EFFECT OF RADIATION ON THE ELASTICITY OF QUARTZ

CLIFFORD FRONDEL,*
Research Division, Reeves Sound Laboratories, New York, N. Y.

CONTENTS

ABSTRACT

When quartz is exposed to *x*-rays, gamma-rays, electron beams, alpha-particles or deuterons it becomes smoky in color, the rate of solution in hydrofluoric acid is decreased, and the elastic constants are altered. The effect of radiation on the elastic properties is revealed by study of irradiated quartz oscillator-plates. In general the oscillation frequency decreases continuously during irradiation to a limiting value, and types of plates dependent on different elastic constants are affected unequally. The effect is being utilized in manufacture to make the final frequency adjustment of oscillator-plates.

The degree of response in color and elasticity is limited and varies among different specimens. Baking at temperatures over about 180°C. restores the original properties, and conditions the response of the quartz on re-irradiation. Luminescence phenomena accompany both irradiation and baking and, together with the response in color and elasticity, are related to the original color and composition of the quartz.

INTRODUCTION

It has been known for many years that quartz becomes smoky in color when exposed to *x*-rays, radium radiations, or cathode-rays (electrons.)[1] Other radiations, both of the wave-type and streams of material particles,

* Department of Mineralogy, Harvard University, Cambridge, Mass. (on leave).

[1] Villard, P., *Compt. rend.*, **126,** 1564 (1898). Georgiewski, N., *Ann. Physik, Beibl.*, **28,** 1015 (1904). Salomonsen, C. J., and Dreyer, G., *Compt. rend.*, **139,** 533 (1904); **138,** 1543 (1904). Egoroff, N., *Compt. rend.*, **140,** 1027 (1905). Bertholet, D., *Compt. rend.*, **143,** 477 (1906); **145,** 818 (1907). Philips, C. E. S., *Nature*, **77,** 535 (1908). Goldschmidt, V. M., *Forh. Vidensk. Seleskr., Oslo*, No. 5, 1 (1906). Brauns, R., *Cbl. Min.*, 721 (**1909**). Doelter, C., *Das Radium und die Farben*, Dresden (**1910**). Newbery, E., and Lupton, H., *Mem. Proc. Manchester Lit. Phil. Soc.*, **62,** 1 (1918). Doelter, C., and Sirk, H., *Sitzber. Ak. Wiss. Wien, Math.-nat. Kl.*, **119,** 1091 (1919). Doelter, C., *Cbl. Min.*, 321 (**1923**). Holden, E. F., *Am.*

also are effective. A photograph of the smoky color produced by a narrow beam of x-rays in a sawn block of quartz is shown in Fig. 1. Recently the writer has found that the change in color is accompanied by variation in the elastic constants of the quartz as well. This is shown by a change in the frequency of oscillation of quartz plates that have been exposed to and colored by radiation.

Characteristics of Irradiated Oscillator-Plates

When a BT-cut quartz oscillator-plate is placed in a powerful beam of x-rays the frequency of oscillation is found to decrease. The frequency change is rapid at first but then slows down and finally approaches a

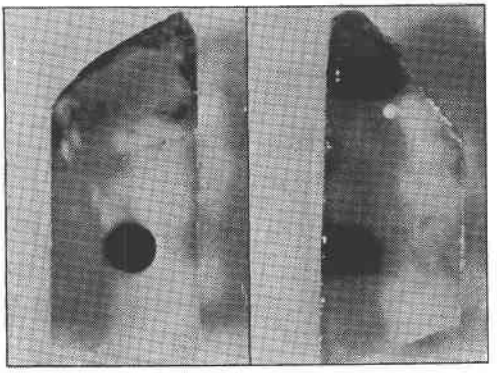


Fig. 1. Two views of a small block of quartz showing the smoky color produced by a narrow beam of x-rays.

point beyond which there is no further change on continued irradiation. The type of curve obtained by plotting frequency change against exposure time is shown in Fig. 2. The total change of frequency that can be effected is variable and depends on a number of factors. Among these are the type of cut of the plate, the treatment given to the plate prior to irradiation, the kind of radiation employed, and the initial frequency, or thickness, of the plate itself. There also is a considerable variation in response among different specimens of raw quartz and hence between different plates of the same frequency cut therefrom. The observed variation in saturation value in 8000 Kc BT plates is roughly

Mineral., **9,** 1 (1924); **10,** 203 (1925). Hoffman, J., *Zeits. Anorg. Chem.*, **196,** 225 (1931); **219,** 197 (1934). Bruce, E. L., *Trans. Royal Soc. Canada*, **28,** Sec. 4, 7 (1934). Twyman, F., and Breck, F., *Nature*, **134,** 180 (1934). Mohler, N., *Am. Mineral.*, **21,** 258 (1936). Shin-Piaw Choong, *Jour. Phys. Radium*, **2,** 41 (1941). The present paper was privately issued under War Department restrictions in Nov., 1943, and July, 1944.

from 500 to 10,000 cycles decrease with an average change of approximately 1400 cycles decrease. Plates that change more than 2000 cycles are rare.

Only part of the total area of a BT crystal has to be irradiated in order to gain the maximum frequency shift. The critical area comprises roughly 30 to 50 per cent of the total area if it is circular and centrally located. Similarly, only a relatively small part of the central portion of a crystal has to be plated with metal in order to gain maximum frequency decrease by loading (Fig. 3). These observations tie in with the well known fact that the corners of shear mode rectangular plates are relatively inactive during oscillation.

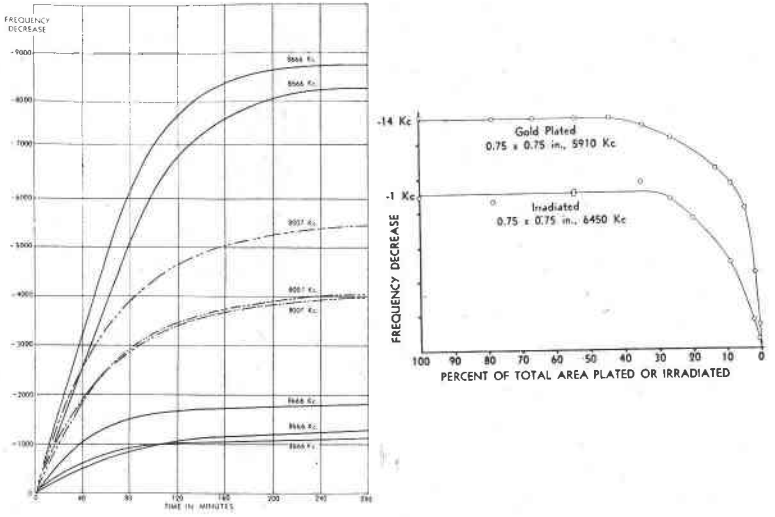


FIG. 2. Curves illustrating the variation in saturation value and rate among two groups of plates. Irradiated at 45 KV, 25 ma, 4 mm window to plate distance, unfiltered Cu radiation.

FIG. 3. Relation between frequency decrease and the area irradiated or gold plated.

The average saturation value is a function of the initial frequency or thickness of the plate and increases with increasing frequency. Preliminary measurements indicate that the average change in BT plates of a particular frequency can be calculated on the assumption that the frequency-thickness constant of irradiated quartz ($K = 100 \times 10^6$ cycles per second per 0.001 inch) is less by about 0.02 per cent than in ordinary quartz. The activity of the plates is usually slightly decreased by irradiation, except when there are material changes in coupling; and the piezoelectric constant is not grossly altered.[2]

[2] Seidl, F., *Ak. Wiss. Wien, Ber.*, **142**, 2a, 467 (1933).

Different types of oscillator-plates respond differently to radiation. Both BT plates oscillating in high frequency shear and X-cut plates oscillating on the high frequency thickness (or X) mode are relatively highly responsive. Y-cut crystals oscillated on the high frequency shear mode are not responsive. AT shear mode plates are less responsive than BT plates. Different elastic constants or combinations of elastic constants are utilized in these types of cuts, and it appears that the constants are affected unequally by irradiation. This might mean that a particular bond in the interlocking SiO_4 tetrahedra composing quartz is more sensitive to radiation than others. Relative variation in the elastic constants leads to the possibility that plates containing closely coupled modes of vibration could be adjusted by irradiation to eliminate activity dips or achieve particular temperature coefficients of frequency. This is ordinarily done by control of dimensional ratios. Other more important practical applications of the irradiation technique are described in an accompanying paper.[3]

A rough idea of the magnitude of the frequency change to be obtained from different pieces of quartz can be gained from the luminescence phenomena described in a later section and from the original color of the quartz. Generally speaking, colorless quartz shows a wide variation from specimen to specimen in the degree of response in color to irradiation. Very low temperature hydrothermal quartz seems to be relatively weakly affected. Deep smoky quartz and citrine uniformly show a relatively small response. Amethyst quartz is entirely unaffected in color. If amethyst is first decolorized by baking, irradiation restores the amethystine color. Rose quartz, which contains traces of titanium in solid solution together with exsolved rutile needles, develops an extremely intense, almost black, smoky color. Chalcedony is weakly affected by *x*-rays and alternate bands in the mineral may become unequally colored. Opal is not affected; tridymite is deeply colored. The depth of smoky color produced in an oscillator-plate by *x*-ray appears to be directly proportional to the frequency change therein. Spectrographic analysis of oscillator-plates made variously from colorless and natural smoky quartz reveals no correlation between the kind or amount of foreign elements present in the quartz and the degree of response in color and frequency on irradiation. The principal foreign elements found in 16 samples of Brazilian quartz examined spectrographically, present in amounts over 0.001 per cent, were Al, Li, B, Na, K, Cu and Mn, listed in order of decreasing magnitude.

Rate of Change of Frequency. The principal factors influencing the rate

[3] Frondel, C., Final Frequency Adjustment of Quartz Oscillator-Plates, *Am. Mineral.*, this issue.

of change of frequency during oscillation are the intensity of the x-ray beam, the distance of the plate from the window and anode of the x-ray tube, the thickness of the plate itself, the choice of wave-length, and the magnitude of the total frequency change that can be effected in the plate in question.

The rate of change of frequency is found to be directly proportional to the intensity of the x-ray beam. The beam intensity itself increases as the square of the voltage and directly as the current passed. Data that illustrate these relations are given in Table 1. A 8007 Kc BT plate was

TABLE 1

	KV:	12.5	25	50	50	50	50
	MA:	25	25	25	6.25	12.5	25
Frequency Decrease in 5 minutes, in cycles		30	120	450	140	270	450
Ratio		1	4	15	1	1.9	3.2

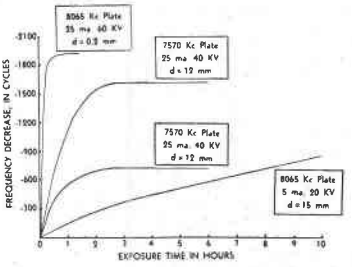
irradiated for 5 minutes at each of the several beam intensities and the frequency change measured. The departure from the theoretical ratios is due to the non-linear relation between irradiation-time and frequency change. Using unfiltered copper radiation from a broad focus copper tube operated at 25 ma and 60 KV, with a crystal to window distance of approximately 1 mm., ordinary unsensitized BT 8 mc plates can be changed in frequency on the average about 40 cycles a minute, calculated on the basis of the initial essentially linear part of the frequency-time curves. Further data are given in Fig. 4.

The distance of the plate from the window and anode of the x-ray tube is an important factor, due to operation of the inverse square law. Broadly speaking, a given frequency change produced in a few minutes when the plate is 0.5 mm. from the window will require an exposure time of hours when the plate is 20 mm. distant and an exposure of many days at a distance of a foot. Measurements that further illustrate this effect are given in Fig. 5. These curves were all obtained on the same plate which was irradiated successively on the various distances cited, with the x-ray intensity kept constant.

It has already been pointed out that the average saturation value increases with increasing frequency of the plate. The average rate at which saturation is reached is also found to increase with increasing plate frequency (or decreasing plate thickness) at constant intensity of

the x-ray beam. This effect is accentuated by the relatively strongly absorbed but weakly penetrating long wave-length components of the incident beam, which, while they penetrate to the same depth in a thin as in a thick plate, expose a larger percentage of the total mass of quartz as the plate thickness decreases.

The rate of change also varies with variation in the total change in frequency, or saturation value, that can be effected in a group of plates of the same nominal frequency, the experimental conditions being held



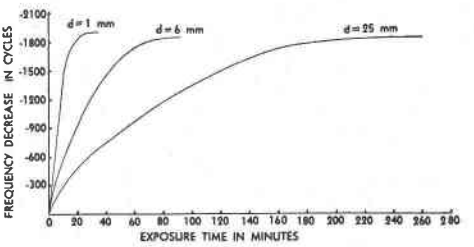


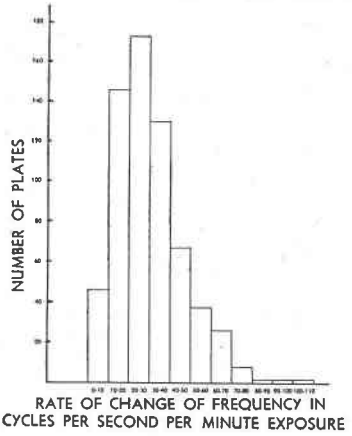
FIG. 4. Curves illustrating the variation in rate of change of frequency with x-ray beam intensity.

FIG. 5. Effect of varying window to plate distance on rate of change of frequency. 8416 Kc plate irradiated at 60 KV and 25 ma, Cu radiation.

constant (Fig. 2). In a group of plates of the same nominal frequency the total frequency change that can be effected varies more than twenty-fold. Generally speaking, the larger the total frequency change the longer it takes to reach saturation, but no rigorous relationship appears to exist between these factors. Production data illustrating the variations in rate observed in a run of 638 BT plates of 8007 Kc frequency that had been irradiated only part of the way to saturation are shown in Fig. 6. The plates had been overshot a few hundred cycles or so during manufacture and were irradiated enough to bring them back down below the upper frequency tolerance. The frequency changes are shown on the graph as rates. These rates are the slopes of the linear portion below the knee of the frequency-time curves.

Effect of Baking. Irradiated quartz decolorizes and reverts back to the original frequency when heated to a sufficiently high temperature. The change is a time-temperature reaction and is similar in this to the decolorization of natural smoky quartz. In the neighborhood of 180° CO a true reversal of frequency begins which is extremely slow and requires a period of weeks for completion. The rate of reversal increases rapidly with increasing temperature and over 450° the change is almost instan-

taneous. The reversal in frequency is accompanied both by a discharge of the smoky color and by a pale blue thermoluminescence. The increase in frequency brought about by baking is found to be exactly the same as the initial decrease brought about by irradiation. The maximum temperature at which the color of natural smoky quartz is stable is about 225° C, and the rate of decolorization, as with artificially colored crystals, increases rapidly with increasing temperature. The decolorization in this case is not accompanied by any change in frequency.



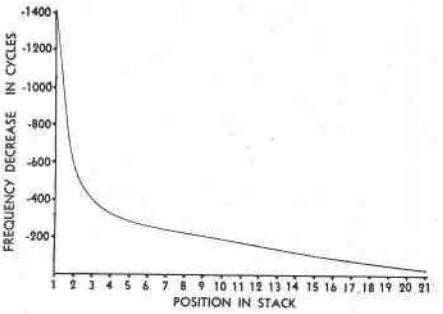


FIG. 6. Observed variation in rate of change of frequency during irradiation of 638 plates of 8007 Kc frequency under constant experimental conditions.

FIG. 7. Frequency decrease in unit time through a stack of twenty-one 8416 Kc plates irradiated at 15 ma and 45 KV with heterogeneous Cu radiation. A 8416 Kc plate is 0.01188 inches thick.

Baking and decolorizing an irradiated plate increases the rate of response of the quartz on re-irradiation. The sensitization is most marked at the limiting decolorization temperature of *ca.* 180° C. and becomes progressively less at higher temperatures until the rate equals and then becomes less than that of fresh quartz. The sensitization may involve the formation of F′ centers, as in the photolysis of the alkali halides.

Rate of Solution of Irradiated Quartz. If a quartz plate is irradiated through a lead shield pierced with a small hole, so that there is a sharply defined smoky area produced in the quartz, and the plate is then deeply etched, it is found that the irradiated area is less affected and appears on the surface of the plate as a slight eminence. In other words the rate of solution of irradiated quartz is less than in ordinary quartz. It is also found under certain not easily reproducible etching conditions that sawn and lapped sections of ordinary quartz sometimes become differentially etched along parallel bands or zones. Crystals with alternately banded

smoky and colorless portions may show marked effects of this kind. The behavior reflects a local variation in the properties of the quartz itself. Oscillator-plates apparently change frequency slightly less rapidly if they have an abrasive-lapped instead of an etched surface. This effect may be due to the existence of a rough skin of quartz in the former case which does not participate in the oscillation and which has a filtering and shielding action on the incident radiation.

Absorption of X-Rays. Stacked Plates. Quartz is relatively transparent to x-rays and a large part of the incident radiation is transmitted through a thin oscillator-plate and does not contribute to frequency changes therein. The efficiency of utilization of the x-ray beam can be increased by appropriately relating the wave-length of the incident radiation to the particular thickness of the quartz plate being treated, or, alternatively, by stacking a number of plates one behind the other and irradiating all of them simultaneously. Since the mass absorption coefficient varies with wave-length, frequency changes measured in successive crystals down through a stack vary markedly from an exponential law due to the filtering action of the preceding plates in removing the softer, easily absorbed and more strongly ionizing components of the incident heterogeneous radiation. Fig. 7 shows the frequency changes effected in unit time in twenty-one 8416 Kc plates stacked together. Three identified plates were measured in each position and the results averaged. Very similar curves are obtained by measuring the absorption of x-rays with a Victoreen roentgen-meter in varying thicknesses of metal or quartz. Oscillator-plates can be used successfully as dosimeters or roentgen-meters in measuring or comparing the intensity and quality of x-ray beams and in measuring the ionizing equivalents of x-rays to other kinds of radiation.

Luminescence Phenomena

Natural quartz when heated in the dark often exhibits a pale bluish-white thermoluminescence (phosphorescence). The luminescence begins at a rather low temperature and after increasing in intensity with increasing temperature to about 300° to 400° C. gradually dies out. The intensity varies widely in different specimens of quartz, including specimens from the same locality, and sometimes is found to be confined to particular growth bands or parts of the crystal. About one half of 232 Brazilian crystals tested showed a noticeable glow on heating. Further observations have been given by Goldschmidt[4] and by Köhler and Leitmeier.[5] The statement of Ichikawa[6] that triboluminescence, a related

[4] Goldschmidt, V. M., *Forh. Vidensk. Selskr., Oslo*, No. 5, 1–19 (1906).

[5] Köhler, A., and Leitmeier, H., *Zeit. Krist.*, **87**, 146 (1934).

[6] Ichikawa, S., *Amer. J. Sci.*, **39**, 455 (1915).

phenomena, occurs in ordinary quartz but not the smoky varieties was not verified; the smoky color only obscures the bright internal reflection of the surface sparks seen in colorless quartz. The effect is relatively weak or absent in smoky quartz. Banded smoky sections of quartz, however, always are more affected by x-rays in the colorless regions whether these parts are thermoluminescent or not. No difference is found in the frequency-thickness constant of natural thermoluminescent and non-thermoluminescent quartz. A complex sequence of luminescence effects is obtained when natural banded smoky and colorless quartz is irradiated and heated.

Irradiated quartz is brightly thermoluminescent when heated. The glow is confined to the irradiated area. The luminescence begins at a lower temperature than in natural quartz, below 100° C., and the intensity is considerably greater. The glow is not accompanied by a change in frequency or in smoky color at temperatures below the 180° C. stability limit previously mentioned. The margins of the irradiated area at first glow more intensely than the interior parts. In an irradiated plate the thermoluminescence may persist after the quartz has been heated to a temperature at which the frequency and smoky color are discharged.

Quartz exhibits a bluish-white fluorescence during irradiation with x-rays. The samples also phosphoresce briefly. The intensity of the fluorescence varies considerably in different specimens. A relatively intense glow is met with in material that is not thermoluminescent and that has an original smoky color, but a number of exceptions were noted. It is usually found that the more intense the fluorescence, the more intense is the total response in frequency and in smoky color produced by the radiation, but several marked exceptions were observed. The intensity of the fluorescence apparently varies during irradiation. If a fluorescent plate is irradiated to saturation (in frequency and color) over a small area, and the plate is then moved laterally so that the incident x-rays overlap a fresh area of quartz the fluorescence in that area is much more pronounced. It may be observed in this experiment that the margin of the initially saturated area fluoresces relatively brightly. An interesting thing about the effect of x-rays on quartz is the lack of consistency in the response of different specimens. The correlation of luminescence phenomena, saturation value in frequency, rate of change of frequency and baking effects with each other, and with characters such as original color, more or less tend to follow general rules but always with marked exceptions. Part of the difficulty is in controlling the wide variety of experimental conditions.

Origin of the Luminescence and the Frequency Change. A possible origin

for the change in elasticity is suggested by the luminescence phenomena. Radiations of relatively low energy content, such as ultraviolet light, when impinging on a crystal may effect a momentary transfer of electrons from lower to higher levels surrounding an atom. The infall of these electrons to their original level is accompanied by a release of energy and this appears as light in the long ultraviolet or visible parts of the spectrum (fluorescence). The displaced electrons also may be frozen in the metastable positions to be later released by thermal agitation of the structure, or otherwise, with emission of light (phosphorescence). With radiations of shorter wave-length and greater energy content as in the x-ray region of the spectrum, electrons may be completed ejected from an atom—an internal photoemission. The change may be permanent under ordinary conditions if the ejected electron is caught elsewhere in the structure, such as by an oppositely charged atom, a vacant lattice position or other type of electron-trap. "Holes" of positive potential to trap photoelectrons may exist in quartz through a Si-deficient defect structure; and atoms of a trivalent metal such as Al in substitution for Si similarly may act as electron receptors. In any case, a transfer of electric charge within the structure would result in a change of the interatomic bonding forces, and this in turn would be reflected by changes in the elastic properties and the piezoelectric oscillations dependent thereon. The limitation in the total change of frequency that can be effected presumably reflects the number of electron-traps available in the crystal. Baking the crystal restores the original properties by supplying energy to lift the trapped, metastable, electrons over their potential barrier and returning them to the atoms, now with a positive potential, from which they were originally ejected. The energy released in the infall appears as thermoluminescence. The color of natural smoky quartz appears to be a separate problem, since it is discharged at higher temperatures and is not then accompanied by a frequency change.

Pigmentation of crystals resulting from the ejection of electrons is well known, as in the blackening of silver bromide and in the production of color-centers in alkali halides and other substances.[7] A wide variety of changes in chemical systems dependent on electron transfer are similarly brought about by ultraviolet light.[8] The variation in elastic constants which is presumed to accompany these changes, however, ordinarily would tend to go unrecognized because of the minuteness of change and the insensitivity of the usual methods of measurement. Ultraviolet rays

[7] Mott, N. F., and Gurney, R. W., *Electronic Processes in Ionic Crystals*, Oxford (**1940**); Rollefson, G. K., and Burton, M., *Photochemistry*, New York (**1942**).

[8] Heyroth, F. F., *The Chemical Action of Ultraviolet Rays*, New York (**1941**).

and x-rays have been found, however, to increase both the rigidity and elastic limit of NaCl crystals.[9]

Color Distribution Phenomena

Evenly irradiated plates sometimes develop alternately light and dark smoky bands up to 1 mm or more in width. Some extreme examples are illustrated in Fig. 8. The bands are arranged parallel to the external faces of the mother crystal and are similar to the color bands commonly seen in natural smoky crystals. When natural quartz containing smoky bands

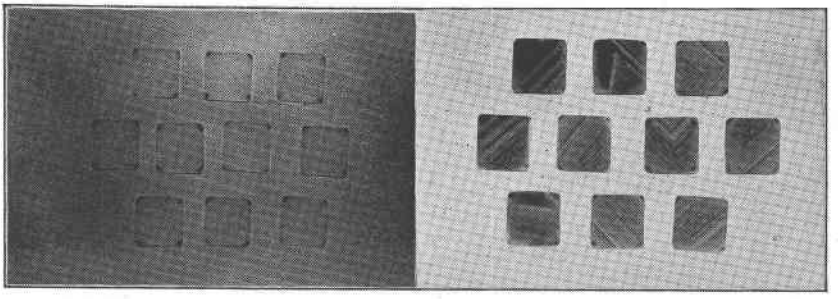


Fig. 8. Smoky bands developed in quartz oscillator-plates by x-rays. Left, before irradiation; right, after irradiation. The color bands are parallel to the rhombohedral faces.

is irradiated, the colorless bands become relatively deeply pigmented (Fig. 9). When banded natural quartz is decolorized by baking and then irradiated the original relatively light colored parts again are more deeply affected by the x-rays. Color bands also may develop in entirely colorless and water clear quartz. Instead of bands some specimens develop irregular or polygonal smoky areas. The color response apparently is conditioned by a sensitizing factor which was areally distributed within the crystal during growth.

A very remarkable type of variation in color response is found in twinned quartz. It is often observed that the two parts of a natural Dauphiné twinned quartz crystal become unequally colored when a broad beam of x-rays is allowed to fall across the twin boundary. The effect is not due to the difference in orientation in the two parts of the twin. Differential coloration is not produced in quartz that has been twinned artificially on the Dauphiné law by heat or pressure. On the other hand, irradiated differentially colored naturally twinned crystals that have been artificially detwinned by heat, and at the same time decolorized, are

[9] Podashevsky, M. N., *Phys. Zeits. Sowjetunion*, **8**, 81 (1935).

found on re-irradiation to become again differentially colored along what was the original twin boundary. Brazilian twins also are differentially colored by radiation. Occasionally natural smoky crystals of quartz are observed in which the color is limited against twin boundaries, and the feature is quite common in amethyst. The origin of the differential coloration of twinned quartz is problematical. The content of foreign elements

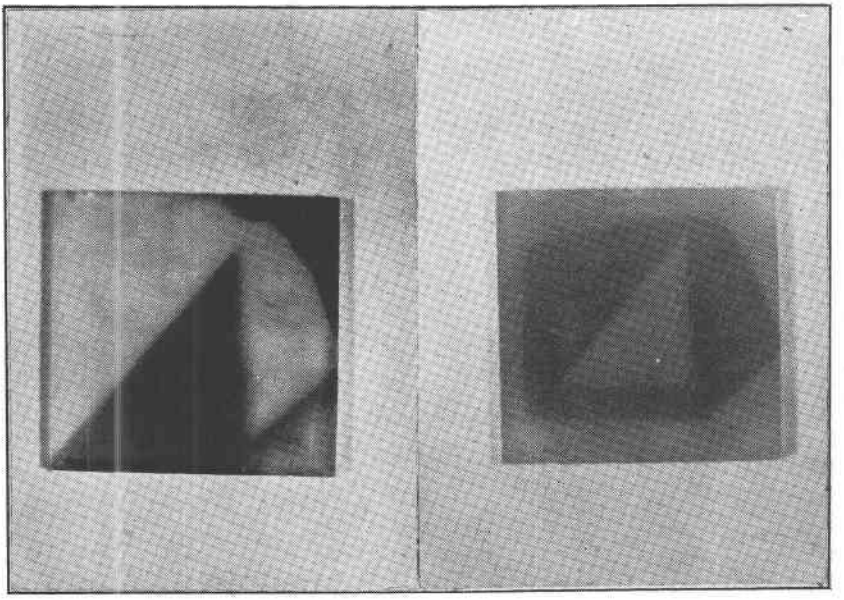


FIG. 9. Quartz plate containing natural smoky banding before (left) and after (right) irradiation. The originally colorless parts are more affected.

in a crystal, and which could possibly act as a sensitizing factor, differs between the various form loci as shown by Frondel, Newhouse and Jarrell.[10] If, now, the twinning brought unlike faces into juxtaposition, such as the effect of Dauphiné twinning in bringing $(10\bar{1}1)$ and $(01\bar{1}1)$ faces together and coplanar, opposite sides of the twin boundary could have differences in composition and hence respond unequally to irradiation. The trouble with this theory, however, is that both Dauphiné and Brazil twins restricted to $(10\bar{1}0)$ prism faces respond unequally over the twin boundaries and in this case the two parts of the twin are chemically identical. Alternatively it can be imagined that there has been a diffusion of a sensitizing factor within the crystal which was dammed up at the

[10] Frondel, C., Newhouse, W. H., and Jarrell, F. R., *Am. Mineral.*, **27**, 726 (1942).

twin boundary; an instance somewhat of this type involving the restriction of color centers to one individual of two intergrown KCl crystals is described by von Hippel.[11]

Effect of Radiations Other Than *X*-Rays

A number of different kinds of radiation have been found that cause color and frequency changes in quartz. These include both radiations of the wave type, more particularly *x*-rays and gamma rays, and streams of material particles including alpha-particles, electrons (both cathode rays and beta-radiation from radioactive decay) and deuterons. Deuterons, which are doubly charged nuclei of heavy hydrogen (deuterium) atoms, are highly effective. Neutrons and radiations in the wave spectrum of wave-length longer than *x*-rays have not been found to pigment or reduce the frequency of quartz oscillator-plates. Of the effective radiations, *x*-rays are the only practical choice for manufacturing operations although the radioactive radiations have in certain circumstances a definite application.

Frequency Adjustment with Radon. Radon, a gas formerly known as radium emanation, is one of the radioactive decay products proceeding from uranium and radium and ultimately ending in the 206 isotope of lead. The members of the series with the exception of the end product, lead, are unstable and transform one into the next at definite rates. The transformation is accompanied by the emission of radiation, either alpha-particles or beta-particles plus gamma rays. Radon itself emits only alpha-particles but the freshly prepared gas progressively disintegrates into RaA, RaB, etc., all present simultaneously in a state of transient equilibrium, and some of these contribute beta and gamma radiation. The decay of radon is exponential and can be expressed

$$N = N_o E^{-\lambda t}$$

where N is the number of relative units of radon (atoms or millicuries) which survive after a time t, N_o the number originally present at time zero and λ is a constant ($=2.097\times10^{-6}$ for radon) indicating the rate of disintegration. Roughly about 16.5 per cent of the activity of radon as measured by its gamma output after transient equilibrium is reached is lost each 24 hours. One half intensity is reached in 3.825 days and only about 0.4 per cent of the initial activity remains after 30 days. The millicurie-hour output of radiation can be calculated from the area under the curve of the decay equation and printed tabulations also are available.[12]

[11] von Hippel, A., *J. Applied Physics*, **8**, 815, Fig. 22 (1937).

[12] Cf. Stuhlman, O., *Introduction to Biophysics*, New York (**1942**), p. 79. Tables also are available from companies supplying radon commercially.

The character of the frequency change is illustrated in Figs. 10 and 11. The data were obtained from 8163 Kc crystals mounted by wire suspension so that they could be oscillated and measured while in direct contact with the gas. The volume of the container was roughly 57 cc. Different crystals of the same frequency respond unequally on irradiation with radon, just as with x-rays. The curves obtained by plotting the

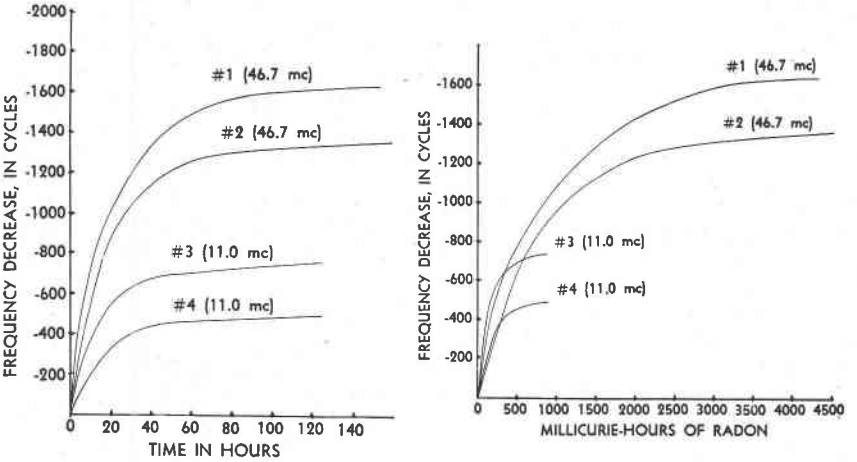


FIG. 10. Frequency change produced by radon in plated suspension mounted 8163 Kc plates. Container volume 57 cc.

FIG. 11. Data of Figure 10 recalculated on the basis of the millicurie-hours of radiation to which the plates were exposed.

frequency change against time of exposure or millicurie-hour exposure differ from the x-ray curves, typified by Fig. 2, in several respects. The radon curves fall into two parts: an initial rapid change which comprises only part of the total frequency change that can be effected by x-rays followed by a slow and relatively uniform change that apparently continues to saturation. The initial rapid change seems caused by strongly ionizing radiations of only limited penetration in quartz which rapidly saturate a certain thickness and thereafter are ineffective. These radiations would comprise the alpha and beta particles. The period of slow uniform change apparently represents ionization by the extremely penetrating but very weakly absorbed gamma radiation. It is also found that the magnitude of the initial bump in the curve increases with increasing concentration of the radon. This effect seemingly is due to an increase in the average penetrating power of the radiation. The increase may arise in the shorter average path distance of the radiation in the air of the con-

tainer as the radon concentration increases, with consequent less loss of energy by absorption and scattering before entering the quartz.

Theoretically the time rate of change of frequency should vary in proportion to the concentration of radon and the millicurie-hour rate of change in plates of a given frequency should be constant and independent of concentration. There are rather marked departures from this due to intrinsic variations in the response of different specimens of quartz and to the penetration effect noted above. The average millicurie-hour rate of change of frequency calculated from the limited data at hand is roughly 2.26 cycles per millicurie-hour in 8163 Kc plates. The rate was calculated on the basis of the initial linear part of the frequency curves.