SOME NEW RESULTS ON THE FREQUENCY CHARACTERISTICS OF QUARTZ CRYSTALS IRRADIATED BY IONIZING AND PARTICLE RADIATIONS

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ABSTRACT

Investigations on the frequency behavior of AT-cut quartz crystals irradiated by X -, γ-rays and fast neutrons have been carried out. Initial instability in frequency for γ- and neutron irradiated crystals has been found. All the different radiations first give a negative frequency shift at lower doses which are followed by positive frequency shift for increased doses. An explanation of the results in terms of the fundamental crystal structure consideration is offered. Applications of the frequency results for radiation hardening are proposed.

INTRODUCTION

Todate, radiation effects in quartz crystals is a widely studied subject. The major stimulus in the area has been largely due to the use of quartz crystals as essential electronic components for various types of communication, time and frequency standardization and other scientific and industrial uses. It is expected that with an increased understanding of the relationship of various structural properties and radiation effects, the performance characteristics of quartz material can be greatly improved according to requirements.

Due to the development of high stability quartz crystals, it is becoming possible to disseminate time and frequency signals by using quartz crystal oscillators in flying clocks, satellites etc.(1). However, for the purpose of dissemination to be achieved, it would become necessary to harden the quartz crystals against natural ionizing (X- or γ -rays) and particle (protons, neutrons, α-particles etc.)

radiations present in space. Alternatively, the effects on frequency characteristics of quartz crystals exposed to the radiation environment can be allowed for if the frequency characteristics of irradiated crystals are properly understood. All this indicates the need to study the effect of ionizing and particle radiations on the frequency and structural characteristics of quartz crystals.

A number of papers (2-19) have appeared on the radiation effects on the frequency characteristics of quartz crystals. The frequency changes due to irradiation by fast neutrons are permanent positive frequency offsets. The change in frequency due to ionizing radiations is of two kinds; transient and permanent. King and Sander (20-24) have extensively studied the transient frequency changes in quartz resonators. The transient frequency shift has been found to last for about 15 minutes in natural crystals and for a greater time (~1 hour) in synthetic crystals and is due to the temporary removal of H^+ ions from Al^{3+} -centers in the quartz lattice under the influence of irradiation and their back diffusion after the termination of irradiation. Bahadur and Parshad (25) have recently given a simple method to demonstrate the transient frequency shifts in X- or Gamma-irradiated quartz crystal resonators. The permanent frequency change in the quartz crystals has been found to be due to the drift of the alkali ions, freed from their charge compensating positions as a result of irradiation, to ion traps in the c-axis. The alkali ions can be brought back to the Al^{3+} -centers only by heating the quartz crystals (say at 200°C). The permanent frequency change in natural quartz due to irradiation with ionizing rays is negative and in the synthetic crystals it is positive. In the case of swept crystals (H^+ ions replacing the alkali ions in the quartz lattice), there is almost negligible change of frequency(8). However, for reasons being investigated perfect radiation insensitivity (radiation hardening) has not yet been achieved (26,27).

This paper presents and discusses some new results obtained on the frequency characteristics of quartz crystals irradiated by X-, γ-rays and fast neutrons. Some limited data on the subject has been published earlier (28).

EXPERIMENTAL

The quartz crystals used for the investigations were of two types; one, rectangular AT-cut plates of dimensions 3.8 x 2.8 x 0.04 cm^3 capable of vibrating in their fundamental thickness-shear mode in the frequency region of 1.87 MHz and the others AT-circular beveled centrally plated disks of frequency around 5 and 10 MHz. The precision in the frequency measurement was ± 1 Hz as the frequencies were measured digitally.

The radiations used were X-rays (30 kV, 12 ma, Cu target, white radiations), γ-rays (^{60}Co. 1000 Ci, 1.25 MeV average energy) and fast neutrons (^{241}Am-Be). Neutron irradiation was done by two different ^{241}Am-Be sources (half life 433 years). One of the sources, following the well known α-n reaction, emitted neutron flux of 2.67×10^5 neutrons/sec/cm^2. The associated γ-dose rate was feeble ($\sim$0.25 mR/sec at one meter from the source). The other ^{241}Am-Be source emitted fast neutrons with average energy of 4.5 MeV and a flux of 10^7 neutrons/sec/cm^2. The strength of this source was 5 Ci having $5 \times 3.7 \times 10^{10}$ disintegrations per second and 40% of these disintegrations were accompanied by emission of 60 keV γ-rays and 13% (of disintegrations) by emission of 100 keV γ-rays. Other γ-energies were quite low. It may be noted here that 60 keV and 100 keV γ-energies are not considerable enough to cause nuclear displacements in the lattice, the main effect being due to neutrons. The γ-rays would infact act like X-rays for their frequency effects.

Irradiation with Ionizing Radiations

The results of X- and γ-irradiation were of the same broad pattern with regard to the permanent frequency offset. However, for γ-irradiation there was observed a period of initial unstable oscillations when the irradiated crystals were first incorporated in the self-oscillating circuit. Fig. 1 depicts a typical result for a natural crystal exposed to 2000 Rads and allowed to oscillate. This instability in the initial oscillations was to a large extent independent of time of idle keeping of the crystal after the termination of irradiation and before the generation of oscillations. For X-irradiation, no such instability of oscillations was noticed, the change in the frequency offset being only steady.

The nature of steady frequency offset for both X- and γ-rays was such that for lower doses the frequency decreased and with continued irradiation, the frequency, after reaching a negative limit started increasing to a value even greater than that of the virgin crystal. Many natural crystals were investigated for this behavior. Figs. 2 and 3 depict the plot of steady frequency change versus irradiation dose for two crystals. It was observed that different crystal specimens, yielded, for the same accumulated irradiation dose, different magnitude of frequency offsets. This of-course indicates, as is widely accepted to be the case, individual variations in the lattice characteristics of the crystal by way of impurity concentration, crystal imperfections etc. Table (I) depicts the frequency offsets for a set of three different crystals which were uniformly irradiated by ^{60}Co with a dose of 48 kRads. Fundamental and third overtone steady frequencies for virgin and irradiated crystals are listed. It can be seen that the offsets in the frequencies of irradiated crystals with regard to those of virgin crystals are always positive. Table (II) depicts the results for a set of four crystals uniformly irradiated with a dose of 5 MRads. The post-irradiation frequencies mentioned are those which were measured after the crystals were allowed to oscillate and their initial instability was culminated by the mechanical oscillations themselves.

Still higher doses (~7.5 MRads) were given to different specimens and it was noticed that for prolonged irradiation, the relative change in the magnitude of frequencies were, in an apparent unexpected manner, much less than the values obtained for relatively lower doses. Also, it would be significant to mention here that the darkening in the crystals irradiated with doses of 7.5 MRads was relatively less than what was induced by 5 MRads, as observed visually. This phenomenon may be attributed to what is called 'radiation bleaching'. Fig.4 depicts the initial frequency instability character for a crystal of 1.87 MHz after it was irradiated with a dose of 7.5 MRads and made to oscillate. The pre-irradiation frequency for this specimen was 1869293 Hz and after 7.5 MRads irradiation the frequency upturned to 1.870031 Hz (a positive offset of 738 Hz). Two other such crystals were irradiated with the same dose. Fig.5 depicts the initial frequency instability character for one of the crystals. The pre-irradiation virgin frequency for this crystal was 1870190 Hz and the post-irradiation frequency was 1870397 Hz (a positive offset of 207 Hz). For the other crystal the pre-irradiation frequencies for fundamental and third overtone modes were 1870160 Hz and 5605502 Hz respectively. Upon irradiation, these frequencies changed to 1870450 Hz and 5605963 Hz for the two modes. The total offset for the fundamental mode was thus +290 Hz while for the third overtone it was +461 Hz. A crystal of 5 HMz (AT-cut) drawn from Bharat Electronics Ltd., Bangalore, India was also investigated for its frequency behavior for heavy irradiation. Fig.6 depicts the frequency instability character in the initial period of oscillations after irradiation.

TABLE (I)

Pre- and post-irradiation steady frequencies of three different quartz crystals oscillated in fundamental and third overtone modes. Each crystal was irradiated uniformly by dose of 48 kRads.

Crystal No.	Pre-irradiation Fundamental Frequencies (Hz)	Pre-irradiation Third Overtone Frequencies (Hz)	Post-irradiation Fundamental Frequencies (Hz)	Post-irradiation Third Overtone Frequencies (Hz)
1	1869451	5605051	1870558	5605421
2	1869381	5604527	1870521	5604927
3	1870206	5602694	1871059	5603927

TABLE (II)

Pre- and post-irradiation steady frequencies of four different quartz crystals after irradiating each crystal uniformly by a dose of 5 MRads using ^{60}Co (Average energy = 1.25 MeV).

Crystal No.	Frequencies of the virgin quartz crystals (Hz)	Post-irradiation Frequencies (Hz)	Permanent offset in frequency (Hz)
1	1750058	1751089	1031
2	1870659	1870949	290
3	1749946	1750742	796
4	1870164	1871256	1092

The frequency of this particular crystal specimen while it was virgin was 4997578 Hz and upon irradiation with a dose of 7.5 MRads, the frequency became 4997811 Hz, thus with a net change in frequency of +233 Hz.

Irradiation with Fast Neutrons

Different virgin quartz crystals, after their frequencies were measured, were given varying accumulated doses of neutrons and their post-irradiation frequency behavior was recorded. Like the case of γ-irradiation, the results fall in two broad classifications. Firstly, following the termination of irradiation, the crystals when put into oscillations (the fundamental mode being used) showed an initial instability for some time when the oscillation frequency was unstable and afterwards the frequency was stabilized. This initial instability also was found to be independent of the time of idle keeping of the quartz crystals spaced between the termination of irradiation and generation of mechanical oscillations. This region of frequency instability in terms of time required for stabilization and the frequency changes again depended upon individual specimen and accumulated dose. The steady frequency offset due to neutron irradiation was the value which the crystals attained after culminating the region of frequency instability. These results resemble the irradiation-frequency character due to γ-rays. However, the distinct results with neutrons and gamma irradiation are the manner in which the frequency stabilizes and is described below.

A crystal was irradiated with a dose of 4 x 10^{10}nvt. It was observed that as the crystal started oscillating, there was a region of unstable frequency which persisted for a period of about an hour. The frequency, during this time, approached towards the pre-irradiation value. After the region of frequency instability was over the frequency offset at a value which was almost that for the virgin crystal. The maximum frequency drift during this period of stabilization was about 50 Hz. Another crystal specimen was irradiated with a dose of 7 x 10^{10}nvt and its frequency behavior is shown in fig.7. In this case the frequency changed by an extent of the order of 90 Hz and after about an hour and half the frequency was stabilized. In this case also, it was found that there was hardly any steady offset within the limit of accuracy of experimental observation. A third crystal was given the dose of 9.2 x 10^{10} nvt. This crystal showed a steady offset of about -50 Hz, the frequency stabilizing after about an hour long mechanical oscillations of the crystal. The maximum frequency drift was of the order of 45 Hz in the region of initial frequency instability. Fig.8 depicts the oscillation characteristics of a neutron-irradiated crystal with a dose of 16×10^{10}nvt. The steady frequency offset in this case was of the order of 110 Hz and the frequency instability region extended for about an hour with a maximum frequency drift of about -90 Hz. With prolonged irradiation, it was found that the extent of steady offset was increasingly negative. A different crystal was exposed to 20.7×10^{10} nvt. The frequency offset was about -200 Hz. The instability continued for about two hours in this case with a maximum frequency drift of the order of 250 Hz (Fig.9).

It may be seen from Figs.7 to 9 that during the course of culmination of region of frequency instability after neutron irradiation, the frequency of oscillations tends always towards the value of the frequency of the virgin crystals and in most cases, in the region of unstable frequency, there is a linear variation of frequency with time. This character is different from that existing for γ-irradiation. There, in the region of culmination of initial frequency instability the frequency variations were up and down before the frequencies got stabilized.

A crystal was irradiated in stages and its frequency character was observed. Fig.10 depicts the plot of steady frequency offsets when accumulated doses were given to the crystal, these doses corresponding to the points A,B,C,D,E and F. At the point A, the accumulated dose given to the crystal was 2×10^{10}nvt. It can be seen that there was not much change of frequency at this stage. As the irradiation proceeded to 20×10^{10} nvt (point B) the frequency decreased by a negative offset of about 100 Hz. With further irradiation up to 36×10^{10} nvt (point C), it may be seen that the frequency decreased still further. As the irradiation was still advanced to 60×10^{10}nvt (point D), the net frequency change started upturning and with 80×10^{10}nvt (point E), the frequency was at a value higher than that of the virgin crystal. With still prolonged irradiation of 4×10^{11}nvt (point F) the frequency continued to increase. This observation (increase of frequency with neutron irradiation) fits well with those reported in literature (29-31).

Two crystals were irradiated by fast neutrons with a dose of 2×10^{12} nvt. Fig.11 depicts the post-irradiation oscillation character, for one of the crystals. The character broadly resembles the one reported in the preceding paragraphs. The crystal required about 40 minutes of mechanical oscillations for the region of initial frequency instability to be culminated. It is significant to mention here that in the specimens irradiated by γ-rays or neutrons, the region of frequency instability could be largely reduced or even eliminated if the crystals were warmed to about 100°C for a period of 20-25 minutes. The crystal of which the frequency character is shown in Fig.11 had its frequency 1869981 Hz when it was in the virgin state and after being irradiated by fast neutrons with a dose of 2×10^{12}nvt the steady frequency of the quartz crystal became 1870288 Hz. It can thus be seen that in this case the net frequency change was positive by 307 Hz. The other specimen, irradiated with the same dose (2×10^{12}nvt), also took about 40 minutes for the frequency to stabilize. During this period the maximum frequency change was about 13 Hz. The pre- and post-irradiation frequencies were 1870007 and 1870218 Hz respectively showing a net positive change of 211 Hz. This crystal was given further irradiation of 6×10^{12} nvt. The steady frequency for this crystal after the second irradiation was 1870377 Hz.

A crystal which was given a dose of 8×10^{12}nvt at a stretch showed the frequency character depicted in Fig.12. The crystal required about half an hour for the culmination of initial frequency instability. The virgin crystal oscillated at the frequency of 1870037 Hz while after the neutron irradiation of 8×10^{12}nvt, the steady frequency value was 1870331 Hz. Thus a net positive steady frequency change of about 300 Hz was caused by neutron irradiation.

SUMMARY OF RESULTS

From the foregoing, it can be seen that the following experimental results about the irradiation characteristics of quartz crystals have been obtained.

(1) The initial frequency instability is caused by γ- and neutron irradiation but not by X-irradiation.
(2) For low doses of neutron, X- and γ-irradiation, the steady frequency shift is negative. For the higher doses, the frequency shift becomes positive for the three different kinds of irradiation. In between, a stage would of-course be reached when there is no resultant frequency shift due to irradiation.

DISCUSSION OF RESULTS

Before proceeding further, it should be mentioned that while the negative frequency shift for ionizing radiations has been observed by a number of workers (2-4,6,8), the positive frequency shift has earlier been observed only in a few investigations (30,32). Again, while the positive frequency shifts by neutron irradiation is already known (29-31), the negative frequency shift for low enough doses of neutrons was not observed before.

In the following we will attempt to explain the above results in terms of the lattice structure of the quartz crystals.

Initial Frequency Instability

The neutron and γ-irradiation cause nuclear displacements. While the displacements caused by neutrons are straightforward knock-on processes, those caused by γ-rays are by indirect mechanism through the high energy Compton electrons released by γ-rays causing Coulombic interaction with the nuclei. In the case of X-rays there will be no nuclear displacement caused since the Compton electrons do not have sufficient energy to penetrate the electron cloud of atoms.

It is our opinion that, among the atomic displacements caused by appropriate radiations, a fraction of these are indeed very near the equilibrium atomic positions and hence the atoms so displaced can be brought back to the original positions by very low activation energies supplied by vibrations themselves or by warming.

Steady Frequency Shifts by Ionizing Radiations

The negative frequency shifts are now well known to be caused by the generation of A-centers from their precursors the Al^{3+} -centers, by ionization of electrons of one or more O-atoms bonded with Al^{3+}. Regarding the positive frequency shift there is no mechanism yet propounded.

We believe that the crystal defects are the source of positive frequency shift in two different ways. When the defects are created due to heavy γ-bombardment or neutrons, a positive frequency shift is produced. Also, these defects, whichever of them are electron traps, will give additional positive frequency shift when electrons are trapped therein. Apart from these crystal defects other impurity centers, except the Al^{3+} like the alkali ions trapped in the c-axis at Ge, Ti or other trapping sites will also give a positive frequency shift when populated by trapped electrons. Incidentally, some of these crystal defects on trapping of electrons will form color centers which apart from giving the optical absorption, also give, unlike the case of A-centers, EPR at room temperature. In our observations, we recorded a number of these paramagnetic resonances both sharp and broad

(33). In our opinion, these EPR resonances are good evidence for the presence of the crystal defects giving the positive frequency shift. As to why the crystal defects when produced should give a positive frequency shift may be seen in the following way. These defects disrupt the otherwise perfect atomic arrangement in the crystal, making it more difficult than before, due to the additional potential barriers created by the defects, for atomic displacements to take place, particularly under a shear stress. Thus, with increase of shear modulus of rigidity, the frequency of the AT-cut and other resonators should increase. A support to this argument is provided by the fact that hardness of the crystals was found to increase as a result of γ- and neutron irradiation. Hardness would of-course increase when it becomes more difficult for the atoms to make transverse displacements due to the hindering potential barriers caused by these defects.

As to why the electron trapping of the defects should cause positive frequency shift, i.e. an increase of elasticity is not clear at this stage. Probably, the trapped electrons contribute to extra electrical forces leading to the increased elastic moduli. At any rate, the fact that electron trapping at the defects causes a positive frequency shift is supported by the following experimental observations.* Quartz crystals were irradiated with ultra-violet light (~3650 Å). In all the cases a positive frequency shift, unaccompanied by any initial negative frequency shift was recorded. The uv light would of-course not be able to form A-centers (which would yield a negative frequency shift), but would be able to fill atleast some of the other crystal defects with electrons ionized off from atoms by uv radiations. Thus, it follows that the electron trapping of crystal defects causes a positive frequency shift.

The role of crystal defects in causing positive frequency shift is also supported by the fact that all those crystals which gave the positive shift were not optically clear quartz, this fact indicating the presence of defects and other impurities in the lattice which become color centers. Optically clear quartz, did not, in accordance with earlier results of Capone et.al.(8), King (4) and Frondel (1,2) give positive frequency shift.

With all the above in view, the following seems to be taking place by γ-irradiation of quartz crystals.

In the early stages of γ-irradiation, the ionization of the Al^{3+} -centers, due to their higher cross section,

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*Details of this work will be published separately.

takes place predominantly giving rise to negative frequency shift. With continued irradiation, crystal defects and other impurity centers having a lesser cross section get increasingly trapped with the electrons, all this producing the turn over of frequency shift observed. For higher doses of γ-irradiation, the magnitude of positive frequency shift is all the greater due to the generation of defects by γ-rays themselves.

Steady Frequency Shifts due to Neutron Irradiation

The role of crystal defects in causing a positive frequency shift straightforwardly explains the effect of higher doses of neutrons in causing the frequency increase.

Regarding the effect of low doses of neutron irradiation causing the negative frequency shift, no straightforward fundamental explanation can be given at the present. However, the decrease of frequency is in accordance with the observation of expansion of the quartz lattice under neutron irradiation (34). The expansion would lead to decrease of interatomic forces and the associated moduli of elasticity.

At the higher neutron doses, the lattice expansion no doubt takes place increasingly but the role of expansion in causing the decrease of frequency seems to be exceeded (atleast for reasonable neutron doses used for obtaining frequency offsets) by the role of defects, so generated by neutrons, in causing the frequency shift. That the role of defects on the elastic moduli particularly the shear modulus is preponderant is shown by the increase of hardness by neutron irradiation referred to above.

APPLICATIONS

The present work, apart from giving explanations for some of the frequency effects of irradiation, also indicates applications towards radiation hardening of crystals, this radiation hardening making it possible to use quartz crystals in space satellites to generate precise frequencies and time intervals. It is clear that at the extreme of the negative frequency shift the differential of the frequency shift with continued irradiation vanishes. It follows that the crystal will assume radiation hardness for some range of irradiation dose if the crystal has already been irradiated enough for obtaining the maximum of the negative frequency shift. This technique would apply both for γ-

and neutron irradiation with the greater returns for neutron irradiation since for this no other technique has yet been developed. In contrast, for γ-irradiation the method of using vacuum swept crystals (35) for satisfactory radiation hardening (though not perfect) has already been established.

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REFERENCES

1. J.C. King, "Hardening quartz resonators to ionizing radiation -a review" Proc. Int. Conf. Evaluation of Space Environment on Materials, Centre Spatial de Tououse, June 17-21, 1974.
2. C. Frondel, Am. Miner. 30, 416 (1945).
3. C. Frondel, Am. Miner. 30, 432 (1945).
4. J.C. King, Bell System Tech. Jour. 38, 573 (1959).
5. R.A. Poll and S.L. Ridgway, IEEE Trans. Nucl. Sc. NS-13, 130 Dec'(1966).
6. D.B. Fraser, Jour. Appl. Phys. 35, 2913 (1963).
7. D.B. Fraser, Physical Acoustics (Ed. W.P. Mason), Academic Press, New York, 5, 59 (1968).
8. B.R. Capone, A. Kahan, R.N. Brown and J.R. Buckmelter IEEE Trans. Nucl.Sc. NS-17, 217 (1970).
9. T.M. Flanagan and T.E. Wrobel, IEEE Trans. Nucl. Sc. NS-16, 130 Dec'(1969).
10. E.P EerNisse,"Radiation Effects in Swept and Unswept Optical Grade Synthetic Quartz AT- Resonators" Tech. Memo. Sandia Laboratories, SC-TM-70-417, July 1970.
11. B. Capone, A. Kahan and B. Sawyer, Proc. Annual Frequency Control Symposium, US Army Electronics Command, Atlantic City, New Jersey, 25, 109 (1971).

12. A. Kahan, B.R. Capone and R.N. Brown, "Radiation Hardness of Electrodiffused (Swept) Electronic Grade Quartz," Tech. Memo. LQ-16 Air Force Cambridge Research Laboratories, Bedford, Mass. 16 March 1973.

13. J.C. King (Ed.), Radiation Effects in Quartz Crystals, Radiation Effects, 26, No.4 (1975).

14. T. Aoki, K. Norisawa and M. Sakisaka, Japanese Jour. Applied Physics, 15, 2131 (1976).

15. T. Aoki, K. Norisawa and M. Sakisawa, Japanese Jour. Applied Physics, 15, 749 (1976).

16. Paul Pellegrini, F. Euler, A. Kahan, T.M. Flanagan and T.F. Wrobel, IEEE Trans. Nucl. Sc. NS-25, 1267, Dec'1978.

17. F. Euler, P.Ligor, A. Kahan,P.Pellegrini, T.M. Flanagan, and T.F. Wrobel, Proc. Annual Frequency Control Symp., US Army Electronics Command, Atlantic City, New Jersey 32, 24 (1978).

18. H.G. Lipson, F. Euler and P.A. Ligor, Proc. Annual Frequency Control Symposium, US Army Electronics Command, Atlantic City, New Jersey, 33, 122 (1979).

19. F. Euler, H.G. Lipson and P.A. Ligor, Proc. Annual Frequency Control Symposium, US Army Electronics Command, Atlantic City, New Jersey, 34, (1980).

20. J.C. King and H.H. Sander, IEEE Trans. Nucl. Sc. NS-19, 23 (1972).

21. E.F. Hartman and J.C. King, Proc. Annual Frequency Control Symposium, US Army Electronics Command, Atlantic City, New Jersey, 27, 124 (1973).

22. J.C. King and H.H. Sander, Radiation Effects, 26, 203 (1975).

23. E.F. Hartman and J.C. King, Radiation Effects, 26, 219 (1975).

24. J.C. King and H.H. Sander, IEEE Trans. Nucl. Sc., NS-20, 117 (1973).

25. Harish Bahadur and R. Parshad, Rev. Scientific Instruments - in press.

26. T.J. Young, D.R. Koehler and R.A. Adams, Proc. Annual Frequency Control Symposium, US Army Electronics Command, Atlantic City, New Jersey, 32, 34 (1978).

27. D.R. Koehler, Proc. Annual Frequency Control Symposium, US Army Electronics Command, New Jersey, 33, (1979).

28. Harish Bahadur and R. Parshad, Indian Jour. Physics, 53(A), 239 (1979).

29. J.C. King and D.B. Fraser, Proc. Annual Frequency Control Symposium, US Army Electronics Command, Atlantic City, New Jersey, 16, 7 (1962).

30. F.B. Johnson and R.S. Pease, Phil. Mag. 45, 651 (1954).

31. J.C. King,"Final Report on Fundamental Studies of the Properties of Natural and Synthetic Quartz Crystals", 15 Jan., 1960, (Contract DA 36-039 Sc- 64586).

32. R.B. Belser and W.H. Hicklin, Proc. Annual Frequency Control Symposium, US Army Electronics Command, New Jersey, 16, 110 (1962).

33. Harish Bahadur, S.K. Gupta and R. Parshad, Proc. 1980 Nuclear Physics and Solid State Physics Symposium, Deptt. Atomic Energy, India - in press.

34. M. Wittels, Phys. Rev. 89, 656 (1953).

35. J.C. King, U.S.Patent no. 3,932,777, Jan.13, (1976).

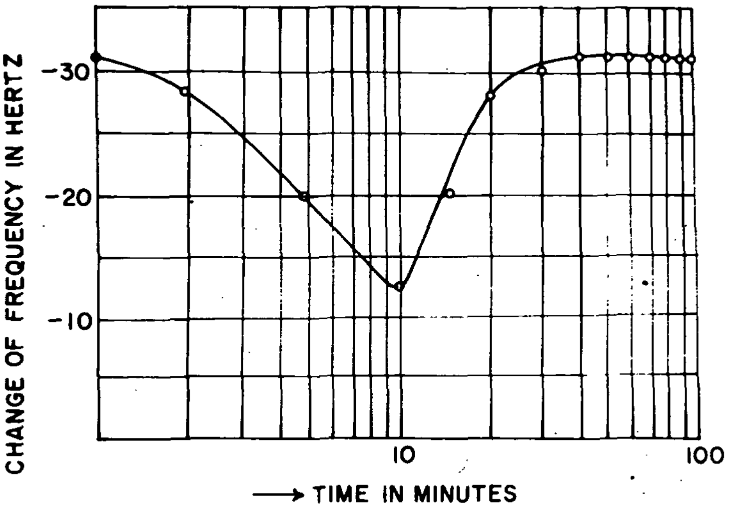

Fig. 1. Frequency behavior of a quartz crystal (AT-cut, 1·87 MHz) after irradiation with gamma rays (dose = 2000 Rads, using ^{60}Co) and allowed to oscillate.

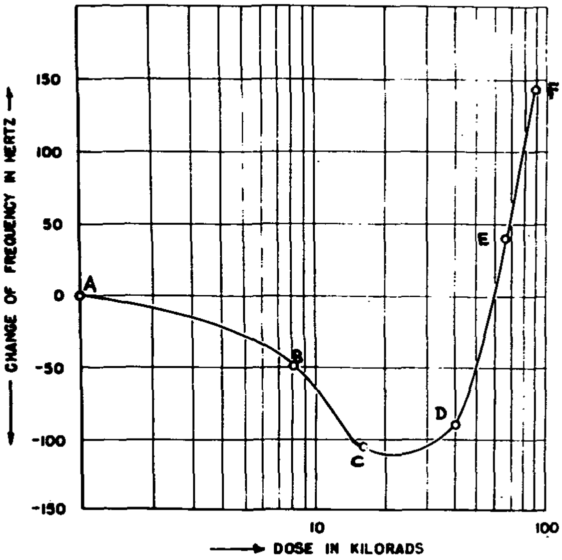

Fig. 2 Steady frequency offsets of a natural AT-cut quartz crystal irradiated in stages A, B,....E. Accumulated doses at B = 8,000 Rads, at C = 16,000 Rads, at D = 40,000 Rads, at E = 64,000 Rads and at F = 88,000 Rads.

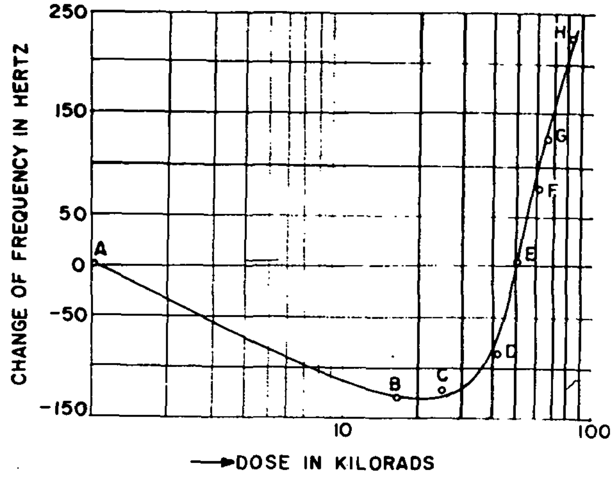

Fig. 3 Steady frequency offsets of a natural quartz crystal (AT-cut, 1·87 MHz) irradiated by gamma rays in stages A, B, C----H (using ^{60}Co). Accumulated doses at A = 8000 Rads, at B = 16,000 Rads, at C = 40,000 Rads, at D = 64,000 Rads, at E = 88,000 Rads, at F = 112,000 Rads, at G = 136,000 Rads, and at H = 160,000 Rads.

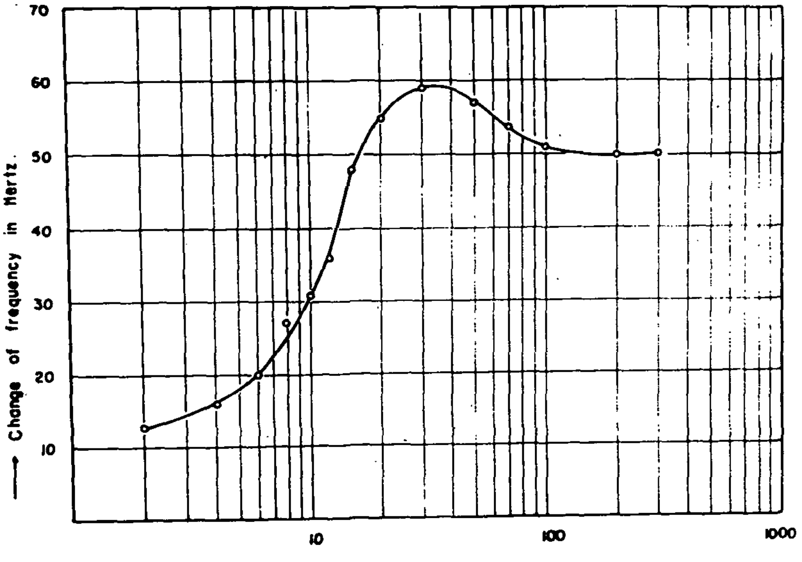

Fig. 4 Frequency behavior of a natural AT-cut (1.87 MHz) quartz crystal after being irradiated by gamma rays (dose = 7.5 MRads) and allowed to oscillate.

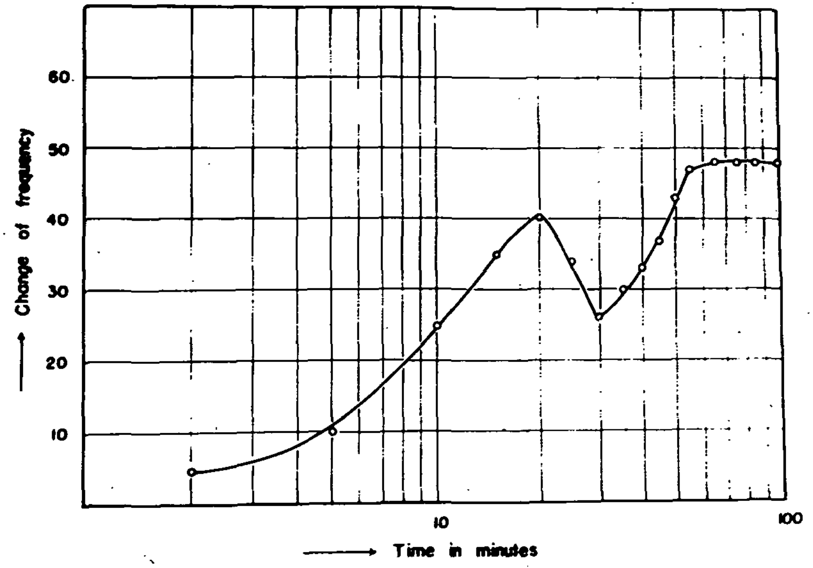

Fig.5 Initial frequency instability characteristics of a natural AT-cut quartz crystal (1.87 MHz) after irradiation by gamma rays (dose = 7.5 MRads) using ^{60}Co and allowed to oscillate.

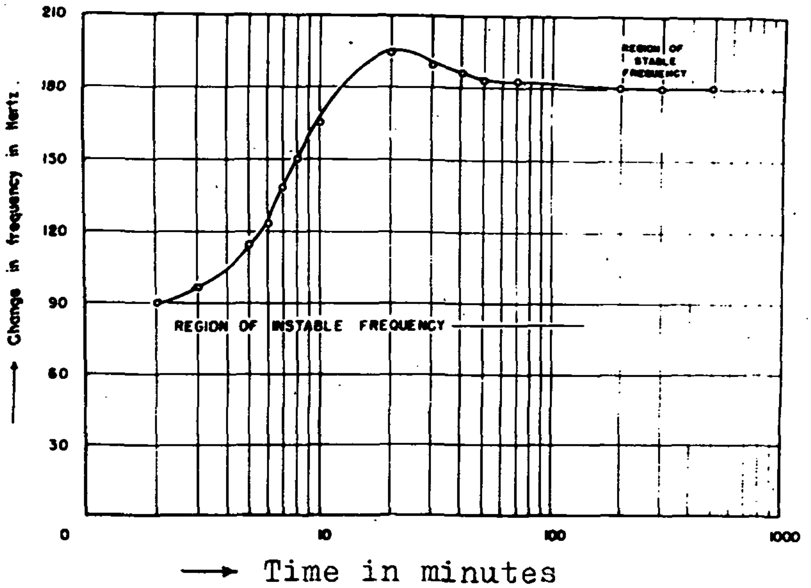

Fig.6 Initial frequency instability characteristics of a natural AT-cut (5 MHz) quartz crystal after irradiation by gamma rays (dose = 7.5 MRads) using ^{60}Co and allowed to oscillate.

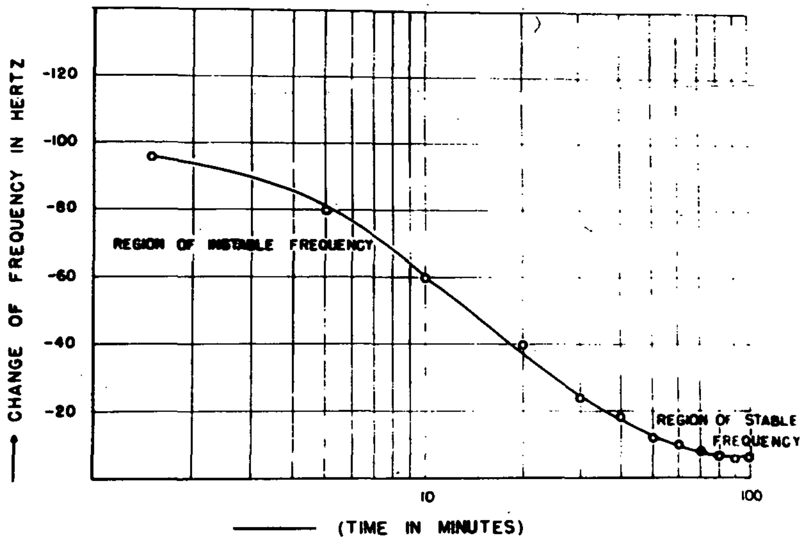

Fig- 7 Frequency stability characteristic of a quartz crystal, irradiated by hot neutrons with a dose of 7×10^{10} nvt, after setting it into oscillations.

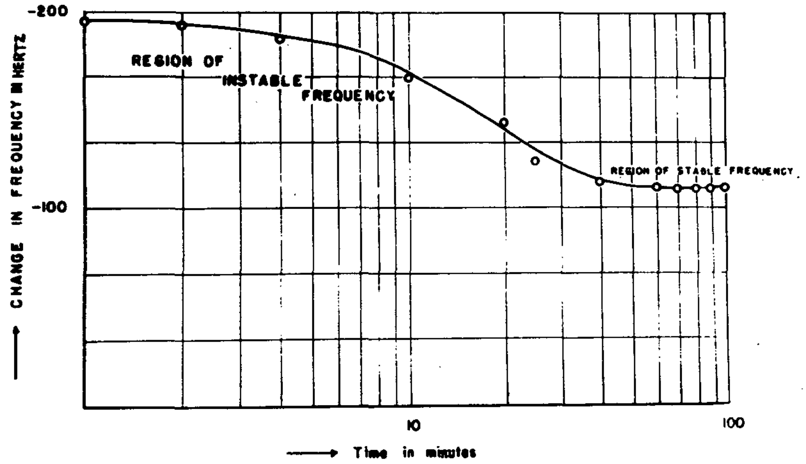

Fig.8 Frequency behavior of a natural AT-cut quartz crystal (1.87 MHz) irradiated by fast neutrons with a dose of 16×10^{10} nvt and allowed to oscillate.

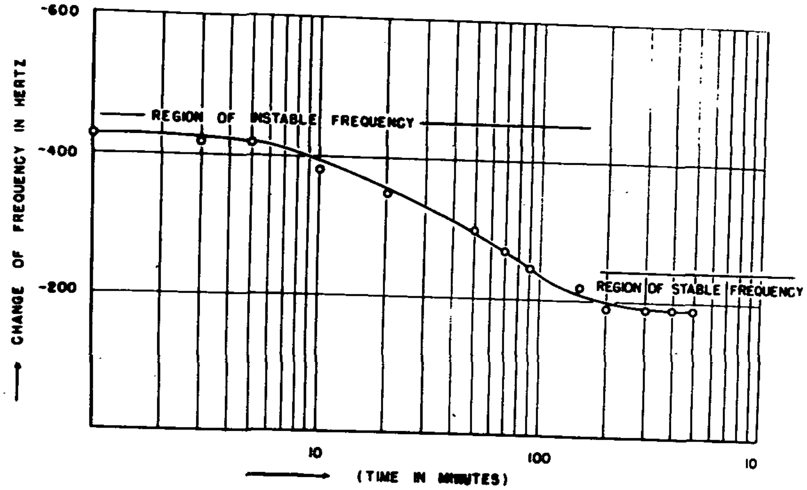

Fig.9 Frequency behavior of a natural AT-cut quartz crystal (1.87 MHz) irradiated by fast neutrons with a dose of 20.7 x 10^{10} nvt and allowed to oscillate.

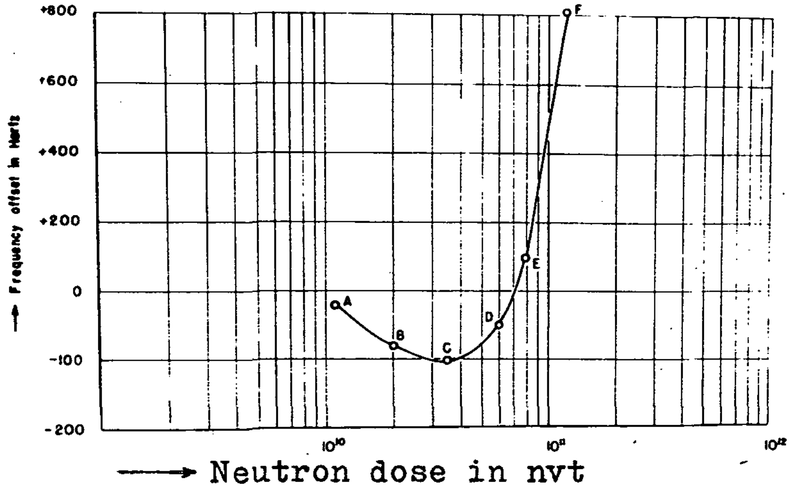

Fig. 10 Steady frequency offsets of a quartz crystal of 1.87 MH_Z irradiated at different accumulated neutron doses, using ^{241}Am - Be, after the temporary frequency isntability has been removed. Doses at A = 2 x 10^{10} nvt, B = 20 x 10^{10} nvt, C = 36 x 10^{10} nvt, D = 60 x 10^{10} nvt E = 80 x 10^{10} nvt, F = 4 x 10^{11} nvt.

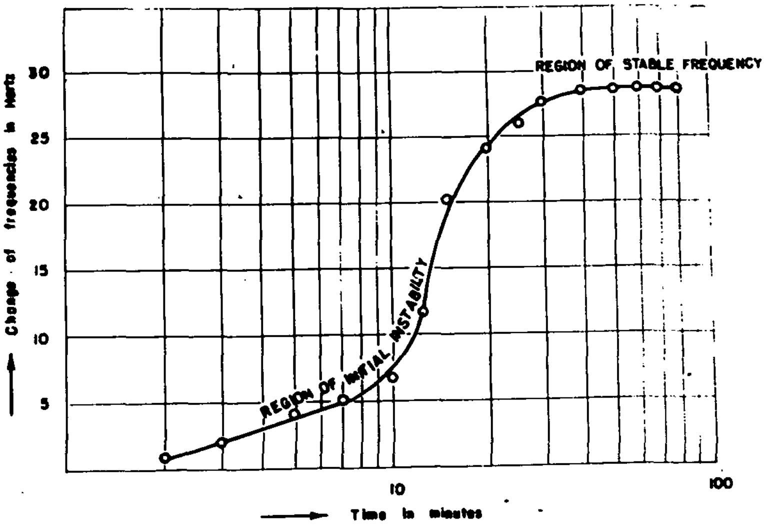

Fig- 11 Frequency behavior of a natural AT-cut (1.87 MHz) quartz crystal with time after irradiation with fast neutrons with accumulated dose of 2×10^{12} nvt using 241 Am - Be.

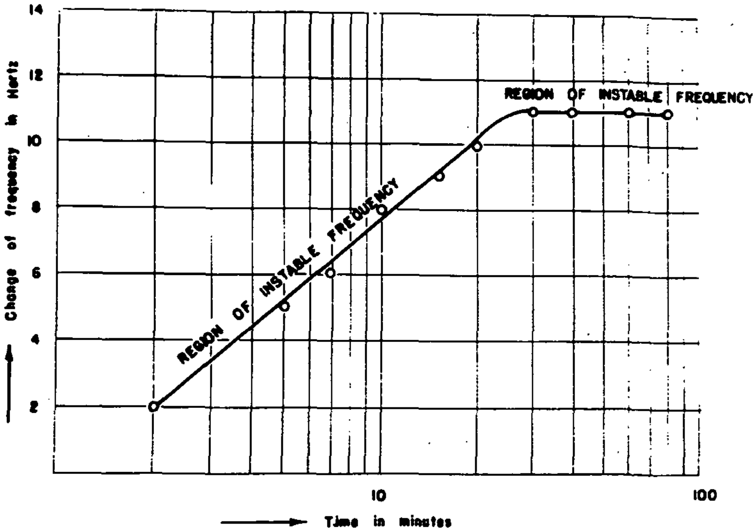

Fig- 12 Frequency behavior of a natural AT- cut (1.87 MHz) quartz crystal with time after irradiation by fast neutrons with accumulated dose of 8×10^{12} nvt using ^{241}Am Be.