negligibly small.

The feasibility of obtaining dew patterns, similar to etching, by condensing water vapor on the unexposed surface of a newly stimulated Z-section plate is something rather peculiar. In view of the fact that this peculiar effect of an electric discharge is easily removable by touching the surface with a moistened cloth or a finger, it must be in some way connected with electricity. For a better understanding of this phenomenon, further investigation is undoubtedly necessary.

Prior to the present work, the crystalline defects of quartz have been detected most readily by the electric method of etching and less readily by the x -ray method. To these, two more methods may now be added, namely, the method of an electric discharge and that of etching by means of the vapors of alkaline metals. Of the four methods, the first appears to be most sensitive.

In conclusion, the writer wishes to acknowledge the helpful assistance of Mr. C. Y. Ling in drawing the diagrams, and of Mr. K. Y. Chou of the Zoology department for the photomicrograph shown in this paper.

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