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3,075,906

## PROCESS FOR IRRADIATING PEARLS AND PRODUCT RESULTING THEREFROM

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My invention relates to a method of processing pearls by the use of high energy, ionizing radiation to produce a novel and superior product and to the novel product so produced.

More particularly my invention relates to the processing of either natural or cultured pearls to produce a new product of different color, better quality and greater value than the original pearl.

One of the principal qualities which determines the commercial value of the precious gem known as the pearl is its color. Genuine pearls, as distinguished from imitation pearls, are formed as an abnormal growth inside the shell of certain mollusks. These may result from natural causes in the normal growth of the mollusk or be produced artificially by the insertion of a nucleus into the tender tissue of the living mollusk around which the pearl is gradually formed by the deposit of nacre. The former are known as natural pearls, while the latter are known as cultured pearls. Natural pearls vary a great deal in color, and while certain colors seem to predominate in pearls found in certain parts of the world, there is no uniformity in this and no known explanation of the color variations. Likewise, with cultured pearls, there is no known way in which there may be predicted or controlled the color of the final pearl after the original insertion of the nucleus in the mollusk. No distinction between natural and cultured pearls is made for purposes of this invention both being covered by the generic terms "natural" or "commercial."

The generally recognized colors of commercial pearls are "white," "cream," "pink," "yellow," "black," "green," and "gold," with some variations and gradations of the above. Thus we may have a "pure white," a "white-cream," a "pinkish-white," a "bluish-pink," and a "bluish-gray" ("metallic-gray") pearl. These designations are generally self-descriptive and familiar to those skilled in the art. They correspond in general to the colors as usually identified with the standard color chart of the visible light spectrum.

The pearl is known to consist primarily of approximately ninety-five percent of calcareous materials, the remainder being protein and some impurities including organic matter. It has been suggested that the latter materials cause the differences in color, although there is no satisfactory accounting for this.

Generally speaking, the more exotic colors have greater commercial value if they are otherwise approximately alike with regard to size, shape, weight, lustre, symmetry, and thickness of coating. Thus certain shades of pink (or "pinkish-white") known as "rosé" are extremely valuable as are certain shades of black which are in reality a "bluish," "metallic-gray," or "gunmetal." It also seems to be the rule that the more exotic the color the more inferior the shape of the natural or cultured pearl. Thus, for example, perfectly shaped or symmetrical black pearls are quite rare, especially those of the "silver-blue" hue. In many cases these rare colors have a commercial value several times that of the more common colors.

Certain attempts have been made to artificially color white pearls by impregnating them with a coloring material or dye. These have, in general, been unsuccessful since besides the difficulty of making the changed color permanent, the coloring often injures the natural lustre or the

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hardness of the pearl and it is almost impossible to duplicate the product which comes from the mollusk. Artificially colored pearls are easily distinguished from those of natural color even by the average person with little skill in the art.

It is therefore an object of my invention to provide a method whereby either natural or cultured pearls may be effectively changed from one color to another without injuring their other desirable qualities.

It is another object of my invention to provide a method whereby a less valuable pearl may be converted to a more valuable one.

It is still another object of my invention to produce, as an article of manufacture, a new, better, more durable, and more valuable pearl.

It is a more specific object of my invention to provide a method of effectively and permanently changing the color and quality of pearls by the use of high energy, ionizing radiation.

It is known that ordinary glass loses its transparency when subjected to gamma radiation. Certain types of glass are similarly known to change color, as for example, Pyrex, which becomes amber, and quartz which becomes purple. Unless specially treated 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 has been made use of to detect radiation by noting the darkening of specially treated glass when exposed to radiation as by McAlpine and Rinehart in U.S. Patent 2,782,319. Various types of glass have been developed, also, to resist the color changing effect of radiation such as that covered by U.S. Patent 2,747,105 to Fitzgerald et al.

The color and transparency of plastics are likewise known to be effected by nuclear irradiation as taught by Rainer, et al. in U.S. Patent 2,855,517 for "Irradiation Treatment of Polyethylene."

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

While it is known, therefore, to those skilled in the nuclear art that the bombardment of materials by subatomic particles often produces changes in color as well as in other properties, neither the reason nor the mechanism of the process are well understood. The results, except in a few well known cases, such as those mentioned above, are neither predictable nor obvious for any given material or type of radiation.

I have discovered that by properly subjecting either natural or cultured pearls of one color to the action of high energy, ionizing radiation I am able to permanently and completely change their color without impairing their other qualities. I have discovered, further, that a pearl so processed cannot be distinguished from one of similar natural color even by experts, though obviously an internal change in the structure of the pearl has taken place.

I have discovered also that by taking pearls of certain given colors and subjecting them to a definite quantity and type of high-energy, ionizing radiation I can produce pearls of other desired and predictable colors as more fully described below.

I have discovered still further that the pearls so produced are equal in lustre, size, and density to the original pearl. The changes which I effect in the pearl are permanent in nature as I will discuss, also, more fully below.

While there are a number of ways in which my invention may be practiced to produce my novel product, I

will describe briefly a typical one which I have used successfully.

In one case, for my source of radiation, I use the isotope cobalt-60 having an intensity of one thousand curies of gamma rays, although those skilled in the art will realize that other sources may be used. I place the pearl to be irradiated in a suitable receptacle, which does not cut off gamma radiation, at a distance of one centimeter from the source and allow it to remain there for about twenty minutes at ordinary room temperature (about 20° C.). I then remove the pearl by any of the means familiar to those skilled in the handling of radioactive devices and materials.

Starting with a white pearl with the above process, I obtain a pearl having a beautiful "bluish-gray" color (sometimes referred to as "metallic-gray"), which is characteristic of what is known commercially as a "black" pearl. A "white-cream" colored pearl when treated as above will yield a "pink" pearl. A natural or "cultured" pink pearl will become "bluish-pink," while a "gold" pearl will become "yellowish-black." The actual variation in intensity of the shade of final color will vary with the intensity of the shade of the original pearl. Thus, the more purely "white" the original pearl is the lighter will be the resulting "bluish-gray." As the "white" becomes more "ivory," the "blue-gray" becomes darker. When "cream" predominates the resultant color becomes "pink." After a large number of trials I am now able to predict what the final color will be and am thus able to produce any desired color by proper selection of the original pearl and subjecting it to the above process.

Those skilled in the nuclear art know that cobalt-60 emits gamma rays having energies of 1.33 and 1.17 mev. making a total of 2.50 mev. per disintegration. The rate of irradiation in roentgens (r.) per hour is given by the formula

$$\frac{r.}{hr.} = \frac{6CE}{d^2}$$

where

C=the intensity of the source in curies  
E=energy of the gamma rays emitted in mev.  
d=distance from the source in feet

In the case described above, therefore, the rate of irradiation was

$$\frac{r.}{hr.} = \frac{6 \times 1,000 \times 2.50}{(0.0329)^2} = 1.39 \times 10^6$$

Since this rate was maintained, in the above example, for twenty minutes, the total amount of radiation absorbed by the pearls was:  $4.63 \times 10^5$  roentgens for the gamma in the above case.

My numerous tests have shown that the color change stabilized or became fixed after about twenty minutes of irradiation. Larger periods of irradiation up to over fourteen hours (total radiation  $2 \times 10^7$  r.) did not produce any appreciable change in the color of the pearl so that the above total radiation represents a saturation point or a fixed color condition of the process.

I have operated the above process at various temperatures up to 100° C. with no noticeable change in results. The effect of the results of changes in the physical size of the pearl is likewise not noticeable.

In order to determine the stability of the pearl in its new irradiated state, as shown by the permanency of its new color, I have conducted a great many tests.

Pearls irradiated by my process were boiled in water for several hours with no noticeable change in color. They were heated to 200° C., 300° C., and 400° C. No annealing nor color reversal was observed until at 500° C. the pearl completely disintegrated.

Pearls so irradiated were likewise cooled to the tem-

peratures of solid CO<sub>2</sub> and liquid N<sub>2</sub> with no annealing nor color change resulting.

On the contrary, identical tests performed on naturally colored pearls resulted in a considerable destruction of the natural color and lustre; in general, a dull grey and drab appearance resulting.

Long exposure to sunlight and to intense artificial ultra-violet and infra-red rays produced the same results as above.

Pearls irradiated by my process were examined under the microscope while whole, and after mechanically disintegrating them and the color was uniform throughout. They were compared with natural and cultivated pearls of the same color which had not been irradiated, both while whole and after disintegration and it was impossible to discern any difference in color properties even by pearl experts except that the color of irradiated pearls was more uniform than that of natural pearls.

In every case the irradiated pearl was equal to the original in lustre. There was no noticeable change in size or density, but the irradiated pearl appeared to be harder than the natural pearl.

It appears that the unusual and unexpected results obtained by my invention may be due to the effect of the irradiation upon the color-forming impurities which are combined with the calcareous base material of the pearl, but since the nature of the former is not well understood, a thorough explanation of what transpires is not possible.

I have, however, definitely established that by following the procedures which I have discovered and disclosed herein, any person of ordinary skill can obtain the same results which I have.

While I have described here one method of processing pearls by means of high energy, ionizing radiation involving the use of cobalt-60, it is possible to use a number of other radioactive isotopes and fission by-products also, as well as charged particle accelerators and irradiation facilities of nuclear reactors. Electrons and other subatomic particles may be utilized as well as gamma rays. Many variations of my invention will be apparent to those skilled in the art and I do not wish to limit myself to the method described herein except insofar as I do so in the claims.

I claim:

1. The process which comprises irradiating pearls with high energy, ionizing radiation to a dosage of at least  $4.5 \times 10^5$  r. until a permanently fixed change in color of said pearls is effected.

2. The process of claim 1 in which the ionizing radiation comprises charged particles.

3. The process of claim 1 in which the ionizing radiation comprises gamma rays.

4. The process of claim 1 in which the ionizing radiation comprises electrons.

5. The process of treating pearls which comprises irradiating said pearls with high energy, ionizing radiation to a dosage of at least  $4 \times 10^5$  r. whereby pearls of improved color are obtained.

6. A process of treating pearls which comprises irradiating said pearls with gamma rays having an energy of about 2.5 mev. to a dosage of at least  $4 \times 10^5$  r. whereby a change in color of said pearls is obtained.

7. A process of treating pearls which comprises irradiating said pearls with electrons to a dosage of at least  $4 \times 10^5$  r. whereby a change in color of said pearls is obtained.

8. As a new article of manufacture, an improved pearl product comprising a pearl which has been irradiated with high energy, ionizing radiation to a dosage of at least  $4.5 \times 10^5$  r. until a permanently fixed change in color is effected.

9. As a new article of manufacture, an improved pearl product comprising a pearl which has been irradiated with

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high energy, ionizing radiation to a dosage of over  $4 \times 10^5$  r.

10. As a new article of manufacture, an improved pearl product comprising a pearl which has been irradiated with gamma rays to a dosage of at least  $4.5 \times 10^5$  r. until a permanently fixed change in color is effected.

11. As a new article of manufacture, an improved pearl product comprising a pearl which has been irradiated with electrons to a dosage of at least  $4.5 \times 10^5$  r. until a permanently fixed change in color is effected.

12. As a new article of manufacture, an improved pearl product comprising a pearl which has been irradiated with charged particles to a dosage of at least  $4.5 \times 10^5$  r. until a permanently fixed change in color is effected.

13. The process which comprises subjecting a pearl to high energy, ionizing radiation to a dosage of at least  $4.5 \times 10^5$  r. whereby a change in the atomic structure of said pearl is effected, said change in structure being characterized inter alia by a uniform and permanent change in color throughout said pearl, said color being further characterized by an improved resistance to change under the action of extreme temperatures and the action of ultra-violet and infra-red rays.

14. The process of claim 13 in which a pearl having a natural white color is converted to a pearl which is substantially black in color.

15. The process of claim 13 in which a pearl having a natural white-cream color is converted to a pearl which is pink in color.

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16. The process of claim 13 in which a pearl having a natural pink color is converted to a pearl which is bluish-pink in color.

17. The process of claim 13 in which a pearl having a natural gold color is converted to a pearl which is yellowish-black in color.

18. As a new article of manufacture the black gem resulting from the high energy, ionizing irradiation of a naturally white pearl to a dosage of at least  $4.5 \times 10^5$  r.

19. As a new article of manufacture the pink gem resulting from the high energy, ionizing irradiation of a naturally white-cream colored pearl to a dosage of at least  $4.5 \times 10^5$  r.

20. As a new article of manufacture the bluish-pink gem resulting from the high energy, ionizing irradiation of a naturally pink colored pearl to a dosage of at least  $4.5 \times 10^5$  r.

21. As a new article of manufacture the yellowish-black gem resulting from the high energy, ionizing irradiation of a naturally gold colored pearl to a dosage of at least  $4.5 \times 10^5$  r.

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