April 6, 1954 A. R. SOBEK 2,674,520

APPARATUS FOR GROWING SINGLE CRYSTALS OF QUARTZ

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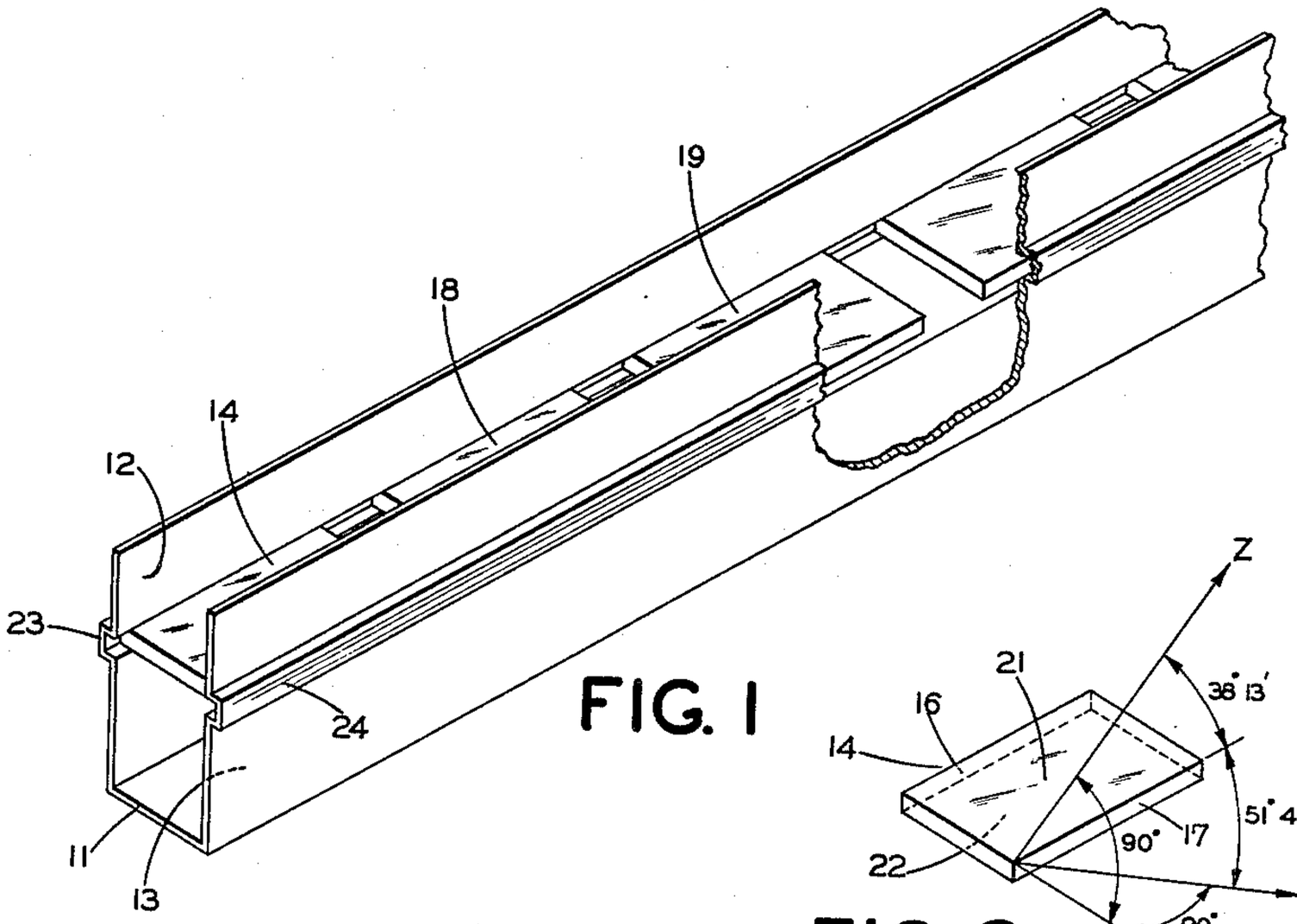

FIG. 1

FIG. 2

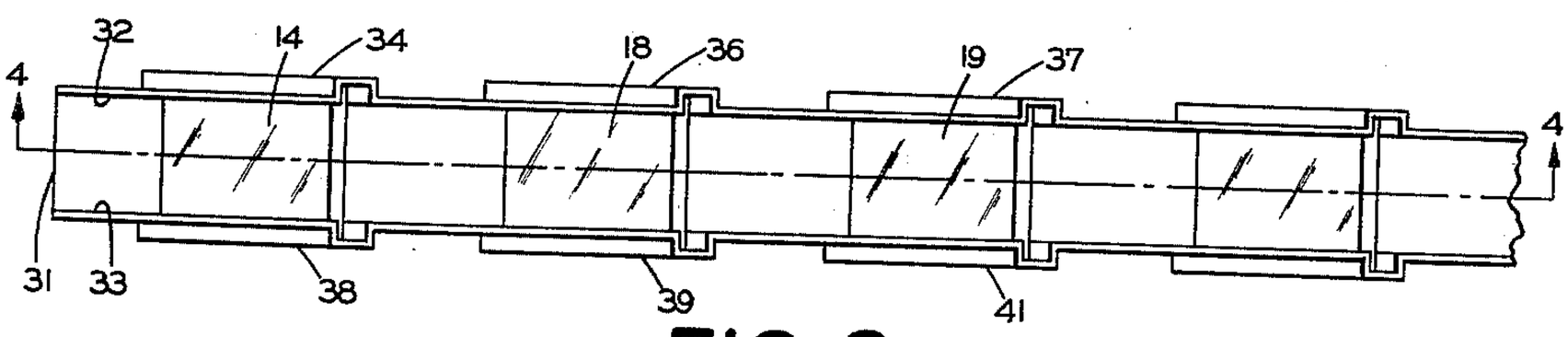

FIG. 3

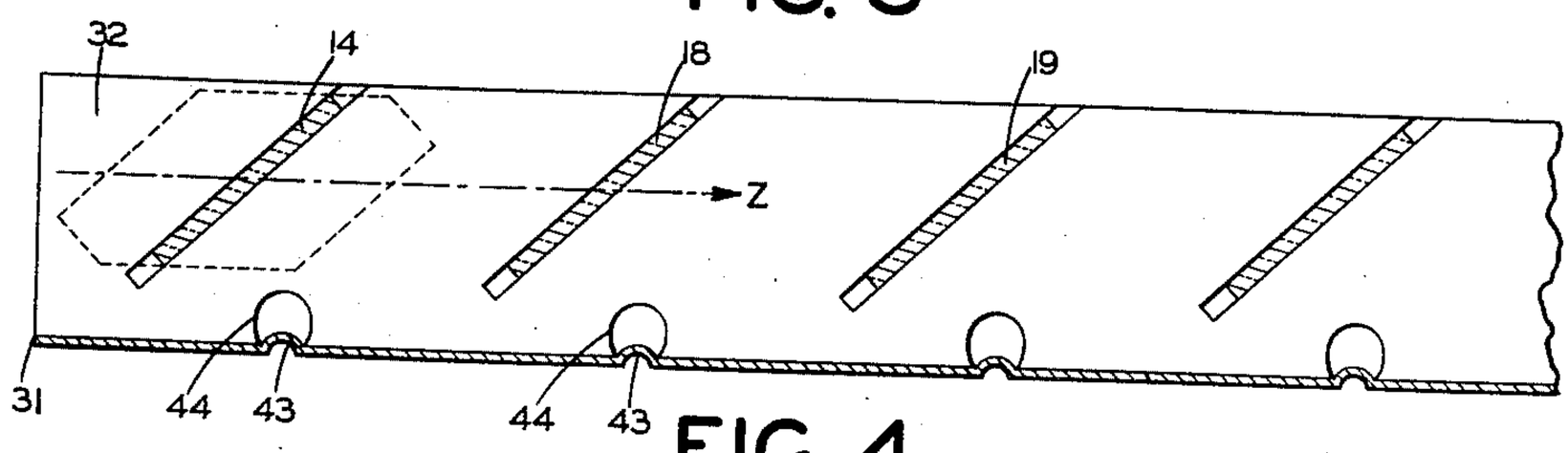

FIG. 4

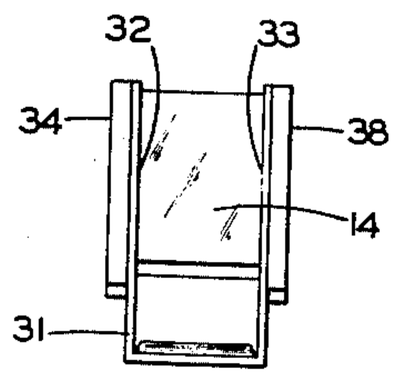

FIG. 5

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APPARATUS FOR GROWING SINGLE CRYSTALS OF QUARTZ

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Application April 11, 1950, Serial No. 155,222

3 Claims. (Cl. 23—273)

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This invention relates to an improved crystal seed arrangement, and more particularly to such an arrangement for incorporation in quartz-growing apparatus.

The synthesis of sizable single crystals of quartz has been attempted by many laboratory investigators during the past fifty years or more. In general, the apparatus and methods used have resulted in only very slow growth of quartz, the crystals produced usually being very tiny or badly flawed. Quite recently, however, procedures have been devised which make possible the synthesis of sizable single crystals of quartz, so that the crystals so produced may well be expected to find use in competition with the naturally occurring quartz of commerce, especially in times of unusually high demand for quartz of electrical grade. Such processes for growing single crystals of quartz are described and claimed in applications Ser. Nos. 94,682 and 94,683 for Letters Patent of the United States, filed May 21, 1949, in the names of Danforth R. Hale and Andrew R. Sobek and assigned to the same assignee as the present invention.

As procedural steps have been developed which are suitable for the efficient synthesis of large and flawless single crystals of quartz, apparatus has been devised which is particularly well adapted for producing quartz. Apparatus for growing single crystals of quartz which has proved quite successful in practice is described and illustrated in application Serial No. 155,221 for Letters Patent of the United States, filed concurrently herewith in the names of Andrew R. Sobek and Danforth R. Hale and assigned to the same assignee as the present invention. As illustrated in a preferred form in the drawings of the application just referred to, this apparatus takes the form of two elongated pressure chambers disposed parallel to each other and interconnected near both end portions of the chambers by two cross pipes. One of these chambers contains a siliceous supply material. The entire system contains a fluid which may be made to dissolve silica from the supply material and to transport the dissolved silica to the other chamber, which may be maintained somewhat cooler so that the fluid becomes supersaturated with respect to silica. With apparatus of this type practically the entire interior volume of this latter chamber may be used for the deposition of quartz on quartz crystal seeds. The present application is directed to crystal seed arrangements suitable for use, for example, in the quartz-growing apparatus described and broadly claimed in the last-mentioned concurrently filed patent application.

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If a quartz crystal seed is suspended freely in the growing region or chamber of a quartz synthesis apparatus, several undesirable results may ensue. Of these perhaps the most obvious is that the crystal, before or during the growing operation, may move, become dislodged, or otherwise shift its position so as to be too close to obstructions, especially after a considerable increase in the size of the crystal. The chemical, physical, or mechanical forces acting upon the crystal may even dislodge it completely from the regions of the pressure vessel in which orderly growth of the crystal can occur. Consequently the crystal seed should be held securely enough so that it will not shift or rotate, even if its dimensions should decrease somewhat during the initial or warm-up stage of the crystal-growing operation.

Another difficulty often experienced during attempts to synthesize quartz results from the tendency of the crystal seed to grow in undesirable directions relative to the shape of the seed or to the crystallographic axes of the quartz substance. Crystal growth in such undesirable directions may tend to be flawed or may produce quartz having a shape and size which do not lend themselves to the efficient production of crystal plates or bars suitable for piezoelectric and electrical circuit applications.

Furthermore, only limited space is available for the deposition and growth of the crystal seeds within the quartz-growing apparatus. If the crystal seed arrangement is not planned to make economical use of the available space, especially in view of the direction and extent of the expected crystal growth, the growing process will be very uneconomical.

Accordingly, it is an object of the present invention to provide a new and improved crystal seed arrangement for quartz-growing apparatus which substantially avoids one or more of the limitations and disadvantages of the type described.

It is another object of the invention to provide a new and improved crystal seed arrangement for quartz-growing apparatus which prevents growth of the seed in undesirable directions.

It is a further object of the invention to provide a new and improved crystal seed arrangement for quartz-growing apparatus in which one or more quartz crystal seeds may be securely disposed and oriented within a quartz-growing chamber so as to make efficient use of the space available within the chamber.

In accordance with the invention, apparatus for growing single crystals of quartz comprises a pressure vessel, adapted to contain a siliceous

fluid, and a seed-holding structure, supported within the vessel, having two extensive spaced opposed surfaces with opposed indentations in each of these two opposed surfaces, these indentations being adapted to engage individually the two opposite edge portions of at least one quartz seed plate and to support these edge portions in proximity to the respective aforementioned opposed surfaces; furthermore, the seed-holding structure of this same apparatus, in accordance with one feature of the invention, is of sheet metal, generally U-shaped in cross section, with the legs of the U forming the two longitudinally extensive opposed surfaces.

In accordance with another feature of the invention, the holding structure of the quartz-growing apparatus, being supported within a longitudinally extensive pressure vessel with the two spaced opposed surfaces extending in the longitudinal direction of the vessel, has a series of elongated protuberances arranged in longitudinal succession along each of the spaced surfaces in a plurality of opposed pairs with each of the elongated protuberances oriented diagonally of the longitudinal direction to support each of a corresponding plurality of quartz seed plates in diagonally offset relationship between the opposed surfaces and to retain the edge portions of the plates in proximity to the respective opposed surfaces.

In accordance with another feature of the invention, the apparatus comprises an arrangement for holding at least one quartz plate of predetermined dimensions disposed transversely between the two opposed surfaces, and this arrangement not only includes protuberances individually in the opposed surfaces for engaging individually the two edge portions of the plate but also includes a frame for positioning the surfaces in opposed relationship and under elastic bias toward each other when the seed plate is disposed therebetween.

The expression "protuberances in the opposed surfaces," as used in the specification and in the appended claims, is not limited to bulges on one of the opposed surfaces extending toward the other such surface, but is intended to signify any groove, channel, rabbet, dimple, bulge, lug-shaped portion, small bracket portion, or any similar treatment of the surfaces of the holder, involving a protrusion in one of the surfaces either toward or away from the other opposed surface, or even simply a cut-out portion of the surface, provided that the protuberance is adapted to engage the seed plate or plates in the required manner.

For a better understanding of the present invention, together with other and further objects thereof, reference is had to the following description taken in connection with the accompanying drawing, and its scope will be pointed out in the appended claims.

Fig. 1 is a perspective view, partially cut away for clarity of illustration, of a crystal seed arrangement embodying the present invention;

Fig. 2 is a view in the same perspective of one of the crystal seeds;

Fig. 3 is a plan view of another crystal seed arrangement embodying the invention;

Fig. 4 is a sectional elevation taken in the direction indicated 4, 4 in Fig. 2; and

Fig. 5 is an end elevation of the arrangement shown in Figs. 3 and 4.

Referring now to Fig. 1 of the drawings, there is illustrated in perspective a crystal seed-holding arrangement for quartz-growing apparatus of the type shown in Figs. 1–3 of the drawings in the above-mentioned concurrently filed application of Andrew R. Sobek and Danforth R. Hale. As noted hereinabove and exemplified in the apparatus described in this concurrently filed application, the proper conditions for growing single crystals of quartz in such apparatus are provided within a longitudinally extensive pressure vessel adapted to contain a siliceous fluid. The crystal seed arrangement utilizes a metallic seed-holding structure 11 which may be supported conveniently within such a vessel. A variety of materials may be used for the seed holder, but the material used should be capable of withstanding alkaline fluids at elevated temperatures. A holder made of sheet metal, generally U-shaped in cross section, is desirable, with the legs of the U forming two longitudinally extensive spaced opposed surfaces 12 and 13 which are arranged to extend, of course, in the longitudinal direction of any suitable elongated pressure vessel. These surfaces should be metallic silver surfaces. The entire holder may be made of silver, or thin sheet steel may be used. However, metallic silver surfaces have been found desirable, since the silver does not stick to or interact with the quartz material. A smooth or polished metallic silver surface also tends to prevent the formation of unwanted quartz nuclei or growth centers on the exposed surfaces during the quartz-growing operation. Accordingly if a stainless or other sheet steel is used the surfaces of the sheet should be well silvered.

This arrangement is adapted to engage and hold at least one quartz seed plate 14 disposed transversely between the opposed surfaces 12 and 13. The plate 14 is shown in Fig. 2 without the seed holder 11, but seen in the same perspective as in Fig. 1. The seed plate has two opposite edge portions 16, 17. Preferably a plurality of quartz seed plates 14, 18, 19, etc. are used to provide a large area on which quartz may be grown in one continuous operation.

The seed plates are cut from unflawed natural or synthetic single crystals of quartz. Generally speaking, suitable seed plates have major surfaces 21 and 22 the normals to which are at least roughly perpendicular to an X-axis of the crystalline substance. As indicated in Fig. 2, the two shorter edge portions of the rectangular plate 14 extend in the direction of an X-axis, while the other two opposite edge portions 16, 17 preferably extend in directions which are at least roughly perpendicular to the X-axis. Preferably the major surfaces of the seed plate are roughly parallel to a major or minor rhombohedral surface of the idealized or fully grown quartz crystal. As shown in Fig. 2, one of the Y-axes of the crystalline substance, at right angles to the last-mentioned X-axis, is illustrated, and the direction of the Z-axis, perpendicular to the XY-plane, also is given. The major surfaces 21, 22 of the plate 14 may be chosen parallel to a minor rhombohedral face of the mother crystal, in which case the angle between the Z-axis and the edge 17 in the YZ-plane is 38° 13′, as illustrated in Fig. 2. This quartz plate is very similar to the so-called CT-cut quartz plate, in which the indicated angle is about 37.5° instead of 38° 13′. Accordingly the plate 14 may be an approximately CT-cut quartz seed plate having two opposite edge portions 16, 17 extending in directions perpendicular to an X-axis.

The holder 11 is provided with protuberances

23 and 24 in the opposed surfaces 12 and 13 respectively. As viewed from the space between the two surfaces 12 and 13, these protuberances appear as opposed groove-like indentations in each of the two opposed surfaces. The indentations 23, 24 individually are adapted to engage the opposite edge portions 16, 17 of the plate 14 and the corresponding edge portions of each of the plates 18, 19, etc. The protuberances or grooves are rather small, so that the holder is adapted to support the edge portions of the plates in proximity to the respective opposed surfaces 12 and 13. Preferably the upper ends of the U-shaped cross section of the holder 11 are spread apart by the seed plates, which are of suitable predetermined dimensions, against the spring action of the sheet metal to cause the seed plates to be supported between the opposed surfaces of the holder 11, whereby these surfaces are maintained under elastic bias toward each other when the seed plates are disposed therebetween. Such spring action is not essential however; it may be necessary merely to slide the seed plates into the grooves 23 and 24 without pressing the surfaces 12 and 13 apart while the plates are inserted in the holder. In any case the arrangement is effective to support the plates with the necessary firmness in the holder. It will be understood that, after the holder carrying the seed plates has been inserted into the pressure vessel so as to be supported therewithin and the silica-transporting fluid has been added, the vessel is sealed and heated to elevated temperatures, as described in detail in the aforementioned concurrently field application, with deposition from the silica-transporting fluid of quartz on the seed and resultant growth of the seed into a quartz crystal; thereafter, to permit recovering the grown crystal from the holding structure, the vessel is cooled and opened and the holder removed therefrom.

In the Fig. 1 arrangement each side of the holder has only one groove engaging in longitudinal succession one of the opposite edge portions of each of the seed plates 14, 18, 19, etc. A somewhat different arrangement is illustrated in Figs. 3–5.

Fig. 3 is a top view, Fig. 4 a sectional front elevation, and Fig. 5 a side elevation of this alternative arrangement. A seed holder 31 has two opposed surfaces 32, 33. A similar plurality of quartz seed plates 14, 18, 19, etc., is provided.

As in the Fig. 1 arrangement, the holder 31 is U-shaped in cross section. However, the seeds are arranged differently within the holder. For this purpose there is formed a series of elongated, diagonally oriented protuberances arranged in longitudinal succession along each of the surfaces 32 and 33. These protuberances in the two opposed surfaces are arranged in a plurality of opposed pairs individually adapted to engage the two opposite edge portions of each of the plates successively so as to hold these edge portions in proximity to the respective surfaces. Thus the surface 32 has diagonal groove-like protuberances 34, 36, and 37 to receive one edge of each of the plates 14, 18, and 19 respectively, while the surface 33 has similar groove-like protuberances 38, 39, and 41 to receive the opposite edges of the respective plates and thus to support the seed plates in diagonally offset relationship between the opopsed surfaces of the holder 31. The desired growth of the crystals usually occurs on both sides of the seed plate in the direction of the Z-axis.

The seeds preferably should be arranged in the holder to obtain the most efficient utilization of the space within the holder during growth of the crystals. To this end the diagonal orientation of the protuberances or grooves should be chosen with respect to the crystal orientation of the seeds, as placed in the holder 31, so that the seed plates are disposed with respect to each other with the directions of the crystallographic Z-axes through the centers of each of the plates approximately coincident. This disposition of the plates is illustrated in Fig. 4 for seed plates of the type shown in Fig. 2. The directions of the Z-axes in all of the plates are coincident, as shown by the dot-dash line indicated to be the Z-axis. This line is parallel to the bottom of the seed holder 31 and ordinarily would extend in the axial direction of a cylindrical quartz-growing chamber. There also is shown in Fig. 4 in dashed lines the outlines of a grown quartz crystal formed on the seed 14.

The bottom of the seed holder 31 has a number of transverse grooves 43, 43 to compensate for the shortening of the side walls 32 and 33 caused by the grooves 34, 38, etc. This permits the use of quite thin sheet metal without a tendency of the holder tc bow outwardly along its length as viewed from beneath. Cutouts 44, 44 permit the desired shortening of the bottom of the holder by virture of the grooves 43, 43.

It will be understood that the seed holder may have numerous shapes, depending on the space available for it and on the material and method of fabrication used. The seed-holding structure having the two spaced opposed surfaces, such as the surfaces 12—13 in the Fig. 1 arrangement or the surfaces 32—33 in the arrangement of Figs. 3–5, also includes, of course, a connecting structure which serves as a frame for positioning and maintaining these surfaces in opposed relationship. This frame may be the bottom of a U-shaped holder as illustrated in Figs. 1 and 5. The two spaced opposed surfaces of the holder, however, need not be formed from one sheet member, but may be separate members held in properly spaced opposed relationship, preferably under elastic bias toward each other as discussed hereinabove, by other structures within the quartz-growing chamber.

It may be pointed out that a seed of the type shown in Fig. 2 shows little tendency to grow on the shorter edges 16 and 17, that is, perpendicularly to the X-axis in the plane of the major surfaces. However, seed holders having extensive surfaces of the types described serve to prevent growth of the seeds in a direction into either of the opposed surfaces 12, 13 or 32, 33. This effectively prohibits growth on the seeds in the general direction of the X-axis. Such growth tends to complete the prism faces characteristic of quartz crystals. Such growth is not oriented in a direction relative to the crystallographic axes which permits most effective utilization of the grown crystal when cut into plates or bars for the usual electrical and piezoelectric applications. Consequently the opposed surfaces of the seed holder should be extensive enough so that the growing crystal is retained between these surfaces during the entire growth of the seed until the grown seed has reached its maximum dimensions. Crystals grown in such seed holders have convenient shapes for efficient utilization of the space within the quartz-growing vessel.

While there have been described what are at

present considered to be the preferred embodiments of the invention, it will be obvious to those skilled in the art that various changes and modifications may be made therein without departing from the invention, and it, therefore, is aimed in the appended claims to cover all such changes and modifications as fall within the true spirit and scope of the invention.

What is claimed is:

1. Apparatus for growing single crystals of quartz, comprising: a pressure vessel adapted to contain a siliceous fluid; and a holding structure, supported within said vessel, of sheet metal, generally U-shaped in cross section, with the legs of the U forming two longitudinally extensive spaced opposed surfaces having opposed indentations in each of said two opposed surfaces, said indentations being adapted to engage individually the two opposite edge portions of at least one quartz seed plate and to support said edge portions in proximity to said respective opposed surfaces.

2. Apparatus for growing single crystals of quartz, comprising: a longitudinally extensive pressure vessel adapted to contain a siliceous fluid; and a holding structure, supported within said vessel, having two spaced opposed surfaces extending in the longitudinal direction of said vessel and having in each of said opposed surfaces a series of elongated protuberances arranged in longitudinal succession along said surfaces with each of said elongated protuberances oriented diagonally of said longitudinal direction, said protuberances in said two opposed surfaces being arranged in a plurality of opposed

pairs individually adapted to engage the two opposite edge portions of each of a corresponding plurality of quartz seed plates to support said seed plates in diagonally offset relationship between said opposed surfaces and to retain said edge portions of said plates in proximity to said respective longitudinally extending opposed surfaces.

3. Apparatus for growing single crystals of quartz, comprising: a pressure vessel adapted to contain a siliceous fluid; and a seed-holding structure, supported within said vessel, having two extensive spaced opposed surfaces, and having an arrangement for holding at least one quartz seed plate of predetermined dimensions disposed transversely between said opposed surfaces, said arrangement including protuberances individually in said opposed surfaces for engaging individually two opposite edge portions of said seed plate in proximity to said respective opposed surfaces and including a frame for positioning said surfaces in opposed relationship and under elastic bias toward each other when said seed plate is disposed therebetween.

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