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RADIOACTIVE PHOTOGRAPHIC ARTICLES

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This invention relates to broadly novel improvements and articles of the photographic arts; more particularly, to radioactive photographic articles and materials, to methods and means for the production thereof, and the like.

It is an object of this invention to make available photographic articles which are characterized as radioactive; for example, photographic materials and emulsions which are radioactive in direct proportion to the reduction thereof.

It is also among the objects of the present invention to provide methods and means for the treatment of photographic media, such that after production of the latent image and its subsequent development and fixing the said image can be rendered radioactive in degree which corresponds to the density of reduction in said photographic media.

It is an object of this invention to provide methods and means for the production of radioactive images upon a variety of surfaces, the said images representing for example the chemical and/or photochemical effects of such agents as light, X-rays, radioactive radiations, heat magnetism, chemical vapors and the like, pressure and mechanical energy, and the like, as set forth in more detail hereinafter.

It is an object of the present invention to supply the medical, dermatologic and like arts and sciences with an improved source of therapeutic radiations; for example, a radioactive photograph source of gamma rays or electrons. In irradiation therapy, as is well known, it is important not to involve healthy, non-malignant tissue which is adjacent to cancerous or other malignant tissue; presently, the methods of irradiation are relatively ineffectual in that healthy tissue may receive harmful doses of say gamma rays when treatment is being given a skin cancer of very irregular configuration. In the present invention, for example, a suitable photograph is taken of the diseased area or growth, which may be beneath the skin in some instances, and this photograph is then made radioactive so that it in turn may be placed against the diseased area and provide an irradiation of field contour which corresponds exactly with the configuration of the area under treatment, not involving adjacent and perhaps healthy tissue. As is set forth in more detail infra, a number of modifications of this method are possible, according to the circumstance.

Other objects of the present invention are described and set forth in detail hereinafter.

Herein, the terms and expressions "photographic articles," "photographic surfaces," "photo-

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tographic media and/or materials," "image bearing surfaces and/or media," "light sensitive, image-forming materials," "emulsions," and the like will be taken in the broad, customary meanings these designations convey to those skilled in the art; such terms will be used herein to broadly designate radiation and/or other energy responsive elements upon which an image is borne or can be produced.

Thus, if photographic articles, supra, are classified on the basis of means and energies which produce the image (latent and fixed, or, non-fixed), then the following are embraced, as examples: visible light (black-and-white or color); luminescence (e. g., arising from a coating or from fluorescent matter injected into an animal (described infra)); X-rays; gamma rays; ultraviolet light; infrared radiations; beta particles and electrons; alpha particles; ions of various energies and atomic and/or molecular species; magnetism (magnetographs); electricity (electrographs); chemical vapors, fumes, gases and the like (e. g., the Russell effect); supersonic energy; hot bodies (called pyrophotography); pressure and mechanical energy (e. g., so-called barophotography), and the like.

If, on the other hand, classification of "photographic media," and the like, supra, is made on a more abbreviated and customary basis, then there is included for example, the optical emulsions (ortho-, pan-, etc.), the infrared emulsions, the ultraviolet and X-ray emulsions (which may involve double coatings), black-and-white, color-blind, color, nuclear, and the like; these, in addition, may be "still" or of the "cinema" variety, also, opaque, translucent or transparent. If a further distinction is made on the basis of carriers and surfaces, then a large number and wide variety of rigid or flexible surfaces are included, for example: paper and cardboard; plastic; wood; metal; fabric; ceramic and glass; and the like, as is well known to those skilled in the art.

Whereas visible images are preferred in the present invention, or, at least images which can be appreciated by simple means, e. g., luminescence, this broadly novel improvement is by no means to be limited to so-called "visible" images and, as described in more detail infra, may include photographic media which bear a substantially invisible radioactive image, an image that is not to be appreciated by methods and means other than those responsive to radioactive radiations, e. g., photographic emulsions, phosphor or other luminescent surfaces (e. g., zinc sulfide phosphor), and, in particular, the radiation de-

tectors such as the Geiger counter and the photomultiplier detector.

As to photographic processes and articles characterized as produced therefrom processes, it is preferred to rely upon the usual silver halide-gelatin emulsions whenever possible, as is set forth in detail in the hereinafter examples; but not to be excluded, as is also described in detail infra, are other photographic processes, examples of which may be briefly listed at this point: (1) silver halides (e. g., bromide, iodide, chloride); (2) chromium compounds (e. g., dichromates in photomechanical processes, the well-known "bichromate system"); (3) baryta processes (which includes, for example, barium halides and barium sulfate); (4) cerium and/or rare earths processes (cerium sulfate in particular); (5) cobalt processes (involving cobalt chloride as a rule); (6) copper processes (usually involving copper sulfate); (7) iron processes (involving such ferric compounds as the chloride); (8) platinum processes; (9) platinum-iron processes (in which the iron is removed by hydrochloric acid); (10) palladiotype processes (using palladium instead of platinum); (11) silver-iron processes (the so-called "kallotype process" using ferric oxalate and silver nitrate); (12) platinum metals processes (other than platinum and palladium, using instead of these metals suitable compounds of say osmium, iridium, ruthenium, rhodium); (13) the well known "salted paper processes"; (14) asphaltic or bituminous processes (using light sensitive bitumens which are precipitated by several methods, well known to those skilled in the art, from asphaltum and the like, Syrian asphalt as an example containing 52 per cent light sensitive bitumens); (15) so-called "oil processes"; (16) so-called "carbon processes"; and the like. In addition, as desired, a number of special and specific photographic processes are well known, and may be employed herein, one example being the so-termed "metal sulfide process" in which the following elements can be employed: Fe, Ni, Co, V, W, Mo, Bi, Pb, Hg, Cu and Ag.

In practically all of the afore-listed photographic processes and media produced therewith, the exposure to light or the like (supra) results in the production of the latent image which upon development is converted into the physical image, the subsequent process of fixing and washing serving to remove photosensitive material which is not involved in the formation of the image, latent and then physical. Whereas this is generally the mechanism, in a qualitative sense, that is involved, the physical image may be produced directly by the light or other energy agent, and the subsequent treatment of fixing or the like serve to preserve it and concurrently remove photosensitive or like material which is not involved, as before. Thus, in the customary silver bromide photographic material, the light exposure produces the latent image, the development results in a physical image of reduced silver, and the fixing removes extraneous silver bromide from the unreduced areas of the material, as is well known. In many other photographic processes this general scheme is also involved.

Briefly, in the present invention a photographic material or the image borne thereupon is rendered selectively and preferentially radioactive in one or more of several ways, as desired, and examples of which are set forth in more detail hereinafter, for example: (a) by the production of radioactivity in the element and/or

compound comprising the image of a developed and fixed photographic material or surface, say in the case of a silver image by bombardment with deuterons (say from a cyclotron) or with neutrons (say from an Ra—Be or Po—Be neutron source), or other elements which comprise the image with other suitable radiations and particles known to induce radioactivity; (b) by partly or completely replacing the silver or other image with suitable metal toners and/or dye and dye-like toners (so-called mordants) and then inducing radioactivity by irradiation with neutrons, deuterons, alpha particles and the like, according to the element involved and the radioactivity characteristic desired, or, by use of radioactive toners and/or mordants to partly or completely replace the silver image, e. g., radioactive gold, cobalt, and the like (described infra); (c) by partly or completely pigmenting in the well known oil-processes and/or carbon-processes with radioactive pigment or pigment subsequently made radioactive by irradiation.

Therefore, the articles produced by the present methods and means, outlined supra, may be classed as (a) those which are susceptible to induced radioactivity, this being produced by suitable irradiation; (b) those which are radioactive by virtue of employment of radioactive matter in treatment of the image or in the production of the image; and, finally, those which are susceptible to fission or are radioactive by virtue of fission, e. g., uranium, thorium, lead, thallium, bismuth and like element bearing photographic media, as described in more detail in the examples infra. When radioactive compounds are used in (c), supra, they may be naturally radioactive and/or artificially radioactive, as desired.

According to the present invention, examples and representative embodiments of which are set forth in the following examples, but which examples and embodiments being understood as subject to considerable variation and modification without violating the spirit or scope of this invention, as desired, both so-called "negatives" and so-called "positives," or any of the conventional forms thereof, e. g., reductions and enlargements, can be used. These, as described supra, may be on practically any kind of carrier, and such a carrier may be rigid or flexible, as desired; moreover, these photographic media may comprise one or more of the many specialty forms of photographic media well known to those skilled in the art, e. g., strippable photosensitive coatings and films. When choice is made of negative or positive, consideration must be given the areas of those articles which are to be rendered radioactive. In the usual case, that of the positive or print, the so-termed "highlights" will be most lacking in radioactivity, whereas the image and parts thereof which are of gradient densities will exhibit radioactivity. The converse is true of the negative, wherein the highlights will show radioactivity. Thus, the term "highlights" will herein mean the densest parts on a negative and the lightest parts on a positive or print. Therefore, it is to be understood that while the present invention may be directed to "radioactive images," these radioactive images may be positive or negative or any conventional combination thereof, as desired. Moreover, by methods well known to those skilled in the art, the present invention is tractable in certain instances where suitable photographic media are used to reversal, wherein one part of a photo material may be radioactive

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and another part may be image bearing in the customary sense of the word.

Example 1.—The present example of this invention comprises the most elementary and simple form; that is, say a silver bromide print (positive) or negative is prepared in the customary manner, by production of the latent image, its development and fixing, with especial care being taken to remove all unreduced silver or the like (other metals for example which may form the basis of photographic processes, supra) from the emulsion. In a print the image will correspond to reduced or elemental silver or the like, and vice versa in the case of the negative. The photographic material, subsequent to the production of the physical image (i. e., changing the latent image to the physical image and removing by well known photographic methods and means all unreacted substance), is then irradiated so that induced radioactivity is produced in the reduced silver or the like areas, corresponding to the image (in a positive) or just the reverse (in a negative).

In the present Example 1, and other examples herein where induced radioactivity is produced by irradiation or the like, choice of the radiation and its source and the conditions of employment will, as is well known to those skilled in the art, vary according to the element(s) involved, the characteristics of the radioactivity to be produced, and the intensity or level of radioactivity to be imparted in a given instance of application. As a preferred source of radiation, of neutrons in particular, the nuclear reactor (e. g., "graphite pile," "uranium pile," "heavy water pile" and the like) proves most convenient. The pile irradiation may be given the photographic article for a period which may range from several hours, to weeks, to months, depending upon the characteristics of the pile (e. g., the neutron flux) and the strength of the radioactivity desired in the final product. In the case of an Oak Ridge-type pile, an irradiation of from say a week to a month will produce in a silver bearing photographic article radiosilver which has a half-life of 225 days, emitting beta particles and gamma rays; the radioactivity will correspond to approximately 8 millicuries per gram of silver. Instead of pile-irradiation, to produce radioactivity by neutron reactions, other sources include the cyclotron and, in particular, mixtures of radium and beryllium or of polonium and beryllium. As desired, alpha particle-irradiation, from say radium or polonium, can be employed to produce artificial radioactivity in the silver or the like.

Radioactive photographic articles and media of practically any radioactivity characteristics, e. g., in the matter of the radiation emitted (e. g., alpha rays, beta particles, gamma rays, X-rays), the half-life (e. g., from minutes (as in radioantimony and radiotitanium) to days (as in radiobarium and radiobismuth), weeks or months (as in radiosilver and radiosulfur), to years (as in radioiron) and centuries (as in radiocarbon, radium, uranium and thorium), and the intensity depending upon the atomic species, the nuclear reactions produced, and the amount of reactant (say the size of the photographic article), can be produced by use of photographic articles and media suitably chosen from a consideration of the element or elements which comprise the photosensitive member of the system or which make-up the image or the highlights. Other photographic

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processes than those involving silver halides are listed hereinbefore and, for example, involve alkaline earth elements, rare earths, and light and heavy metals, all of which are tractable to irradiation by neutrons, deuterons, alpha particles, and the like, the consequence being radioisotopes more or less characteristic of the atomic reactant. Variation of the photoreactant may be made as desired, and any such variation will not be construed as violating the present invention.

Example 2.—The present example is like Example 1, supra, in that the photographic article is prepared as before, but is characterized as fissionable; that is, the induced radioactivity arises from the process of atomic fission, and the isotopic debris and the like may be said to account for the resulting radioactivity. According to the present form hereof the photographic article is prepared by a photographic process, examples of which are given supra, which results in an image (or highlights or the like, as explained hereinbefore) comprising a fissionable element. As examples of fissionable elements, in addition to those of atomic numbers above 90 and including transuranium elements, the elements covering the range of atomic numbers 83 (bismuth) to 73 (tantalum) are included.

These elements (numbers 83 to 73) respond by spallation to such projectiles as alpha particles, deuterons and neutrons of energies up to 400, 200, and 100 m. e. v., respectively. Such radiations are preferred as originating in a frequency modulated cyclotron, e. g., the 184-inch Berkeley cyclotron, or other convenient and well known sources. In the case of platinum, alpha particles of say 400 m. e. v. (million electron volts) produce spallation and this is especially desirable in the case of platinum prints, platinotypes and like photographic articles. In the case of thallium, both deuterons and alpha particles are effective. The spallation reaction of these lighter elements with high energy particles obtained in the cyclotron differs in some important respects from that resulting from the fission of such elements as uranium, thorium, plutonium, protoactinium and the like, when slow or fast neutrons are employed. There is, for example, the absence of a chain reaction that characterizes high energy fission, but the yield is high and a more even split is produced in spallation.

Example 3.—In contrast to the previous Examples 1 and 2, wherein a physical image is produced in a photographic article and then by suitable methods and means rendered radioactive, generally without greatly altering the physical image in the usual sense, the present Examples 3 and 4, for example, rely upon modification of the physical image; e. g., partial or complete replacement or chemical alteration of the say reduced silver, as an example, which may be said to comprise the physical image.

Briefly, the present example, as well as others of a like nature, may be said to embrace "radioactive toning," the purely photographic or chemical process, as contrasted to staining, wherein the image of a photographic article is partly or fully replaced or modified by said (a) certain radioactive compounds and/or (b) certain substances which by suitable irradiation (cf. Examples 1 and 2) are rendered radioactive in the desired quality and quantity. In the silver bromide print, for example, toning is directed to changing the nature of the reduced silver and, as is well known, this general method of changing the color of a photographic article,

may be directed to inorganic salt replacement of the silver, or chemically changing the silver (for example) so that it can be mordanted or at least in a chemical sense with basic dyestuffs or, in general for present purposes, basic compounds, usually of an organic character. Radioactive toning and radioactive mordanting may cause not the slightest color or physical change, at least insofar as appearance to the eye is concerned.

The methods of toning are well known to those skilled in the art, and vary according to the toning agent and the photographic material, so detailed instruction as to employment is not deemed necessary in the present specification. A large number and wide variety of toning agents, compounds, and formulations are known, and examples are as follows: (1) ammonium molybdatesulfide; (2) ammonium carbonate; (3) sodium tetraborate; (4) auric chloride; (5) sodium aurochloride; (6) vanadium chloride; (7) uranyl chloride; (8) gold-platinum mixtures; (9) lead salts such as the acetate or the nitrate; (10) potassium chloroplatinate; (11) selenium; (12) sodium polysulfides and the like; (13) sodium thioantimonate; (14) sodium tungstate; (15) cobalt salts such as the chloride; (16) copper(II) salts such as the sulfate; and the like, one or more of which toners may be used in combination, as desired.

In the present Example 3, the photographic article is toned, by methods well known to those skilled in the art, with material which is radioactive, the radioactive ingredient finally appearing as part or all of the image or the like of the photo article under toning. In the case of alkaline earth element toners, e. g., Ba, Mg, Ca, Sr, I prefer to replace part of the alkaline earth element with the alkaline earth element radium, in compound form corresponding to the non-radioactive analog(s). The radioactivity of the toned article will then depend upon the quantity of radium which has been taken up in the toning process, and this may range from levels of micrograms, through milligrams, to multigrams; the microgram level is useful in research and tracer studies, for example; the milligram level in therapy and photocopying (using a master copy of the radioactive photograph and placing in contact therewith prints or negatives or the like to be reproduced); and, the multigram level for special applications of industrial radiography. The radium is conveniently a double salt of say barium.

Otherwise, it is preferred to use artificially radioactive elements, i. e., radioisotopes, in the toning process. Radiocobalt-60 may be used for the non-active cobalt; radioantimony (122 and 124) for the usual antimony in thioantimonates; radiocopper-64 for non-active copper; radiogold (especially Au-199) for non-active gold in the conventional gold toners; radioiron (say 55 and/or 59) for ordinary iron; radiomercury in mercury toners; radioselenium-75; and radiontrogen (say 185 and/or 187). These radioisotopes can be produced in a variety of reactions, but it is preferred that they be formed by pile-irradiation. As desired, the radioisotope can be introduced into the toning compositions or solutions, or, the same if not too complex, can be exposed to neutrons with the production of the radiotoner, thus eliminating the necessity of

chemical operations with dangerous radioactive matter.

Example 4.—The present Example 4 is related to Example 3, supra, in that photographic articles are toned with the metal or inorganic or like toner, chosen from a consideration of its qualities to acquire radioactivity, e. g., the elements listed under Example 3, and, finally rendered radioactive by suitable irradiation, e. g., pile derived neutrons, or the production of a nuclear reaction with deuterons, alpha particles or the like, effective to form a radioisotope which may or may not be the atomic species initially used. Examples 1 and 2 set forth means for the production of artificial radioactivity which are to be applied in the present example.

Moreover, elements susceptible to fission, for example those set forth in Example 2 are to be included in the present example and embodiment hereof. Thus, the conventional uranium toners of the photographic art are applied in the customary manner to the photographic article and then exposed to neutrons so as to produce fission, with the consequence of liberation of radioactive fission products therein photographic article. As desired, the ordinary uranium or enriched (with U-325) may be employed. In specific and special instances of application of the present invention, relatively pure or pure uranium-235 isotope may be used, which isotope is responsive to thermal neutrons, as is well known. However, it is anticipated that the need for this improvement or form hereof will be very slight, and restricted to research.

It is in this connection that in any or all of the present examples hereof improvement wherein nuclear reactions provide the radioactivity the reactant may, as desired, be in pure isotopic form or in material enriched in a given isotope, so chosen because it exhibits special and desirable properties well known to those skilled in the art.

Example 5.—The present Examples 5 and 6 correspond to the previous Examples 3 and 4 in that radioactive toning processes are employed, or, toning processing susceptible to induced radioactivity by say irradiation or fissioning are involved. In the present embodiment say the silver image is chemically altered, e. g., to brown silver sulfide or silver ferrocyanide or the like, and subsequently, at option, mordanted with basic substance such as a dyestuff. Furthermore, as desired, the radioactive toner or ingredient may comprise the material such as sulfide which changes the metal image and/or the mordanting substance, the basic material corresponding to the basic dye in the usual method.

Thus, taking silver halide photographic media as examples the reduced silver of the image or the like is chemically altered so as to form a compound such as silver ferrocyanide, silver sulfide or the like, and this in turn can be used as the mordant for basic dyes and chemical compounds. In one embodiment hereof, the use of the basic dye or the like is optional, since the production of a silver or like derivative may serve satisfactorily. Thus, in for example the so-called liver of sulfur toning bath, e. g., 60 grains of potassium polysulfides (liver of sulfur) in say 10-15 ounces of hot water, with 1 drop of ammonium hydroxide for each 4 ounces of the mixture, part or all of the sulfur may be replaced by radiosulfur-35 (the radioisotope made, say, by the pile-irradiation of sulfur) and/or radiopotassium-42 (prepared in the same way).

The prints or other like articles are merely immersed in this radioactive toning composition until the desired level of activity has been imparted to the product. Likewise, in the gold toners, of which a number of formulations are well known and not novel with me, radiogold may be substituted for the usual gold, in part or completely, with or without other elements, e. g., sulfur, being radioactive.

In yet another version hereof example, the photographic article is converted, say by the afore-described procedure, and is then dyed with a basic dyestuff containing one or more species of radioisotope. As examples of basic dyes, which list is not by far exhaustive, there is included safranin A; auramine; methylene blue; methyl violet; Victoria green; and chrysoidine 3R; and the like. It is not anticipated that this particular embodiment will be useful for other than research and specialty applications of the present invention, but accordingly the basic dyestuffs or the like (which may or may not be colored) serve as means for the introduction of radiocarbon-14 into the photographic medium, in large amounts in view of the number of carbon atoms which make-up such compounds. The radiocarbon-14 is preferably derived from neutron irradiation in the graphite pile, and the radioactive basic substance is synthesized previous to employment in the mordanting process by methods and means well known to those skilled in the chemical art.

Example 6.—This embodiment is like that set forth in Example 5, except that the photographic material is changed to a mordant or mordantable form, with the optional employment of basic dyes or the like, supra, and is then rendered radioactive by (a) nuclear reaction and/or (b) by production of fission with the consequence of radioactive debris and fission products being deposited. The methods for producing radioactivity by either of these agencies are set forth in more detail hereinbefore. For the radioactivity that results from fission by neutrons, the uranium toners are used, and the uranium in such formulations may, as desired, be enriched in uranium-235 isotope.

Example 7.—The present embodiment hereof comprises somewhat of a departure from previous examples and is based on what are known as "pigment processes," a variety of which are known; for example, the oil process, the bromoil process, the dichromate or gum-dichromate process, and the like, including the so-called carbon process. For purposes of the present specification and disclosure, a representative process (pigment process), the bromoil method is hereinafter set forth; as desired, variations and modifications hereof will be taken to include other species of the general category of pigment processes without violating the spirit and scope of the present invention.

The present Examples 7 and 8 correspond to pigment processes in which (a) radioactive pigments are used and/or (b) radioactivity is produced subsequently by say irradiation or fission, as described supra.

The bromoil process is based on the principle that a gelatin or like coated member, e. g., paper, wood, glass, accepts or repels oleaginous matter, e. g., greasy ink, in proportion to the amount of water present in the article. For example, an ordinary bromide print is made and bleached out so that the visible image of the regular print disappearing, leaves an image in gelatin. The bleached bromide print is soaked with water and blotted off. Radioactive oil pigment, or, as de-

scribed in Example 8, radioactively susceptible oil pigment, is applied via a brush and where parts of the article hold a large amount of water, as in the highlight areas, the ink or oleaginous matter is repelled. The deeper tones and shadows hold progressively lesser amounts of water, and therefore accept the oily matter, e. g., the ink, in proportion to the exact quantities. Much control can be exercised, and modifications of the amount taken up are readily possible. Such a product is as permanent as an oil painting, since the silver or other image or the like is actually replaced with one of pigment and oil. Amidol is generally the best developer, and a number of formulas as well known. Furthermore, bromoil transfers can be made, resulting in radioactive duplicates; this is done from a freshly inked print to another paper or like support, placing the two in contact by means of a press.

As examples of radioactive pigments which are used in the present form hereof, there is included: radiobarium-131, which when pile produced usually contains radiocesium-131 (for barium white, used in making "invisible" images); radiocalcium-45 (for gypsum or satinite); radiozinc-69 (for China White or zinc white); radioantimony (for antimony white); radiotitanium-51 (for titanium white); radiobismuth-210 (for bismuth white); radioiron-55 (for sienna, ochre); radiochromium-51 (for chrome yellow and the like); radiocadmium-115 (for cadmium yellow); radiocobalt-60 (for cobalt yellow); radiomercury (for cinnabar red); radiocopper-64 (for Bremen Blue); radiomanganese (for manganese black); radiocarbon-14 (for carbon black); and the like. These radiopigments are preferably produced by pile-irradiation but, as desired, may be derived from other sources and nuclear reactions. Those elements or pigments which are fissionable, as set forth supra, are employed likewise.

Example 8.—Instead of introducing radioactive matter in the pigment process, as set forth supra, the photographic article is prepared with a radioactively susceptible or fissionable pigment or material, and is then rendered radioactive by suitable treatment, e. g., pile-irradiation, exposure to alpha particles, deuterons or the like, as explained hereinbefore. As in Example 7, the pigments set forth are employed and then the article subjected to radioactivity inducement agency; furthermore, aside from the pigments listed under Example 7, fissionable pigments include uranium dioxide (uranium brown); uranium black or green (U_3O_8); uranium red (uranium trioxide); uranium yellow (sodium uranate); uranium orange (potassium uranate); also, complex compounds involving neutron or deuteron susceptible elements such as zinc, copper, and cobalt. Moreover, instead of uranium as the fissionable "pigment," lighter elements, say as oxides or silicates, can be employed in suitable comminuted forms in the customary oil or greasy vehicle, as well as heavier fissionable elements such as thorium dioxide and thorium silicate; in addition, compounds having more than one fissionable element may be employed, e. g., thorium platinocyanide, thorium uranate, and the like.

According to Examples 1 to 8, the radioactive photographic media will exhibit radioactivity characteristics which in part will depend upon the method and means of production. Thus, the method wherein radioactive reactants, mordants, dyes and compounds of say a basic nature, and pigments and the like, are introduced into the photographic media in a radioactive form will re-

sult in a product which does not exhibit extraneous radioactivities, e. g., radioactivities in other elements and materials making up the photographic article such as carbon. Hence, the image, highlights, or the like, will be more purely radioactive than those products wherein photographic media are irradiated by say neutrons, with the production of secondary and extraneous radioactivities; in the case of this invention where fissioning is responsible for radioactivity, then there will be a multitude of radioactivities, corresponding to the plurality of radioisotopes and fission products. Therefore, choice of a given method, form, article or embodiment must be made out of consideration of the utilization desired.

Whereas the terms and expressions "image," "physical image," "highlight(s)," "latent image" and the like are used in their customary physical and photographic meanings herein, it is seen that the present improvement has as its consequence the "radioactive image," "radioactive physical image," "radioactive highlight(s)," and the like, which are to be construed as embracing any sort of images or representations in gradiently radioactive areas upon or of a photographic medium, visible or invisible to the eye, detectable or substantially non-detectable by other chemical and/or physical methods and means of appraisal, or the like. Hence, a radioactive image may range from simple and complete reductions and non-reductions to any and all degrees and gradations thereof. The nature, characteristics, choice and mode of production of the image will depend upon the application desired, and this in turn may range from simple geometric figures, dots and lines, to "radioactively black-and-white images," to more complex images such as may be represented by numerals, letters, reading matter and printed matter and the like. In short, the term "image" and the like is herein equated with "intelligence" and its conveyance.

According to the present invention, a variety of refinements, embodiments and structural and geometric forms and modifications can be produced. Thus, a combination of radioactivities in a given image or intelligence conveyance can be produced, e. g., alpha emitters in the presence of neutron-reactive elements, e. g., gadolinium, lithium, boron, cadmium, which neutron reactive elements in turn emit special radiations, e. g., soft electrons; or, the radioactive image may be carried on a member of nuclear reactive metal, supra, or other desirable material, say containing radiation sensitive chemical compounds, e. g., phenylenediamine (which turns blue in the presence of short wavelength ultraviolet light); or, the radioactive image may be covered with responsive films or coatings, including sputtered or evaporated (vacuum for example) metals and/or compounds, e. g., those supra, also fluorides (with the consequence of hiding of any visible image that may exist), as well as luminescent solids such as zinc-o-silicate, zinc sulfide, calcium tungstate, and like phosphors; or, covered with strippable films or rigidly carried photosensitive emulsion; or, living matter such as bacterial culture media, diseased skin or other tissue; or, ultraviolet emitting solids such as calcium tungstate to act as intensifying screens in photographic applications; and the like, such modifications and variations needing no further description herein. Moreover, the radioactive image may be employed in specialized photocopying and photoduplicating processes by merely placing photographic material in con-

tact with the radioactive image and allowing same to remain for a suitable exposure, developing and fixing according to the art; in this phase, a stack or plurality of photographic papers or films, for example, may be used when the radioactive image emits gamma rays.

This invention is, in addition, not limited by geometric considerations; that is, while it is anticipated that plane surfaces will be of most general utility, involving say one dimension, multi-dimensional forms and geometries of any kind are not to be excluded. This is taken to include not only surface coatings and films, which in turn may be of more than one dimension and geometry, but also "solid geometries" and "three dimensional radioactive images" and radioactive photographic articles. Thus, the radioactive image(s) may comprise part or all of a cylinder or a sphere, for example, or say bodies corresponding to crystals of any kind, the solid mass thereof comprising three-dimensional or stereographic-like radioactive articles. In the matter of variations in geometry, the simplest may be represented by pictures and flat photographs, the more advanced by cameo and intaglio surfaces (which may or may not be plane, cf. supra) and the most advanced by analogs of statues and solid representations of this nature.

Furthermore, any of the specialty physico-chemical treatments, processing and the like known in the photographic and related arts may be directed to the present invention; thus, the radioactive image may be rigidly fixed on or within ceramic carriers; or, extraneous organic matter may be pyrolyzed or burned out, eluted out by solvents, changed by chemical reaction; or, the like, as desired.

I claim:

1. The method of irradiating an object with radioactive radiations which comprises the steps of taking a photographic image of the said object, causing the said image to become radioactive by treating same with radioactive material, and applying the thus treated image to the said object.

2. The method of irradiating an object with radioactive radiations which comprises the steps of taking a negative photographic image of the said object, the said image being approximately the real size of the object, causing the said negative image to become radioactive by treating same with radioactive material, and applying the thus treated image to the said object.

3. The method of irradiating an object as set forth in claim 1 wherein the said radioactive material emits gamma radiation.

4. The method of irradiating an object as set forth in claim 1 wherein the said radioactive material emits beta radiation.

5. The method of irradiating an object as set forth in claim 1 wherein the said radioactive material emits alpha radiation.

6. The method of irradiating an object as set forth in claim 1 wherein the said radioactive material emits neutron radiation.

7. A radioactive structure which comprises a photoimage member containing radioactive matter having a half-life of between approximately one minute and 2×10^{10} years, the quantity of radioactive matter in the said photoimage member being in direct proportion to the reduced areas of the photoimage member, said radioactive photoimage member being carried upon non-radioactive support means.

8. The radioactive structure set forth in claim

7 wherein the said radioactive matter emits gamma radiation.

9. The radioactive structure set forth in claim 7 wherein the said radioactive matter emits beta radiation.

10. The radioactive structure set forth in claim 7 wherein the said radioactive matter emits alpha radiation.

11. The radioactive structure set forth in claim 7 wherein the said radioactive matter emits neutron radiation.

12. In the process of preparing a photographic image including the steps of forming a latent image upon a photographic article and processing the said latent image into a physical image, the improvement of imparting radioactivity to the said physical image in direct proportion to the reduced areas of the said image, said radioactivity characterized as having a half-life of between approximately one minute and 2×10^{10} years, whereby a radioactive photographic image is produced.

13. In the process set forth in claim 12 wherein the said radioactivity is gamma radiation.

14. In the process set forth in claim 12 wherein the said radioactivity is beta radiation.

15. In the process set forth in claim 12 wherein the said radioactivity is alpha radiation.

16. In the process set forth in claim 12 wherein the said radioactivity is neutron radiation.

17. The method of irradiating an object as set forth in claim 1 wherein the said radioactive material emits gamma radiation.

18. The method of irradiating an object as set forth in claim 1 wherein the said radioactive material emits beta radiation.

19. The method of irradiating an object as set forth in claim 1 wherein the said radioactive material emits alpha radiation.

20. The method of irradiating an object as set forth in claim 1 wherein the said radioactive material emits neutron radiation.

21. As a new article of manufacture, a source of radioactive radiation comprising the combination of a non-radioactive supporting member and an element bearing a photographic image carried upon the said member, said photographic image containing radioactive matter of half-life between approximately one minute and 2×10^{10} years, the amount of radioactive matter in the said image being in direct proportion to the reduction thereof.

22. The article set forth in claim 21 wherein the said radioactive matter emits gamma radiation.

23. The article set forth in claim 21 wherein the said radioactive matter emits beta radiation.

24. The article set forth in claim 21 wherein the said radioactive matter emits alpha radiation.

25. The article set forth in claim 21 wherein the said radioactive matter emits neutron radiation.

26. As a new article of manufacture, a source of radioactive radiation comprising the combination of a non-radioactive supporting member and an element bearing a photographic image carried upon the said member, said photographic image containing radioactive matter having a half-life of between approximately one minute and 2×10^{10} years, the amount of radioactive matter in the said photographic image being in proportion to the reduction thereof.

27. The article set forth in claim 26 wherein

the amount of the said radioactive matter in the said photographic image is in direct proportion to the reduction thereof.

28. The article set forth in claim 26 wherein the amount of the said radioactive matter in the said photographic image is in inverse proportion to the reduction thereof.

29. In the process of preparing a photographic image including the steps of forming a latent image upon a photographic article and processing the said latent image into a physical image, the improvement of causing the said physical image to become radioactive in proportion to the reduction thereof, said radioactivity having a half-life of between approximately one minute and 2×10^{10} years, whereby a photographic source of radioactive radiation is produced.

30. In the process set forth in claim 29 wherein the said physical image is radioactive in direct proportion to the reduction thereof.

31. In the process set forth in claim 29 wherein the said physical image is radioactive in inverse proportion to the reduction thereof.

32. A radioactive structure which comprises a photoimage member containing radioactive matter having a half-life of between approximately one minute and 2×10^{10} years, the quantity of radioactive matter in the said photoimage member being in proportion to the reduced areas of the photoimage member, said radioactive photoimage member being carried upon non-radioactive support means.

33. The radioactive structure set forth in claim 32 wherein the quantity of the said radioactive matter in the said photoimage member is in direct proportion to the reduction thereof.

34. The radioactive structure set forth in claim 32 wherein the quantity of the said radioactive matter in the said photoimage member is in inverse proportion to the reduction thereof.

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REFERENCES CITED

The following references are of record in the file of this patent:

UNITED STATES PATENTS

Number	Name	Date
821,655	Lieber	May 29, 1906
1,406,488	Pugh	Feb. 14, 1932
1,882,426	Kalix	Oct. 11, 1932
2,206,634	Fermi et al.	July 2, 1940

FOREIGN PATENTS

Number	Country	Date
5,182	Great Britain	of 1912

OTHER REFERENCES

Wall: "Intensification and Reduction," pp. 15-18, American Photographic Publishing Co., 1927. Copy in Div. 67.

Ellis et al.: "Artificial Radioactivity," Proc. Royal Soc. (London), vol. 146, pp. 206-7 (1934). Copy in Patent Office Library.

Towler: "The Silver Sunbeam," published in N. Y. (1864) by J. H. Ladd, pages 272-273. Copy in Scientific Library.

Saint-Victor: Comptes Rendus, January-June 1858, tome 46, pages 448-452. Copy in Sci. Library.

Ser. No. 395,760, Schopper et al. (A. P. C.), published May 11, 1943.