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⑤④ **Process for irradiating topaz and the product resulting therefrom.**

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## Description

### BACKGROUND OF THE INVENTION

#### Field of the Invention

This invention relates to a method for processing topaz by the combined use of high energy neutron radiation and electron bombardment to produce a novel and superior product and to the novel product so produced by this unique processing method.

More particularly, this invention relates to the processing of colorless or pale colored topaz to produce a new product of a different color, particularly of the moderately deeper blue, which I describe as "American Blue" which color is of better quality and greater value than the original topaz gemstone. A slightly modified method may be used to produce a lighter blue color than the "American Blue" which I describe as "Super Sky Blue".

#### Description of the Prior Art

While it is well known to those skilled in the nuclear art that the bombardment of various gems, glasses and plastics by sub-atomic particles often produces changes in color as well as in other properties of these materials, neither the reason nor the mechanism of the process are comprehensively understood. Except in a few well known cases, the results of such subatomic particle bombardment are neither predictable nor obvious for any given material or type of radiation.

For example, it is known in the prior art that ordinary glass loses its transparency when subjected to gamma radiation. Pyrex, a type of glass, becomes amber and quartz becomes purple. Without special treatment, these colors gradually disappear if the glass is allowed to stand for a prolonged period or is subject to the action of intense light rays. This particular property has been used to detect radiation by noting the darkening of specially treated glass when exposed to further radiation as described by McAlpine and Rinehart in U.S. Patent No. 2,782,319. On the other hand, various types of glasses have been developed also which resist the color changing effect of radiation such as disclosed in U.S. Patent No. 2,747,105 which was issued to Fitzgerald, et al.

Likewise, the color and transparency of plastics have been known to be effected by nuclear irradiation as detailed in U.S. Patent No. 2,855,517 issued to Rainer, et al.

The phenomenon is frequently described as being caused by the displacement of electrons from one part of the material to another resulting in the entrapment of some of the electrons in the color or "F" center of the material thereby changing its isotropy and consequently its color. (Stephenson, "Introduction to Nuclear Engineering," pp. 222, 256, and 350).

In the instant invention, I have discovered that by taking topaz gemstones which are colorless or

pale-colored and by properly subjecting to the combined two step action of neutron bombardment, and then by electron bombardment, that I am able to permanently and completely change their color without impairing their other qualities.

Further, I have discovered also that by taking topaz gemstones which are colorless or pale-colored and subjecting them first to high energy neutron bombardment and then bombarding them with high energy electrons I can produce topaz gemstones of a highly desirable, predictable, and permanent color.

As is well known, Topaz is a natural aluminum fluorosilicate which can be written in the formula,  $[Al F SiO]$ . It commonly occurs in high temperature veins probably formed in igneous intrusions in the presence of fluoride and water vapor. As a result, some of the fluoride is usually replaced by hydroxyl, giving it the variable composition,  $[Al (F,OH) SiO]$ .

Topaz occurs naturally in a wide range of colors (blue, yellow, green, orange, violet, pink and the highly desirable gold to sherry), but it is most commonly colorless.

The crystalline structure of topaz was determined independently by Pauling (1928) and by Alston and West (1928). It was described as belonging to the orthorhombic centrosymmetric space group  $pbnm$  and the structure has been successfully refined in that space group (Ribbe and Gibbs, 1971).

An important optical property of many gemstones including topaz is that known as dichroism or pleochroism. A stone possessing this property, when observed in different directions will show different colors or shades of color which may resemble each other more or less closely, or may differ considerably. The pleochroism of natural topaz is distinct but not strong.

The production of blue topaz from colorless stones by irradiation was first reported by F. H. Pough in 1957 as one of a large number of color changes observed in a variety of materials subjected to radiation treatment. The process was rediscovered and reported by Nassau in 1974 and by Nassau and Prescott in 1975.

Many people in the gemstone business believed the reporting of irradiation treatment to produce blue topaz provided an explanation for the large increase in the quantity of blue topaz available at that time. During the last ten years, more than a million carats of blue topaz have entered the world market.

Several types of irradiation can be used to alter the color of topaz including X and gamma rays, high energy charged particles, and neutrons. To date, however, the only generally useful forms of ionizing radiation include gamma rays from Co-60, high energy electrons from accelerators, and neutrons from nuclear reactors.

One of the principal qualities which determines the commercial value of a topaz stone is its color. Naturally occurring blue topaz stones are pale in color and their value in the market is therefore limited.

Only a small percentage of the topaz stones which have been irradiated by electrons will be a sufficiently intense blue color to be marketable. Therefore, the cost of producing the stones that will

eventually be sold is increased by the cost for producing stones that are not salable.

When colorless or pale colored topaz stones are irradiated with neutrons, the pleochroism which results in different colors being seen when looking along different axes frequently results in an undesirable gray or "inky" appearance to many of the stones. The value of these stones is much less than the top quality blue gemstones.

When a colorless or pale-colored topaz is exposed to gamma rays, several colors of the yellow to reddish-brown to brown tones develop in some of the stones at relatively low radiation doses, that is, doses of less than one megarad. As the dose is increased, a green-yellow to green blue color will develop in some stones, depending on the nature of the particular stone. If these particular stones are subsequently heat treated, a blue color can be brought out in the stone.

When by the nature of some stones an intense blue color is produced, it is generally a gray or "steely" blue.

In order for electrons to penetrate a significant thickness of topaz, energies in the range of 10 to 20 million electron-volts are required. To obtain these energies, it is necessary to accelerate the electrons in any one of a variety of machines including linear accelerators, Van de Graaff generators, and betatrons. A significant advantage in the use of electron accelerators is that the dose rates available are much higher than with gamma ray sources, enabling sufficient exposures to be given to the stones in only a few hours instead of several months. Another advantage is that the gray blue color produced by gamma rays and neutrons does not generally occur. Instead, in those stones which possess the needed natural property to turn blue, a very clear "sky" blue color is produced.

Neutrons can be produced by a variety of means, but the most practical method to generate the neutron intensities required to color meaningful quantities of topaz in a reasonable time is to use a nuclear reactor.

In general, all colorless or pale colored topaz will turn a blue color if given sufficient exposure to high energy neutrons. The color can be made darker than most intense color from either gamma rays or electrons. The pleochroism of topaz is very evident in neutron irradiated stones with the color axis appearing dark blue, light blue, and gray. This property results in a wide range of colors depending on the orientation of the crystalline axis with respect to the stone and the type of cutting used relative to the particular stone.

#### SUMMARY OF THE INVENTION AND OBJECTS

This invention relates to a process for enhancing the color of colorless and pale-colored topaz crystals by novel techniques and to the superior products so produced.

The present invention relates to a process for the irradiation of topaz and the product produced thereby wherein the colorless or pale-color topaz is first irradiated in a neutron environment at an expo-

sure level sufficient to produce some blue, brown, yellow or green color or a steely, dark blue or gray color therein. The method is characterized in that the neutron bombarded topaz is irradiated with electrons at an exposure level from 1 000 to 10 000 megarads to produce the desired blue color. Occasionally, some of the topaz stones may require heating in order to obtain the desired blue color; however, in most cases, the heating step is not necessary.

#### DESCRIPTION OF THE PREFERRED EMBODIMENT

Fundamentally, this invention relates to a new and novel method for processing topaz by the combined use of high energy neutron radiation and electron bombardment to produce a novel and superior topaz and to the particular topaz produced by this unique processing method.

More particularly, this invention relates to the processing of colorless or pale colored topaz to produce a new product of a different color, particularly of the moderately deeper blue, which I describe as "American Blue". American Blue topaz is of a better color, better quality and is of greater value than the original topaz gemstone. A variation of this process can be used to produce a topaz gemstone which has a very definitely enhanced blue coloration which I prefer to call a "Super Sky" blue. The "Super Sky" blue is a color which falls between the "Sky" blue and the "American" blue.

The purpose of such a process is to enhance the value of the relatively "normal" sky-blue topaz gem by moderately deepening the blueness of the color of the topaz gemstone to a relatively "rare" and more desirable blue color.

#### METHOD FOR PRODUCING SUPER SKY BLUE TOPAZ

My process for improving the color and quality of the topaz product to produce a "Super Sky" blue colored topaz includes the following steps:

First, the colorless or pale colored topaz gemstone is immersed in a neutron environment to irradiate the topaz at an exposure level sufficient to produce some color in the topaz so that when the topaz is subsequently exposed to electron bombardment in the range of approximately 1,000 to 10,000 megarads a blue color is obtained in the topaz. In most cases, the neutron bombardment of the topaz will produce a light blue color. However, it is not unusual for the topaz to be changed from its original colorless, or near colorless, state, to a brown, yellow, green, or blue color, or varying shades of these, and other, color combinations. The level of neutron irradiation to produce the light blue color will, of course, depend on a number of factors, including, just to name a few of the determinative factors involved, the amount of topaz being irradiated, the duration of the radiation exposure, the specific nature and character of the particular topaz being bombarded, and the relative sizes of the topaz gemstones. One of the key and most important elements is that the topaz be treated with a sufficient level of neutron radiation

bombardment. Such a degree and amount of neutron radiation bombardment is achieved when some color is induced in the topaz. Of course, it has been found that such level of neutron radiation bombardment of the topaz is preferred to be when the stone is turned to a medium blue color.

Second, the neutron irradiated medium blue colored topaz is then irradiated with electrons at an exposure level in the range of from approximately 1,000 to 10,000 megarads to produce a remarkable and enhanced "Super Sky" blue colored topaz gemstone.

Occasionally, some of the neutron and electron irradiated topaz gemstones due to the particular nature and characteristics of the material forming the topaz, will not obtain the desired "Super Sky" blue coloration, but, instead, will become greenish-blue or greenish-yellowish. When this occurs, the desired "Super Sky" blue color can be obtained by further treating the topaz by heating the topaz for a few hours, that is, from one to six hours, at between 250 to 600 degrees Fahrenheit (121°C to 316°C). By heating, I find that nearly all of the topaz stones heat treated in this manner will have the very desirable color and are highly marketable.

#### METHOD FOR PRODUCING AMERICAN BLUE TOPAZ

The process which I have discovered for improving the color and quality of the topaz product to produce an "American" blue colored topaz includes the following steps:

First, the colorless or pale colored topaz gemstone is immersed in a neutron environment to irradiate the topaz at an exposure level sufficient to produce some color in the topaz. Sometimes, this involves the production of a steely, or dark blue, or gray color. The level of neutron irradiation to produce the blue color will, of course, depend on a number of factors, including, just to name a few of the determinative factors involved, the amount of topaz being irradiated, the duration of the radiation exposure, the specific nature and character of the particular topaz being bombarded, and the relative sizes of the topaz gemstones. One of the key and most important elements is that the topaz be treated with a sufficient level of neutron radiation bombardment. Such a degree and amount of neutron radiation bombardment is achieved when some color is induced in the topaz. Of course, it has been found that such level of neutron radiation bombardment of the topaz occurs when some color appears in the topaz. In some cases, this level is achieved when the topaz gemstone is turned a steely or gray color.

Second, the neutron irradiated dark blue colored topaz is then irradiated with electrons at an exposure level in the range of from approximately 1,000 to 10,000 megarads to produce a remarkable and enhanced "American" blue colored topaz gemstone. "American" blue is a new, extremely bright, intense blue color.

Occasionally, as in the previous method which I have described to produce a "Super Sky" blue topaz, some of the neutron and electron irradiated to-

paz gemstones because of the particular nature and characteristics of the material forming the topaz, will not obtain the desired "American" blue coloration, but, instead, will become brownish or yellowish. When this occurs, the desired "American" blue color can be obtained by further treating the topaz by heating it for a few, that is, two or more hours, at about 250 to 900 degrees Fahrenheit (121°C to 482°C). As a practical matter, the preferred time period for heating the topaz is from one to six hours. Further, it should be clearly understood and noted that heating of the topaz to obtain the desired final color change may be accomplished at lower temperatures. Such lower temperatures are from about 250 degrees Fahrenheit, but, this, of course, requires a longer period of heating than is accomplished at higher temperatures. Also, if the temperature is raised to approximately 900 degrees Fahrenheit, the desired color change can be accomplished in a shorter time frame. Through this process of heating, I find that nearly all of the topaz stones heat treated in this manner will have the very desirable color and are highly marketable.

It should be noted that the within described process and product is intended to include all variations in composition of topaz from any and all locations in the world and all forms of topaz including, but not limited to, rough and preformed stones, all manner of carved stones, and all manner of polished and cut stones. The neutron irradiation described includes treatment by neutrons of all energies produced by any means including but not limited to isotopic sources, accelerator sources, and nuclear reactors. The electron irradiation includes irradiation by electrons produced by any technique and accelerated by any method.

It should be clearly understood that certain obvious modifications will occur to those skilled in the art to which this invention pertains. However, such obvious modifications are intended to be within the scope and purview of the present invention herein, and the outer boundaries of the scope of the instant invention are intended to be limited and determined only by the scope of the claims appended hereto.

#### **Claims**

1. A method for producing color change in topaz, said method including the step of irradiating colorless or pale-colored topaz in a neutron environment to an exposure level sufficient to produce some blue, brown, yellow or green color or a steely, dark blue or gray color; and characterized by the step of irradiating the neutron bombarded topaz with electrons at an exposure level of from 1 000 to 10 000 megarads.

2. The method of Claim 1, wherein said neutron radiation level produces a steely, dark blue or gray color.

3. The method of Claims 1 or 2 further characterized in that the color produced by the electron radiation is a bright blue color.

4. The method of Claim 1 wherein said neutron radiation level produces a light blue color and when the topaz is subsequently irradiated with electrons

from 1 000 to 10 000 megarads a lesser blue color than a bright blue color will be obtained.

5. The method of Claim 1, 2, 3 or 4 further characterized by the step of heating the topaz to a temperature sufficient to produce the desired color change in the topaz.

6. The method of Claim 5 wherein said temperature is at least 250 degrees Fahrenheit (121°C).

7. The method of Claim 5 wherein said temperature is from about 250 to 900 degrees Fahrenheit (121°C to 482°C).

8. The method of Claim 5, 6 or 7 wherein said temperature is maintained at said temperature from a period of about one to six hours.

9. The method of Claim 5, 6 or 7 wherein the topaz is maintained at said temperature for a period of at least two hours.

10. A bright blue topaz produced by the method of any one of the preceding claims.

#### Patentansprüche

1. Verfahren zum Herbeiführen einer Farbveränderung in Topaz, mit dem Schritt, in dem ein farblos oder blassfarbener Topaz in einer Neutronenumgebung bis zu einer absorbierten Energiedosis bestrahlt wird, die genügt, um etwas blaue, braune, gelbe oder grüne Farbe oder eine stahlartige, dunkelblaue oder graue Farbe zu erzeugen, gekennzeichnet, durch einen Schritt, in dem der neutronenbestrahlte Topaz mit Elektronen bis zu einer absorbierten Energiedosis von 1 000 bis 10 000 Megarad bestrahlt wird.

2. Verfahren nach Anspruch 1, dadurch gekennzeichnet, dass die infolge der Neutronenbestrahlung absorbierte Energiedosis die Erzeugung einer stahlartigen, dunkelblauen oder grauen Farbe bewirkt.

3. Verfahren nach Anspruch 1 oder 2, dadurch gekennzeichnet, dass die durch Elektronenbestrahlung erzeugte Farbe eine hellblaue Farbe ist.

4. Verfahren nach Anspruch 1, dadurch gekennzeichnet, dass die infolge der Neutronenbestrahlung absorbierte Energiedosis die Erzeugung einer hellblauen Farbe bewirkt und dass bei der darauffolgenden Bestrahlung des Topazes mit Elektronen bis zu einer absorbierten Energiedosis von 1000 bis 10 000 Megarad eine blaue Farbe erzeugt wird, die heller ist als eine hellblaue Farbe.

5. Verfahren nach Anspruch 1, 2, 3 oder 4, gekennzeichnet durch einen Schritt, in dem der Topaz auf eine Temperatur erhitzt wird, die zum Herbeiführen der gewünschten Farbveränderung in dem Topaz genügt.

6. Verfahren nach Anspruch 5, dadurch gekennzeichnet, dass die Temperatur mindestens 121°C beträgt.

7. Verfahren nach Anspruch 5, dadurch gekennzeichnet, dass die genannte Temperatur zwischen 121 und 482°C liegt.

8. Verfahren nach Anspruch 5, 6 oder 7, dadurch gekennzeichnet, dass der Topaz ungefähr eine bis sechs Stunden auf der genannte Temperatur gehalten wird.

9. Verfahren nach Anspruch 5, 6 oder 7, dadurch gekennzeichnet, dass der Topaz mindestens zwei Stunden lang auf der genannten Temperatur gehalten wird.

10. Hellblauer Topaz, der nach dem Verfahren nach einem der vorhergehenden Ansprüchen erzeugt worden ist.

#### Revendications

1. Procédé permettant de produire un changement de couleur dans une topaze, ledit procédé comportant l'opération qui consiste à irradier une topaze incolore ou de couleur pâle dans un environnement neutronique jusqu'à un niveau d'exposition suffisant pour produire une certaine couleur bleue, brune, jaune ou verte ou bien une couleur acier, bleu foncé, ou gris; et caractérisé par l'opération consistant à irradier la topaze bombardée par des neutrons à l'aide d'électrons à un niveau d'exposition de 1 000 à 10 000 mégarades.

2. Procédé selon la revendication 1, où ledit niveau de rayonnement neutronique produit une couleur acier, bleu foncé ou gris.

3. Procédé selon la revendication 1 ou 2, caractérisé en outre en ce que la couleur produite par le rayonnement électronique est une couleur bleu vif.

4. Procédé selon la revendication 1, où ledit niveau de rayonnement neutronique produit une couleur bleu vif et, lorsque la topaze a ensuite été irradiée à l'aide d'électrons de 1 000 à 10 000 mégarades, il est obtenu une couleur moins bleue qu'une couleur bleu vif.

5. Procédé selon la revendication 1, 2, 3 ou 4, caractérisé en outre par l'opération consistant à chauffer la topaze jusqu'à une température suffisante pour produire le changement de couleur voulu dans la topaze.

6. Procédé selon la revendication 5, où ladite température est au moins de 250°F (121°C).

7. Procédé selon la revendication 5, où ladite température est d'environ 250 à 900°F (de 121°C à 482°C).

8. Procédé selon la revendication 5, 6 ou 7, où la température est maintenue à ladite valeur pendant une durée d'environ 1 à 6 h.

9. Procédé selon la revendication 5, 6 ou 7, où la topaze est maintenue à ladite température pendant une durée d'au moins 2 h.

10. Topaze bleu vif produite par le procédé selon l'une des revendications précédentes.

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Title PROCESS FOR IRRADIATING TOPAZ AND THE PRODUCT RESULTING THEREFROM

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