

Effects of solvent and growth conditions on mechanical Q of quartz single crystals

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The effects of basic sodium carbonate solvent, various types of lithium salt dopants, and growth conditions on mechanical Q of synthetic quartz crystals have been investigated. Varying concentrations of basic solvent were used, with and without addition of measured quantities of lithium salt impurities. The impurities were lithium carbonate, lithium nitrite, lithium nitrate and lithium fluoride. Large Y -bar single crystals of quartz were synthesized in runs of about one month's duration. Development of opacity in the crystals by X-ray irradiation and heat-treatment was investigated and the average Q values of the crystals were determined from infrared absorption measurements. It is concluded that low growth rate resulting from fine control of experimental variables yields better crystals and that there are certain optimum concentrations of impurities and solvents which help in improving quality of the crystals.

1. INTRODUCTION

For many years the quality of quartz single crystals synthesized in different laboratories was inferior to that of flawless natural quartz single crystals, and much endeavour was therefore directed to improving their quality. Adding Li^+ to the hydrothermal solution, tried by King, Ballman & Laudise (1962), resulted in a marked improvement in quality of the synthetic crystals. Further research along this line (Ballman *et al* 1966, Rudd *et al* 1967, Toyocom 1970), using NaOH solvent and LiNO_2 dopant, culminated in the production of crystals having a mechanical Q as high as 2.5×10^6 . Sawyer (1972) has shown that synthetic crystals grown at a slow rate in a Na_2CO_3 medium, designated by him as *High Q*, possess a maximum Q value of about 2.5×10^6 . *Premium Q Grade Quartz*, reported by Capone, Kahan & Sawyer (1971) has an even larger value of Q , i.e., about 3×10^6 . It is certain that a lithium salt was used as dopant for production of the latter, but it is not known whether it was grown in Na_2CO_3 medium. The International Electrotechnical Commission (1971a, 1971b) has recently proposed that the minimum Q for the best grade of synthetic quartz should be raised from $Q > 2 \times 10^6$ to $Q > 3 \times 10^6$.

Mechanical Q is the reciprocal of acoustic loss or internal friction (Q^{-1}) of a crystal. Internal friction is a measure of the energy that is lost in a crystal in the form of heat as the crystal unit operates and is related to imperfections in the

crystal structure. Because of the small numerical values obtained, the reciprocal of internal friction, namely mechanical Q , has been widely used in describing the quality of quartz provided the frequency and temperature ranges are specified.

The effects of growth condition, and varying concentrations of basic sodium carbonate solvent and four types of lithium salt dopants (Li_2CO_3 , LiNO_2 , LiNO_3 and LiF) on mechanical Q of the crystals synthesized have been discussed in this paper. Large Y -bar (about 3"-6" in length along the Y -axis) single crystals of quartz were grown for this purpose in runs of about one month's duration. Seven runs of this series have already been reported in some other connection in an earlier paper from this laboratory (Chakraborty & Saha (1973), Table 3).

2. EXPERIMENTAL TECHNIQUE

The method of growing the crystals has been described in great detail in papers already published from this laboratory (Chakraborty *et al* 1973, Bandyopadhyay *et al* 1966).

Some of the Y -bar seed rods (approximately 0.1" \times 0.2" in cross-section) were sawed from imported synthetic crystals. Some other flawless natural quartz Y -bar seed rods were kindly supplied by Messrs. Bharat Electronics Ltd., India. Seed-processing technique has been described in detail in another paper (Chakraborty *et al* 1973).

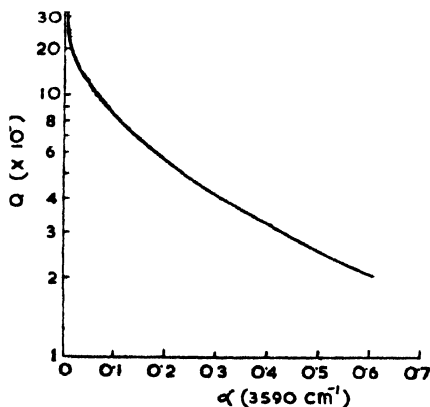


Fig. 1. Relation between absorption coefficient of infrared radiation and Q -values of synthetic quartz (after Toyocom 1970)

The grown crystals were sectioned perpendicular to the Y -axis of quartz, and then ground and polished to exactly 5mm. thick plates. Care was taken to avoid the capped portions of the crystals since the natural face cappings (predominantly the major and minor rhombohedra of quartz) have been reported to be covered by a very imperfect layer of growth (Lang *et al* 1967) which would interfere with mechanical Q measurements of the thick polished plates.

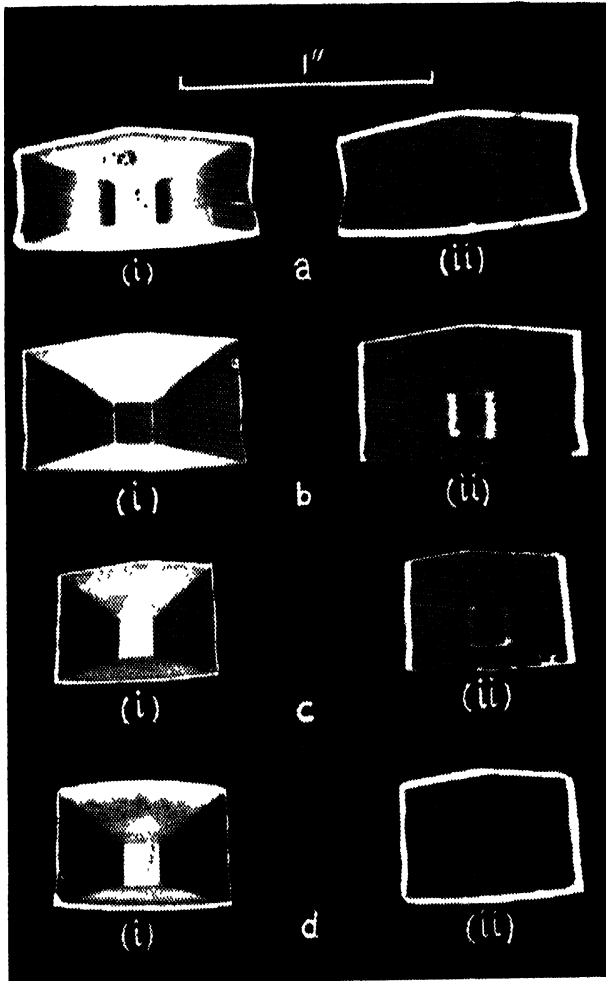


Fig. 2. Irradiated (i) and heat-treated (ii) processed synthetic quartz single crystals ($X-Z$ plane)

- (a) Crystal of run No. 1, table 1: $Q = 0.25 \times 10^6$
- (b) Crystal of run No. 1, table 1: $Q = 0.32 \times 10^6$
- (c) Crystal of run No. 18, table 1; $Q = 0.55 \times 10^6$
- (d) Crystal of run No. 3, table 1; $Q = 0.80 \times 10^6$.

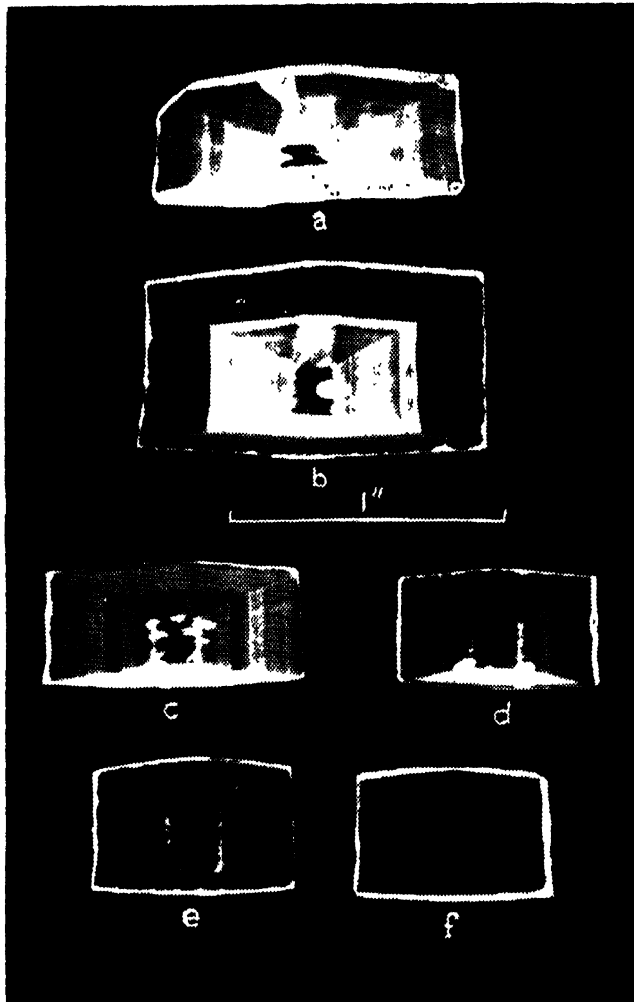


Fig. 3. Heat-treated processed plates from synthetic quartz single crystals ($X-Z$ plane)

- (a) Crystal of run No. 21, table 1 $Q = 0.15 \times 10^6$
- (b) Crystal of run No. 13, table 1 $Q = 0.16 \times 10^6$
- (c) Crystal of run No. 5, table 1 $Q = 0.26 \times 10^6$
- (d) Crystal of run No. 6, table 1 $Q = 0.51 \times 10^6$
- (e) Crystal of run No. 8, table 1 $Q = 0.60 \times 10^6$
- (f) Crystal of run No. 3, table 1 $Q = 0.80 \times 10^6$

Two photographs of each of the X-ray irradiated and heat-treated specimens (the methods of X-ray irradiation and heat-treatment are described below) were taken by transmitting a beam of parallel light through the polished plates mounted on glass plate and exposing the photographic paper directly to the beam. The plates of figures 2 and 3 therefore show the reverse print-out effect, i.e., the opaque and translucent regions becoming white and the transparent regions becoming dark.

(i) *X-ray irradiation*

Polished plates processed from ten crystals (run nos. 1, 2, 3, 4, 10, 14, 16, 17, 18 and 19, table 1) were irradiated with X-ray from an instrument operating at 50 KV, 30 mA. Exposure time given for each sample was 1 hour and tungsten target was used*. Some of the irradiated crystals have been shown in figure 2.

(ii) *Infrared absorption measurement*

The method of obtaining the mechanical Q values of the crystals involves measurement of optical absorbance of the polished plates in the near infrared (3590 cm^{-1}) region. Some of the measurements were carried out with the highly sensitive Hilger Watts infrared spectrophotometer (Model No. H 800) of the institute with a $0.8\text{ cm} \times 0.2\text{ cm}$. vertical slit-type aperture arrangement; most measurements were however carried out with another Hilger Watts infrared equipment with a 2 cm. circular type of aperture arrangement.** Those two types of measurements have been distinguished in table 1.

The optical absorption data are then converted to absorption coefficients ($\alpha_{3590}\text{ cm}^{-1}$). The absorption coefficient is measured by subtracting the logarithm of the absorbance at 3590 cm^{-1} , where the OH^- absorption frequency is located, from logarithm of the absorbance at 3800 cm^{-1} and dividing the result by the thickness of the sample in centimeters. It can be assumed that loss due to reflection and scattering at the two neighbouring wavelengths are similar. Hence their effects will be effectively removed by subtraction of the logarithm of 3800 cm^{-1} absorbance (background absorption) which includes no OH^- absorption effect. The relation between the absorption coefficient ($\alpha_{3590}\text{ cm}^{-1}$) and Q for 5 MHz (5th Overtone) crystal units, at room temperature, is shown in figure 1, adopted from

*We are grateful to Dr. S. Taki, Research Director, Toyo Communication Equipment Co. Ltd., Japan, for preparing two adjacent polished plates from each of the crystals of those ten runs, for measuring the mechanical Q from one set of those plates, and for carrying out later heating experiments with the same set of plates, when one of the authors went to Japan in 1970 on a JSPS fellowship. The other set of plates was sent by Dr. Taki for irradiating them with X-ray to Dr. M. Yoshimatsu, Research Director, Rigaku Denki Co. Ltd. We are indebted to Dr. Yoshimatsu for kindly irradiating this set of plates.

**Kind courtesy of Bose Institute, Calcutta.

Table 1

Run No.	Actual molar composition	Temperature T°C	Pressure P psi	Temperature gradient T°C	Growth rate (GR) (mils/day)		Duration days	Average mechanical $Q \times 10^6$	Run Nos. of Chakraborty <i>et al.</i> (1973a)
					basal	Av. x			
1	2	3	4	5	6	7	8	9	10
1.	0.6 NC	343 ± 3.5	7300 ± 200	31 ± 4	30.5	11.5	27	0.32	45
2.	0.4 NC 0.4 LN ₂	346 ± 4.5	7750 ± 1175	41 ± 9	36.5	14	24	0.25	50
3.	0.6 NC 0.04 LN ₂	341 ± 3.5	6250 ± 500	34 ± 2	23.5	12	26	0.80	51
4.	0.4 NC 0.08 LN ₂	342 ± 3.5	7550 ± 400	43 ± 1.5	41	15	23	0.23	
5.	0.4 NC 0.08 LN ₂	336 ± 2.5	5800 ± 200	33 ± 2	33	10	26	0.26	
6.	0.4 NC 0.08 LN ₂	338 ± 8	5700 ± 1300	33 ± 2	30	11	20	0.51	
7.	0.4 NC 0.08 LN ₂	334 ± 2	6000 ± 200	58 ± 2	32 30.5	9.5 9.5	27 27	0.51 (Top Crystal) 0.54 (Bottom Crystal)	
8.	0.4 NC 0.08 LN ₂	340 ± 4.5	7100 ± 750	29 ± 5	30	n.d.	21	Top—0.60 Bot—0.73	
9.	0.4 NC 0.08 LN ₂	332 ± 2	5750 ± 100	37 ± 2	23	8.5	34	Top—0.48 Bot—0.80	
10**.	0.5 NC 0.08 LN ₂	342 ± 13	6600 ± 1250	42 ± 12	35	17.5	24	0.26	
11.	0.5 NC 0.08 LN ₂	337 ± 2	6000 ± 200	28 ± 1	30	11.3	26	Top—0.43 Bot—0.60	
12.	0.6 NC 0.08 LN ₂	340 ± 1.5	6700 ± 200	41 ± 1	29	12	33	0.24	
13.	0.7 NC 0.02 LN ₂	345 ± 2	6000 ± 200	44 ± 2	36.5	14	33	0.16	53

Table 1—contd

1	2	3	4	5	9	7	8	9	10
14.	0.4 NC 0.17 LN ₂	345 ± 4	6200 ± 500	39 ± 4	28	8.8	28	0.55	
15.	0.6 NC 0.17 LN ₂	335 ± 1	6000 ± 100	40 ± 1	32	13	28	Top—0.23 Bot—0.27	
16.	0.4 NC 0.04 LC	340 ± 2.5	6500 ± 400	40 ± 2.5	44	14	33	0.10	
17.	0.5 NC 0.04 LC	340 ± 2.5	7000 ± 500	29 ± 1	34	14	29	0.25	47
18.	0.6 NC 0.04 LC	334 ± 2.5	6900 ± 350	38 ± 2	23	11.5	24	0.55	48
19.	0.6 NC 0.04 LN ₃	339 ± 3.5	8000 ± 200	31 ± 5	30	14	25	0.30	49
20**.	0.6 NC 0.04 LF	333 ± 2	5700 ± 350	37 ± 2	39	13	26	Top—0.10 Bot—0.21	
21.	0.6 NC 0.04 LF	336 ± 3	6000 ± 500	66 ± 3	44.5	n.d.	27	0.15	

Abbreviations used : NC Na₂CO₃, LN₂ = LiNO₂, LC = Li₂CO₃, LN₃ = LiNO₃, LF = LiF.
n.d. = not determined, bot = bottom.

**Cracked crystals.

***Growth zone temperature

Italicised mechanical Q values of Column (9) are those measured by our Institute's Infrared equipment; the others were measured by Bose Institute's (Calcutta) Infrared equipment.

Toyocom (1970) with the kind permission of M/s. Toyo Communication Equipment Co. Ltd. Japan (personal communication with Dr. Taki). The mechanical Q values of our crystals were derived by plotting our absorption coefficients on this standard curve***. A method for determining the mechanical Q of synthetic quartz, similar to the method described above, has been adopted by International Electrotechnical Commission for inclusion in a synthetic quartz specifications brochure which is under preparation (Sawyer 1972).

It would have perhaps been better to plot our data on the standard computer-smoothed curve for *slow-growth* cultured quartz using sodium carbonate as the basic solvent medium, for which the standard equation has also been derived by Sawyer (1972). However, since it had not been possible to use a beam condenser and N_2 purge for obtaining precise measurements with the infrared equipment, and since most of the heat-treated specimens showed growth layering, it was felt that no significant advantage could be gained by using Sawyer's curve. Whatever data that have been obtained should be treated as *average* Q of the crystal over a very large area of the sample, which has an inhomogeneous distribution of impurities in the seed, Z , $-X$ and $+X$ regions, and which includes inhomogeneous growth layers.

It has been observed by Dodd & Fraser (1965) that the impurities (presence of which results in imperfections in the crystals) responsible for internal friction at and around room temperature are hydrogen and hydrogen bonded OH which has an infrared absorption band at 3590 cm^{-1} . So it is easy to determine qualitatively OH^- content of a crystal and thereby quality, i.e., mechanical Q , of the crystal can be evaluated. It has been reported that absorption band at 3590 cm^{-1} is not affected by alkali content of the crystal (Toyocom 1970). Impurities like iron do however increase acoustic loss in quartz, specially in low Q quartz (Sawyer 1972).

It was necessary for the 6" long crystals to process the plates from the top and bottom portions (with respect to location in the growth zone of the liner) and carry out measurements on them, in view of the lengthwise variation of mechanical Q reported by Rudd & Lias (1967). These data have been incorporated in table 1. The plates were sawed from the middle in the case of the smaller crystals ($2\frac{1}{2}$ "- $3\frac{1}{2}$ " in length).

(iii) Heat treatment

The polished plates used for infrared absorption measurements were then heat-treated to investigate the development of opacity due to impurities present in the crystals. They were slowly heated in a furnace to a temperature of 550°C

***Dr. Taki's measurements of mechanical Q of the ten crystals mentioned in a previous footnote were exactly duplicated for the same crystals in our laboratory using our highly sensitive infrared equipment and fairly concordant results were obtained.

(attained in 5 hrs) and allowed to remain at that temperature for 24 hrs. Some of the heat-treated crystals have been shown in figures 2 and 3.

3. RESULTS AND DISCUSSIONS

(i) Nature of the impurities

Figure 2 shows the irradiated polished plates of crystals of some runs and their heat-treated equivalents, the heat-treatment having been performed on adjacent polished plates. All the seed crystals of those runs were cut from natural quartz crystals, kindly provided by Bharat Electronics Ltd. Those natural seed rods came in two batches, figure 2(a, c and d) showing seeds of one lot and figure 2(b) showing a seed of the other. An interesting thing to observe is that seed portion of the X-ray irradiated plate of figure 2b(i) does not show any development of opacity whereas X-ray irradiated plates of figure 2 (a(i), c(i) and d(i)) show development of considerable opacity, usually brownish in colour. It is obvious that the distribution of impurities which are affected by X-ray irradiation, is different for the two different lots of natural quartz seed rods. Moreover, the adjacent heat-treated plates of figure 2 show that those impurities are not affected by heat-treatment. Some of those impurities are also present in the $+X$ and $-X$ zones of the irradiated plates and to a lesser extent in the Z zones of a low grade synthetic crystal (figure 2a(i), $Q = 0.16 \times 10^6$).

The heat-treated polished plates have been arranged in order of increasing mechanical Q in figure 3. The degree of development of opacity, milky-white in appearance for heat-treated crystals, decreases with increasing mechanical Q . It is well-known that this development of opacity in heat-treated crystals is mainly due to the OH^- radical (Dodd *et al* 1965). This is also quite obvious from the fact that the milkiness of the heat-treated crystals can be directly correlated with the mechanical Q values of those crystals derived from infrared absorption measurement of the 3590 cm^{-1} absorption band for OH^- , as in figure 3.

However, the results of figure 2 show that OH^- radical is not the only factor governing mechanical Q . Natural quartz crystals are known to contain very little OH^- . In spite of this the seed portions of the irradiated crystals of figure 2 show high degree of development of opacity, presumably due to presence of some other types of impurities. The grown portion of those crystals, specially the $+X$ and $-X$ zones, show considerable irradiation darkening but not heat-treatment translucency. Thus it is obvious that some impurities other than OH^- must have also been incorporated in the synthetic crystals. The only possibility seems to be iron in the ferric state substituting for Si^{4+} , the charge deficiency being compensated for by substitution of O^{2-} of the tectosilicate framework by the OH^- radical. Since mild steel liners were used for growing our crystals, this seems to be quite possible. This leaves behind Na^+ and Li^+ as the

only other possible structural impurities. The ionic radius of Na^+ is too large to fit into the α -quartz channel. Li^+ seems to be a good possibility. Nevertheless, Rudd & Lias (1967) have shown that the synthetic crystals grown in Li^+ doped solvent medium do not contain more than 5 ppm of Li^+ over undoped synthetic crystals. Also in a previous paper from this laboratory Bandyopadhyay *et al* (1966) have demonstrated that no Li^+ could be detected in a synthetic crystal grown in a Li^+ doped solvent medium.

The heat-treated crystal of figure 3(b) is quite interesting. It is difficult to realise how the experimental variables could suddenly assume such a steady state, or the interior of the autoclave and liner could suddenly become so free from impurities, so as to give a sharply defined almost clear outer core after such a heavily zoned opaque inner core. The average mechanical Q of this crystal considering both zoned and unzoned portions is 0.16×10^6 .

(ii) *Nature of experimental variables*

The experimental conditions of the runs, the growth rate along the optic axis considering both sides of the seed, the average of growth rates along the $+X$ and $-X$ directions, and the *average* mechanical Q of the grown crystals have been listed in table 1.

Before analysing the data of table 1 it would perhaps not be out of place to characterize the nature of the experimental variables and the dependence of growth rate on them in a summary form. Details would be available in two papers already communicated from this laboratory (Chakraborty *et al* 1973, Chakraborty *et al* 1974). This would make it easier to properly assess the relative importance of the variables in affecting the quality of the crystals.

The variables that will have to be considered for such long runs are

- (1) temperature
- (2) fluctuation of temperature
- (3) temperature gradient
- (4) fluctuation of temperature gradient
- (5) pressure
- (6) fluctuation of pressure
- (7) nature and concentration of solvent and dopant.

These can be examined one by one.

Temperature is a purely independent variable. There will be two independent temperature variables, i.e., growth zone temperature and nutrient zone temperature, but their degree of inter-dependence depends on the number of controls used. For the system used in this laboratory only the growth zone temperature can be considered as a purely independent variable, because only one control thermocouple (for the growth zone) was used. The nutrient zone temperature

depends on (a) growth zone temperature, and (b) the rate of heating. Mention must be made here of the fact that furnace characteristics are assumed to be constant because the same type of furnaces were used for all the runs.

The range of temperature fluctuation depends on the nature of controls available. This was fairly constant for most of the cases except for few runs which were affected by short-time power failure.

Pressure under normal circumstances should also be treated as an independent variable. But for such closed-system hydrothermal experiments it is completely controlled by percentage fill of the autoclave (Bandoyadhyay *et al* 1966), that is, by the P-V-T relationship of aqueous alkaline solutions. Obviously, therefore, provided there is no leakage in the run, pressure and its range of fluctuation would be completely dependent on temperature and its range of fluctuation. Pressure, then, becomes a dependent variable for such experiments.

Temperature gradient and its range of fluctuation depend on (a) rate of heating, (b) furnace characteristics (for experiments such as ours), (c) temperature and its range of fluctuation. It was curious that temperature gradient should be dependent on rate of heating, but this was repeatedly found to be true. A fast rate of heating usually ensures a high temperature gradient and a slow rate of heating a low temperature gradient. As a figure of merit it can be stated that a heating period of $3\frac{1}{2}$ hours causes a temperature gradient of 66°C (run no. 18, table 1) whereas the normal gradient was about 40°C for a 6 hours heating period. As noted before, the furnace characteristics being the same for all the runs, the experimental set-up did not affect temperature gradient appreciably. Temperature also indirectly influences temperature gradient. Usually a high temperature results in a high temperature gradient. As a figure of merit it can be stated that a temperature of 345°C causes a gradient of about 44°C whereas a temperature of 333°C results in a gradient of 37°C where the same rate of heating was employed. Range of temperature gradient fluctuations will mainly be dependent on range of temperature fluctuations.

The nature and concentration of solvent and dopant can, of course, be completely controlled and therefore it is a purely independent but fully controllable variable.

The discussion will perhaps make it obvious that for an experimental set-up like that of ours the independent variables on which long-time operation of the process will depend are (1) temperature, (2) range of temperature fluctuation, and (3) rate of heating.

The growth rate is a variable which is completely dependent on those independent variables. It would therefore be necessary to have very fine control of temperature and its fluctuations, rate of heating, and the other controllable

variables (composition of solvent, percentage baffle opening etc.) to make a proper assessment of the growth rate data.

In view of these facts it would be impossible to distinguish the influence of growth rate, a fully dependent variable, on the mechanical Q of the crystals, from *individual* effects of the independent variables. The subsequent conclusions drawn from the data of table 1, given below, should be judged by this criterion.

4. SUMMARY AND CONCLUSIONS

The following conclusions could therefore be drawn by investigating the data of table 1.

(1) At low concentration of the basic solvent, mechanical Q increased with either increasing concentration of the lithium nitrite impurity or with decreasing growth rate (run nos. 2, 4-9, 14).

(2) At low concentration of the impurity, whatever be the nature of the impurity, mechanical Q increased with either increasing concentration of the basic solvent or with decreasing growth rate (run nos. 2, 3, 16-18).

(3) At high concentration of the impurity, however, mechanical Q decreased with increasing concentration of the basic solvent (run nos. 9, 10, 12-15). Exception is run no. 11.

(4) Concentrations of basic solvent and impurity remaining the same and growth rate being similar, lithium nitrite yielded better crystals than lithium carbonate (run nos. 3, 18). Lithium nitrate and lithium fluoride yielded inferior quality crystals (run nos. 19, 20).

(5) For the same concentration of basic solvent and same nature and concentration of the impurity, quality of the crystals obtained were better for low growth rates (run nos. 4-9). The different growth rates obtained in those runs were probably due to short-range fluctuations of the experimental variables which could not be recorded.

(6) For the 6" long crystals (synthesized in an 8" long growth zone) mechanical Q of both top and bottom (with respect to location in the growth zone of the liner) portions were measured (run nos. 8, 9, 11, 15 and 20). A very wide range of Q was obtained in the case of run no. 9. However, growth rates of both the basal and X directions of this crystal for the top and bottom portions were identical. This is rather unusual in view of the fact that incorporation of crystal gives rise to imperfections and therefore should normally increase the impurities in the growth rate.

--- explanation of this feature can perhaps be put forward. The growth zone temperature recorded in those runs was an average of the temperatures of the top and bottom of the growth zone. Normally the temperature at the top of

the growth zone is lower than that of the bottom of the growth zone, and the reverse is true for temperature gradient. Those two effects have probably compensated for each other in this particular case, rather fortuitously it can be imagined, to give rise to the same growth rate for the top and bottom portions.

Regarding the nature of impurities present in the synthetic quartz crystals, it would not be possible to make any generalised comments, because different investigators might be using different types of solvents and dopants, and liners made of materials other than mild steel. In our case OH^- seems to be the common impurity and Fe^{3+} is also likely to be present.

The general conclusions that can be arrived at from this study is that low growth rate, resulting from very fine control of the experimental variables, mainly temperature, its range of fluctuations and moderate temperature gradient, certainly yields better crystals, and that certain optimum concentrations of basic solvent and impurity selected are also helpful in improving the quality of the crystals.

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