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LUCAS HEIGHTS

AUTORADIOGRAPHY OF RADIOACTIVE PHOTOGRAPHIC IMAGES – APPLICATIONS TO INTENSIFICATION, RESTORATION, PRECISION ETCHING, PHOTOMECHANICAL REPRODUCTION AND PHOTOGRAPHIC RESEARCH

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ABSTRACT

Applications of nuclear science to photographic techniques are described. They are related to the observation that autoradiography of radioactive silver images confers three unique benefits on the process of photography:

(i) Information can be retrieved from a much smaller number of silver grains in a developed negative than is required using the transmission of visible light. This means that lower exposures are acceptable.

(ii) Films used to record radiations of reasonable penetrating power (as in radiography with X-rays or neutrons) are no longer required to be

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transparent. This permits a much wider selection of materials for use in their fabrication.

(iii) Photographs toned with isotopes which emit densely ionising radiation will produce a radiation damage pattern in any surface which contacts them. In many such surfaces, etching techniques can reproduce the photograph as a high fidelity intaglio image. Such etchings can provide much more faithful relief reproductions of the photograph than were possible by previous methods.

Experimental work is described on the application of these principles to the intensification of underexposed and faded photographs, to precision etching and to the multiple reproduction of photographs by printing.

A literature survey is also included, together with some speculation on the possible future interactions between nuclear science and photography.

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The following descriptors have been selected from the INIS Thesaurus to describe the subject content of this report for information retrieval purposes. For further details please refer to IAEA–INIS–12 (INIS: Manual for Indexing) and IAEA–INIS–13 (INIS: Thesaurus) published in Vienna by the International Atomic Energy Agency.

ALPHA PARTICLES; AUTORADIOGRAPHY; CALIFORNIUM 252; ETCHING; HALIDES; IMAGE INTENSIFIERS; IMAGES; NEUTRONS; PHOTOGRAPHIC FILMS; PHOTOGRAPHY; SILVER COMPOUNDS; X RADIATION

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1. INTRODUCTION

Information stored on photographic films is nearly always retrieved by the transmission of visible light. Whenever this method of information retrieval is used, three very important limitations are imposed on the photographic process. These are:

(i) The image must be visible. Usually, to be acceptable, the silver grains of the image must change the light transmission of the other film constituents (gelatine and base) by at least ten per cent.

(ii) The matrix in which the silver grains are suspended must have high transparency so that the image can be seen clearly.

(iii) The image can be copied directly only on materials which are themselves photosensitive, *e.g.* silver halide film, diazo type paper, blue print paper, *etc.*

The first condition of image visibility demands that image grain densities should be in the region 10^7 - 10^9 per cm^2 (owing to the necessary small diameter of individual grains - usually 0.5 - 2.0 μm). However, much of the information represented by these high grain densities is redundant for many applications. (For example, in a high quality journal reproduction of a photograph, the dot structure might have only about 10^4 ink spots per cm^2.)

The second condition, demanding transparency of the emulsion (after processing), has tied the photographic process almost exclusively to the clear, water-permeable materials gelatine and polyvinyl alcohol. It has also prevented addition to the emulsion of any solid or strongly coloured substances which cannot be dissolved out in the processing solutions. Since most increases in photographic sensitivity have come about by the addition of dyes and traces of sensitising agents to the emulsion, and since even the best modern photographic films have a low detected quantum efficiency for visible light (Bird, Clarke-Jones & Ames 1969) and for X-ray quanta having energies of industrial importance (see Table 1), the transparency condition imposes a severe limitation on photographic research.

The third condition is particularly limiting to the preparation of printing surfaces used in rotogravure or lithography for the multiple reproduction of photographic images. Only by complicated procedures, using diffusing screens and etch resists which can be photopolymerised, can photographic images be transferred to surfaces of metal, glass, ceramics or plastics as relief images. These processes cause the information content of the photograph to be reduced from > 10^7 grains cm^{-2} to < 10^4 grains cm^{-2}.

Methods other than light transmission have been used to derive information

from developed photographs. Enomoto & Tominaga (1971) have used X-ray fluorescence to determine silver in photographic dosimetry, but this method appears incapable of high spatial resolution of photographic images and is unlikely to compete with autoradiography in most applications.

Stevens (1948) prepared photographic test patterns to show how these could be toned with radioisotopes and used to prepare autoradiographs. These autoradiographs permitted a numerical estimate of the resolution which might be achieved when different radioisotopes were used in biological autoradiography. These measurements are by no means simple, depending on such factors as exposure time, grain size, emulsion thickness and the type of nuclear radiation involved. The application of the technique was studied in more detail by Herrmann, Hartmann & Brust (1962) and by Bleeken (1968a, b). De Ment (1952) described many other methods of making photographic images radioactive, and suggested very many applications for radioactive photographs. Tellez-Plasencia (1959) used radiotoning or neutron activation to analyse for silver in films to levels as low as those encountered in photographic latent images. Later Ostroff (1966) showed how neutron activation of old faded photographs of historical interest followed by autoradiography could be applied as a safe and useful method of restoration. Chatters & Jacobs (1970) have reported further work in this field. Herman, Hartmann & Schumpelt (1968) and Thackray and associates (Thackray 1968; 1972a, Thackray *et al.* 1972; Thackray & Roman 1972) have shown how autoradiography of underexposed photographs can be used as a method of photographic intensification.

The deposition of uranium compounds on the silver grains of an underexposed photograph has been used for many decades as a method of photographic intensification. The method depends entirely on the increased size and light absorption of the toned silver grains and, surprisingly, no report of autoradiographs being prepared from such uranium-toned images can be found. Methods of toning with several other alpha-emitting isotopes, and with the spontaneously fissile isotope californium-252, were described by Thackray (1968) (see also Appendix A).

Autoradiographs can be prepared from photographs toned by these latter methods, without the use of normal photographic materials or darkroom techniques, since Fleischer *et al.* (1965, 1972) showed that damage trails produced by charged particles in non-conducting solids can be etched to produce pits which can be seen clearly under the microscope. It became obvious that these discoveries made possible a new technique for preparing intaglio images and for photomechanical reproduction (Thackray 1972b, 1974).

2. METHODS OF MAKING SILVER IMAGES RADIOACTIVE

2.1 Preliminary Treatment of Films

Where an increase in photographic speed is sought from the application of autoradiographic intensification, it is necessary to keep non-image silver fog to a minimum since this is the major limiting factor in the procedure. Of particular importance is the use of fresh film, the elimination of all extraneous light during handling and exposure, and processing in total darkness. Wherever possible, an unexposed section of film should be developed and processed in parallel with the film to be intensified so that the effect of fog can be assessed (usually by means of a radiation counter) at all stages of the process. This is particularly desirable when reducers, such as dilute potassium cyanide or other complexing agents, are employed to lower the general fog level after radiotoning.

The photographs to be intensified should be freed from any grease, fingermarks, *etc.* by washing in dilute aqueous detergent or petroleum ether. They should then be refixed and, preferably, hardened (with formaldehyde) in fresh photographic fixer, to remove any residual traces of ionised silver, then thoroughly washed. Heavily fogged films frequently give better results if the fixation step is replaced by slight density reduction with a photographic reducer. How far these preliminary steps can be applied to old faded photographs has to be decided on individual merit, since images which have oxidised almost completely can easily be lost by dissolution.

There are three general methods of making a silver image radioactive: by nuclear reaction, by radiotoning and by silver isotope exchange.

2.2 Nuclear Reactions

Nuclear reactions can be induced by sub-atomic particles or high energy gamma rays. Of these only the reactions of thermal neutrons are of practical importance, since no other reactions can induce sufficient radioactivity without destruction of the photograph. When untreated silver images react with thermal neutrons, only the following reactions are important:

$$^{107}Ag\ (n,\gamma)\ ^{108}Ag \quad (t_{½} = 2.4 \text{ minutes})$$

$$^{109}Ag\ (n,\gamma)\ ^{110}Ag^{m} \quad (t_{½} = 253 \text{ days})$$

The half-lives involved are either inconveniently short, or inconveniently long. (Long half-lives are usually impractical for activation procedures, since a long irradiation period is necessary to produce appreciable radioactivity and the photograph becomes damaged by radiolytic decomposition of gelatine and film base.) If time is no object, good autoradiographs can be

obtained from low levels of silver-110m activity when all other forms of radioactivity have decayed but normally one has to use the short-lived isotope.

Irradiation of low density photographs at a thermal neutron flux of 7×10^{12} neutron $cm^{-2}s^{-1}$ for 5 minutes followed by a cooling period of 2 minutes and autoradiographic exposure for 10 seconds, has given dense autoradiographs, but the procedure is inconveniently hasty and estimation of the correct autoradiographic exposure is difficult. Full automation of irradiation, activity estimation and autoradiography could remove most of these objections but is not yet warranted by the quality of the results obtained. All the photographs produced by this technique using silver-108, became heavily fogged owing to activation of impurity elements in the gelatine. The most troublesome isotope was chlorine-38, and no techniques could be devised to remove this impurity completely from photographic negatives. This has also been noted by Russell (1967).

The disadvantages arising from neutron activation of silver images can be removed by first toning the images with elements which give rise to more convenient radioisotopes when irradiated. Examples of such elements which give softer radiation, more convenient half-lives or more intense activity are mercury, indium and uranium.

2.3 Radiotoning

The silver is made to react chemically with a radioactive material so that the silver grains become coated with a radioactive compound or are replaced by such a compound.

The apparatus used differs somewhat from that used in normal photography. Open trays are not very suitable for handling radioactive toning solutions. It is preferable to treat films in a vertical position either coiled around the wall of a cylindrical vessel or supported in a specially designed rectangular tank. Magnetic or gas stirring is convenient for cylindrical vessels and gas stirring for rectangular tanks.

Toning procedures for many elements are described in the literature of photography and usually require only slight modification for use with radioelements. The modification is usually in the direction of decreasing concentrations and increasing reaction times, since the recommended concentrations at the necessary specific activity would give rise to curie levels of activity in every toning solution, even though only microcuries of activity are required for individual photographs. The low concentration of reagents frequently demands that static treatment of the film be replaced by vigorous shaking or stirring.

The simplest toning reaction is the displacement of a more noble metal, usually in solution as the thiocyanate complex, by the silver of the film image. Unfortunately, no radioisotopes with suitable properties are readily available in this group, but we have tested the procedure with gold-195. Toning with elements of the sulphur group (*i.e.* sulphur, selenium, tellurium and polonium) is most convenient from a radiochemical point of view, since a highly insoluble silver compound can be formed in a single stage treatment. Most other toning reactions are more complex, frequently involving the initial formation of silver ferrocyanide and the precipitation of other metal ferrocyanides. These compounds generally have appreciable aqueous solubility and some loss of fine detail may occur on photographs of very low density.

When the toning reaction is complete, any unreacted activity must be removed from the gelatine. Simple washing with water is rarely able to accomplish this. In the case of many elements (*e.g.* sulphur), washing with an inactive solution which is identical with the toning solution will cause the activity adhering to the gelatine to exchange much more rapidly than that in the toned silver grains. In other cases, complexing agents must frequently be used to remove activity from the gelatine.

Toning procedures have been developed for sulphur-35, promethium-147, cadmium-115m, iron-55, nickel-63, mercury-203, silver-110m, gold-195, polonium-210, uranium-233, americium-241 and californium-252. Untreated silver images and silver images toned with mercury, indium and uranium have been activated with neutrons. Several procedures are described in more detail in Appendix A.

After radiotoning, washing and drying, films usually behave as sealed sources and do not spread contamination, but it should be remembered that the gelatine is easily destroyed by heat and by many chemicals.

2.4 Silver Isotope Exchange

The exchange of silver between the photographic image and silver nitrate solution labelled with silver-110m is slow and incomplete, and much activity becomes fixed in the gelatine. If the silver is complexed with ammonia or thiosulphate, better results are obtained but the exchange still proceeds very slowly. Preliminary conversion of the image to silver bromide by the frequently used photographic bleach, consisting of a mixed solution of potassium ferricyanide and potassium bromide, allows a more complete exchange of silver isotopes.

When high specific activity silver nitrate solution is employed (several Ci g^{-1}), the silver can be added to the bleach solution without the formation

of a permanent precipitate and exchange occurs while the silver bromide is forming, as a rapid one stage procedure. More prolonged stirring with the same solution helps to reduce silver fog on the original photograph.

This has proved to be one of the most convenient methods of activating a silver image. Although the gamma ray emission of silver-110m is inconvenient from a handling point of view, this has a negligible effect during autoradiography. Most of the exposure can be attributed to the soft beta fraction having E_{max} = 0.087 MeV.

Combinations of these general activation procedures are also possible and often can be advantageously applied. For example, the silver image can be toned before irradiation with neutrons, using an element which has a larger cross section or more convenient half-life than silver. Indium, iodine and mercury are of particular importance for such applications.

Alternatively, the two latter general procedures can be combined by first toning with an inactive element and then allowing exchange to occur with the radioelement in solution. This procedure is of particular importance when the radioelement is in a carrier-free or almost weightless form, which makes it difficult to exceed the solubility product of any of its compounds so as to bring about precipitation. Cations generally exchange more rapidly than anions.

In some cases, autoradiography can be performed concurrently with neutron irradiation where a toning element gives rise to instantaneous corpuscular radiation on neutron capture (*e.g.* lithium, boron, uranium and plutonium; see Section 4).

Usually, procedures involving toning the silver image with a radioactive isotope are the most convenient, since they do not require access to a source of neutrons. Radioisotopes with any desired properties can be selected for rendering the photographic image radioactive. In general, it is better to employ soft beta emitters of high specific activity.

2.5 Radioactive Development

The procedures described in Sections 2.1 to 2.4 must be applied to silver images which have been developed and fixed in the usual manner, although a few of them can be modified to apply to photolytic images which have been fixed without development. It is probable that toning procedures could be devised to proceed concurrently with fixation of the photograph. However, such fixing solutions are limited by technology and would show little advantage over separate fixing and toning solutions. If toning can occur concurrently with development, several important possibilities arise and, for some

elements, it is not too difficult to envisage procedures since they can be based on existing systems of colour photography.

Possible methods of making silver images radioactive while the images are developing, and the possible benefits from such a system, are discussed in Appendix B.

3. AUTORADIOGRAPHY

All silver halide films are sensitive to ionising radiation, but they show a very large range of sensitivity and resolving power when used for autoradiography. The fastest X-ray films allow an autoradiograph to be obtained in the shortest possible time, but the large grain size and double coating frequently produce unacceptable results. Usually it is convenient to produce such an autoradiograph after an exposure of one or two hours and, from an estimate of the density and contrast produced, to select a finer grain single-coated film for a longer (overnight) exposure. For very soft beta radiation (nickel-63, tritium), the correct exposure can only be estimated by such trials, but with other isotopes a radiation counter operating at a fixed distance from the photograph will permit a good estimate of the exposure time. The contrast of the autoradiograph can be greatly influenced by the choice of film. The toning elements should have specific activities greater than 1 Ci g^{-1} to obtain autoradiographs on fine grain X-ray films with an overnight exposure.

The best resolution of detail down to about 1 micron can be achieved with soft beta emitters such as tritium. Caro & Schnös (1965), using electron microscope autoradiography, obtained a limit of resolution of about 0.1 micron. With more penetrating radiation, where the range exceeds the emulsion thickness, resolution is apparently more a function of the emulsion and exposure time than the radioisotope used. With fine grain emulsions, resolving power of 4 - 5 microns can be readily achieved. Owing to the rapid decrease of radiation exposure with radial distance from a toned silver grain, one can achieve quite good photographic reproduction even with a pure X-ray emitter such as iron-55.

For reasons such as these the choice of a radioisotope for radiotoning is usually dictated by factors other than resolving power.

3.1 Intensification of Underexposed Negative

Autoradiography with most radioisotopes can locate individual silver grains, and carrier-free isotopes such as sulphur-35 or polonium-210 (which have a strong affinity for silver) can produce dense autoradiographs with an overnight exposure when the surface concentration is as low as 10^{-13} g cm^{-2}.

This concentration is less than the levels at which silver is considered to be present in photographic latent images before development. It has not proved possible, however, to apply these intensification procedures to normally exposed latent images. This is probably because normal fixation procedures dissolve a large fraction of the latent image. Tellez-Plasencia (1959) has described an elaborate fixation procedure in an inert atmosphere which, he claims, must be followed to preserve the latent image prior to neutron activation. Tests with silver nitrate solutions of known concentration dried on porous materials showed that artificial silver patterns on inert surfaces could be autoradiographed after treatment with polonium-210 down to levels of about 10^{-11} g cm^{-2}. We were also able to intensify grossly overexposed (although still invisible) latent images by autoradiography after normal fixation but, as this procedure appeared to have no practical value, most work was concerned only with photographs which had been developed and fixed in the usual manner.

Normal photographs for visible light work, even when developed without exposure, usually produce fog levels greater than 10^{-5} g cm^{-2} silver. Very fast films, old films and films which have been exposed for long periods to darkroom 'safe-lights' can produce much higher levels of silver fog. It is important to remember that, if fog can be clearly seen on a film and no image can be seen, fog grains will outnumber image grains by at least 10 to 1 and it is most improbable that any intensification procedure will extract worthwhile information. This developer-produced fog seems to be an unavoidable attribute of fast films for visible light work, but it can be considerably reduced by such precautions as using fresh film and special developers, and eliminating safe-lights and other forms of extraneous radiation. With many medium speed films, the limit of underexposure which can be usefully intensified is about 2 per cent of the optimum exposure and this is mainly due to the problem of fog. The experience of nuclear physicists has shown that emulsions for recording densely ionising radiations can be made virtually fog free (Barkas 1963), and some development procedures allow proton tracks to be recorded in the presence of very high gamma fields without undue fogging (Bishop & Saha 1968). More recently Dostes (1972) has described the manufacture of X-ray films which are not fogged by light and can be processed outside the darkroom. It seems certain that pinhole camera images using such radiations and such films could be intensified at silver density levels well below those which are currently used in visible light photography. Such applications might be of considerable value to astronomers now that pinhole

cameras can be deployed above the Earth's atmosphere; films might even be designed specifically for use with autoradiographic intensification which would respond to radiations of lower ionising power without producing high levels of background fog. Since the original images are not required to be visible,the high amplification factor of normal development which produces most of the fog might be discarded. Alternatively, non-silver photographic systems, which have proved impractical owing to inadequate development factors, might be reconsidered for use with autoradiographic intensification.

3.2 Restoration of Faded Photographs

Photographic prints kept over many decades frequently show fading of the black silver grain images through sepia tones to almost complete invisibility. The rapidity with which this process occurs depends, mainly, on how thoroughly the prints are washed free of traces of the processing solutions and the type of atmosphere to which they are subsequently exposed. Fading involves the oxidation of metallic silver to compounds of low visibility such as silver sulphate. No silver is lost from the photograph and, provided there is no diffusion of soluble silver compounds, the information content is unchanged.

Examination of very old photographic negatives from the NSW Archives showed that they fall into two categories. In one the medium supporting the silver image was collodion and in the other it was gelatine. The collodion photographs are completely impermeable to aqueous solutions and the only practical method of activating the silver image was by neutron irradiation. Many of the gelatine photographs were so deteriorated that they would be destroyed by treatment with aqueous solutions and these too would only be suitable for neutron activation. Several photographs, where the gelatine was in good condition, were restored by toning them with sulphur-35 followed by autoradiography. However, it was shown by Mr. D. Roman (AAEC unpublished paper), that photographs in this category could frequently be restored by simply redeveloping the faded image. As this much simpler procedure is less likely to damage the photograph, it appears likely that neutron activation is the only nuclear technique that will find application in the field of restoration. Here too, in the case of thick glass-based negatives, the technique will only be worthwhile in extreme cases since very high sodium-24 activities must be allowed to decay before autoradiography becomes possible. For photographs not based on glass, neutron activation can be more easily applied and Ostroff (1966) and Chatters & Jacobs (1970) have restored several photographs of historical interest.

3.3 Use with Loaded Emulsions as a Possible Method of Increasing Photographic Sensitivity

The removal by autoradiography of the two limiting conditions of image visibility and matrix transparency will allow further photographic research on methods for increasing photographic speed. The case of radiographic films is of particular interest. Films which can be processed in an acceptable time contain insufficient silver bromide to have high stopping power for the X-ray energies, which are of value in medical and in industrial radiography, and are not much affected by neutron beams. They therefore have low photographic efficiency for recording such radiation.

Salts of heavy metals have been used for many years to improve the sensitivity of X-ray films (Tomado 1959, Larson *et al.* 1953). This efficiency can also be increased by exposing the films in contact with screens of heavy metals or salts which fluoresce when exposed to X-rays. Heavy metal screens intensify by virtue of the photoelectrons which they emit and which strike the photographic emulsion. Fluorescent salt screens convert some of the X-ray quanta to visible light which is more effectively stopped by the film. The very short range of photoelectrons limits the use of metal screens to the higher range of X-ray energies. The use of fluorescent screens is limited mainly by resolution loss as screen thickness is increased. With the removal of the transparency condition, it becomes possible to incorporate into the emulsion, materials of the type used for intensifying screens. Closer proximity of the heavy material or phosphor to the silver bromide grains may then permit more efficient energy transfer and improvements in resolving power.

TABLE 1

PER CENT X-RAY ABSORPTION BY NORMAL AND LOADED EMULSIONS

X-ray Energy (keV)	Normal[(a)] Emulsion	Gold Loaded[(b)]	Phosphor Loaded[(c)]
12	17	87	47
25	6.0	39.5	12.7
50	2.5	9.0	5.5
100	0.37	5.5	0.88

(a) *3.5 mg cm^{-2} AgBr*

(b) *3.5 mg cm^{-2} AgBr + equal volume of Au (10.4 mg)*

(c) *3.5 mg cm^{-2} AgBr + equal volume of $CaWO_4$ (3.28 mg).*

Table 1 shows the calculated X-ray absorption of a film containing 3.5 mg cm^{-2} of silver bromide, and of similar films loaded with metallic gold and calcium tungstate phosphor present in equal volume with the silver bromide. In the most favourable case, gold increases the stopping power of the film by a factor of 15 at 100 keV. The addition of calcium tungstate increases the stopping power by a factor of about 2.5 at 12 keV and 100 keV but it is less effective at other energies.

When an X-ray photon is stopped by gold, one photoelectron of limited range is produced in the emulsion, but when an X-ray photon (or photoelectron) is stopped by the calcium tungstate phosphor, a large number of visible light photons is produced to which emulsion has considerable transparency. The sensitivity of photographic emulsions loaded with heavy metals only is, thus, critically dependent on geometrical conditions which permit photoelectrons produced by the heavy metal to interact with the silver bromide; in practice, this means a very fine grain size and a low gelatine content. In emulsions loaded with phosphor, or heavy metal coated with phosphor, geometrical considerations may not be so critical.

In a similar manner, the loading of films for special purposes (*e.g.* boron for neutron radiography and particular elements for high energy physics research) should be a much simpler task, now that removal of the transparency requirement permits the use of finely divided inert solids.

While the task of obtaining materials which are chemically compatible with emulsions, and in a suitable state of subdivision, will present many problems, it seems certain that methods of information readout from photographic films, which do not rely on light transmission and of which autoradiography is currently the most useful, will permit considerable development in photographic technology (Thackray 1973, 1974).

4. PRODUCTION OF HIGH FIDELITY INTAGLIO IMAGES FROM ETCH TRACKS

The emission of densely ionising radiation, such as fission fragments or alpha particles, from photographic images toned with radioisotopes permits reproduction of the photograph in the form of a radiation damage pattern in most surfaces with which the photograph is placed in close contact. In many materials, subsequent etching of this damage pattern may cause the photograph to be reproduced in the form of a series of visible etch pits.

These etch pits bear a close positional relationship to silver grains in the original photograph and permit its reproduction with high fidelity on many surfaces which are not normally photoreceptive.

Since fission tracks can be etched easily in a large number of materials

(glass, plastics, ceramics, *etc.*) while alpha tracks can only be etched in a few esters of cellulose, alpha particles are currently of much less importance in the practical application of this technique than fission fragments. The simplest method of employing the technique is to use photographs toned with californium-252.

4.1 Method Using Spontaneous Fission

4.1.1 Toning photographs with californium

Photographic images are first bleached to silver ferrocyanide and then treated with an acidic solution of Cf^{3+}. (For details see Appendix A.)

4.1.2 Quantities and costs

The amount of californium which must be deposited depends mainly on the time one is prepared to allow for the autoradiographic exposure and the purpose for which the etching is required. A mean value of about 10^6 etch pits per cm^2 will give a good full tone reproduction of a photograph as an etching on a glass surface if the etch pits are developed to about 10 μm diameter (~ 15 min. in 1:10 HF). To obtain 10^6 etch pits with an exposure time of one hour, approximately 10^{-9} g of californium-252 must be deposited on each cm^2 of film. The cost of this quantity of isotope is quite trivial, even without recycle, and is likely to fall continuously as production increases (USAEC 1969).

4.1.3 Exposure and etching

As with ordinary autoradiography, good contact between all parts of the emitter and receiver is essential for good resolution. Few problems arise with small plane surfaces, but larger or more complicated surfaces benefit greatly from the application of vacuum. Strippable emulsion could possibly be applied to still more complex surfaces.

Unlike ordinary autoradiography and photography, in which there is a fairly closely defined exposure time and development time, the exposure latitude when producing these intaglio images is surprisingly wide. This is due to the fact that varying etching times can produce etch pits ranging from less than 1 μm to more than 50 μm in diameter. This represents a pit area variation of > 2,500 and, if the mean area of the picture which is covered by etch pits is to remain the same (as it must to achieve optimum contrast), the exposure time must be varied by a similar factor. The best looking etchings are produced,however, when the etch pits are in the range 5-15 μm diameter. Glass and mica etched with hydrofluoric acid, and nitrocellulose and polycarbonate etched with strong solutions of caustic alkali have been the materials most frequently used for detecting nuclear tracks,

but the number of materials and etchants will probably increase considerably in the future.

4.2 Method Using Neutron-Induced Fission

While californium-252 is the only available isotope which shows appreciable decay by spontaneous fission, a considerable number of isotopes (*e.g.* uranium-233, uranium-235, plutonium-239 and americium-242) are available which undergo fission with neutron irradiation.

Photographs toned with these isotopes produce fission-damage autoradiographs in exactly the same manner as californium, provided they are irradiated with neutrons while in contact with suitable materials. Uranium has been used for many years to intensify underexposed photographs by a non-radioactive toning procedure, and the method described for californium is applicable also to americium and probably to plutonium (Appendix A).

The irradiation procedure has been tested using photographs toned with uranium-233. A nuclear reactor is necessary, at present, to provide a sufficiently high thermal neutron flux to obtain an adequate etch pit density in a reasonably short time (about 10^{10} n s^{-1} for one hour). High levels of radioactivity are induced in the sample and the method is much less convenient than the use of californium. However, as new types of neutron source become available to industry, it is possible that radiological protection would be less costly for large scale work using this method in preference to californium toning.

4.3 Method Using Alpha-Ray Etch Tracks

It is more difficult to etch alpha-ray damage trails than those produced by fission fragments. Even for the most sensitive material, nitrocellulose, the alpha-ray energy must be below 3 MeV before the ionisation density becomes high enough to allow etch tracks to be produced. Although this requirement has no noticeable effect when using toned photographs, because the gelatine degrades the energy of many alpha particles below this value, the tracks appear smaller and are more difficult to develop.

4.4 Method Using Boron-10 and Neutron Irradiation

All the etching methods discussed so far involve the use of highly toxic alpha emitters as toning materials, and only well-equipped radiochemical laboratories can handle such materials without hazard to personnel. In each of these cases, radiological protection would be an appreciable overhead cost on the operation of any commercial application of the etching process.

Only one method of operating the process can be envisaged in which contamination hazards will be negligible. This is to employ the prompt alpha

emission produced by the neutron irradiation of boron or lithium. This method of autoradiography has been frequently employed to measure the surface concentrations of these elements in metallurgical specimens (Hughes & Rogers 1967) and in glass (Wood & Robinson 1970). Although this alpha emission can produce direct intaglio images in only certain esters of cellulose at present, it seems probable that other plastics susceptible to alpha track etching will be developed. As discussed later (Section 6), it is also possible to use thin layers of these polymeric materials as etch resists so that high fidelity intaglio images can be etched in most materials.

4.5 Possible Applications of Etch Track Images

In addition to the direct use of toned photographs to produce useful and decorative etchings, several other applications have also been explored.

4.5.1 Production of metal relief photographs

The particle track etching procedure described in the previous section can also be applied to produce relief images in which the image is raised above the surrounding surface.

One method of preparing such relief images is to make replicas of intaglio images produced by the methods described in Sections 4.1 - 4.4. One of the most convenient of these methods is to prepare an intaglio image on a sheet of glass using a photograph toned with californium-252. A thin layer of silver is then deposited on the glass from a reducing solution of the type used in the manufacture of mirrors. The thin metal layer is then reinforced by electroplating until it is thick enough to be separated from the glass. The initial metal coating can also be applied by vacuum evaporation and it seems probable that casting techniques could be developed to prepare replicas.

In this particular application, since one radioactive photograph can produce many glass plates bearing intaglio images and each glass plate can produce many replicas, the cost component for handling radioactivity to produce these metal relief photographs would be negligible.

An entirely different method of preparing these relief images is to coat a metal plate with an etch resist in which fission track holes can be generated (*e.g.* polycarbonate or nitrocellulose). Further material is then electrodeposited on the plate through the track holes. When these relief images are prepared using precious or non-corroding metals, very permanent and decorative photographs can be obtained which may be of value in the manufacture of jewellery and the preservation of archival records.

4.5.2 Use of intaglio images in printing

Comparison with current methods of printing. There is a great and continuing need to transfer photographic images to surfaces which are not normally photoreceptive. This need is illustrated by the multiple reproduction of photographs in newspapers, magazines and books, by the transfer of decorative pictures or patterns to packaging materials, glassware, pottery, ceramics *etc.*, and by the reproduction of durable scales on measuring instruments. Faithful reproduction of photographic images on such surfaces presents no serious problems for line drawings or half-tone reproductions which do not require high resolution.

The method employed generally is to coat the material which is to receive the image with a thin layer of a substance which can be made insoluble (usually by polymerisation) when exposed to light. Such materials are generally referred to as photopolymerisable etch resists. When such a thin layer is exposed beneath a two-tone (black and white) photograph, the parts of the layer which are exposed to light become insoluble and adhere strongly to the underlying surface, and the parts which are screened by the black parts of the photograph are unchanged and can be washed away later to leave the corresponding areas of metal surface unprotected. These unprotected areas can then be marked by suitable inks or chemical etchants so as to reproduce the original photograph.

With full-tone photographs consisting of many shades of grey, this process cannot be applied in a direct manner since the etch resist is either present on the surface or it is absent. It cannot be partially present to give limited protection to the surface against subsequent chemical attack.

To overcome this problem, a gridded screen is normally used to break up the light passing through the photograph into a large number of small black and white areas. This causes the etch resist to remain soluble in places and later to be washed away as a large number of small pits.

In rotogravure, depending on the particular process, the surface density of the number of etch pits subsequently produced or the depth of the pits is roughly proportional to the density of the part of the photograph which overlayed them; but there is no direct positional relationship between the silver grains on the original photograph and the pits which later form in the corresponding area of the etched plate. In addition, screens having 200 lines per inch are about the maximum which can be used and, even for this level of resolution, considerable skill is required on the part of the engraver. As discussed earlier, the technique of using photographs toned with certain

radioisotopes permits the production of relief images of much higher fidelity, which raises the question whether these processes could be advantageously applied to photomechanical reproduction.

In lithographic printing, screens and etch resists are employed in a similar manner to rotogravure but, instead of etching macroscopic pits in the underlying surface, its chemical properties are changed so that the image areas can be wetted by a grease-based printing ink. The remainder of the surface remains hydrophilic. In Appendix C, the old method of preparing trimetallic lithographic printing plates is compared with a hypothetical new process using fission track etching.

The main factor inhibiting the direct application of these intaglio images to rotogravure type printing is the small volume of the individual pits. When etching a material such as glass in dilute hydrofluoric acid, the etch pits increase in size from submicroscopic dimensions until a depth of about 5 μm and a diameter of about 10 μm are reached. Thereafter, although the pit diameter continues to increase, the depth slowly decreases; by the time the etch pits are about 50 μm in diameter their depth is about 1 μm. Only plates having etch pits close to the maximum depth of about 5 μm can be satisfactorily inked. Even then, the volume of ink in each etch pit is so small that only low density images can be transferred to paper or plastics unless the ink contains a very high percentage of pigment. By using carbon black moistened with hydrocarbons and high pressure against a smooth white plastic surface, we were able to show that printing is possible using a glass plate, but problems in ink technology may be severe for any colour printing process using these small etch pits. The high pressures necessary can also cause difficulties with non-metallic plates.

Use of barrier layers to produce larger etch pits and to etch metals. One method employed to produce larger pits and to etch metals was to deposit a thin barrier layer (3 - 4 μm) of nitrocellulose or polycarbonate on the metal surface. Such layers can be easily obtained by multiple dipping in dilute solutions of these polymers (nitrocellulose dissolved in ethyl acetate or polycarbonate dissolved in dichloromethane have been found satisfactory). To obtain satisfactory adhesion of such thin layers under etching conditions, the metal surface must be very carefully prepared as in the technology of current commercial etch resists, and the edges of the plate must be protected with a resistant paint or wax.

After a suitable exposure to the californium-toned photograph, holes are etched through the barrier layer; the metal can then be etched at these

points to produce pits of any desired size. To have good control over the process, it is clearly desirable that the metal should remain unaffected by the liquid used to etch the barrier layer and should be subsequently etched by a liquid which does not attack the barrier layer. Owing to the ease with which the fine holes in the barrier layer can become blocked with gas bubbles, it is desirable to use an etchant which produces no gas. Anodic etching can also be employed.

The author has not yet obtained very good intaglio images on metal surfaces by these processes, but this may be only a problem of improved technology. It was found that barrier layers adhere best to porously anodised aluminium, but this metal is also attacked by the sodium hydroxide used to etch the barrier. On the other hand, films on copper are not yet sufficiently adherent and some attack occurs in areas which have not been exposed to fission fragments.

Problems in rotogravure would be greatly simplified if the protective oxide films which form on such metals as aluminium or zirconium could be used in place of plastics as an etch resistant barrier layer. It was not possible to etch damage tracks in any oxides formed on these two metals by anodising or by heating in air; however, there was some indication that this goal could be achieved. As a last resort, samples of aluminium which had been used in a separate experiment to study aqueous corrosion were tested. The samples had been exposed to circulating, oxygenated water (pH5) for several weeks at 50°C and quite a thick transparent oxide had built up on them. When these samples were exposed to fission fragments from californium-toned test patterns, the patterns could be clearly etched in the oxide layer with a phosphoric-chromic acid etchant. Transitory patterns were also observed in anodised titanium when anodically etched in ammonium bifluoride solution.

Since damage trails in silica can be easily etched, it is probable that oxide barrier layers on silicon chips, which are important in integrated circuit components, could be etched in a similar manner leading to possible improvements in this important branch of technology.

5. RESULTS AND DISCUSSION

5.1 Increasing Photographic Speed

Experience over the last few years at AAEC Research Establishment, Lucas Heights, has shown that intensification by autoradiography is much superior to chemical intensification on films having low fog levels. On such films, and with high contrast subjects, it has been possible to reduce the level of

exposure by a factor of fifty and still produce reasonable prints both with visible light and with industrial type radiographs. Some of these results are shown in Figures 1 and 2.

Petrakev & Dimitrov (1968) have claimed that detection limits in emission spectrography can be lowered by chemical intensification of the plate, and Hayward (1970) has examined several image enhancement techniques for extracting more information from photographic negatives. Because the main aim of the program was to study the radiochemical aspects of the technique and factors affecting autoradiography of photographs, negatives having low fog levels were obtained using medium speed film, usually Ilford Ortho (ASA80) or Ilford F-type X-ray film. Admittedly, the use of such films is not a fair test of the use of this technique to increase photographic speed. High speed films employed with high contrast developers could probably have obtained good photographs with very little intensification at the exposures employed.

When it was finally studied, the intensification of these very fast films by autoradiography proved to be no better than some methods of chemical intensification and, in some cases, the results were worse. This was really not surprising since all these fast films, employed without strict adherence to the principles listed in Appendix E, produce very high levels of fog (usually having densities greater than 0.2) and any image must be rather dark initially to show above this fog. Only a small intensification factor, perhaps X5, is required to make the negative of acceptable density and chemical intensifiers are quite capable of this.

The occasional poorer performance of autoradiography with such negatives probably occurs because the fog takes up a disproportionate amount of the radioisotope (which is usually present in the toning solutions in low weight concentrations) leading to lowered contrast in the autoradiograph. This problem would not arise with neutron activation of the image. The adaptation of this procedure or increased concentrations in the toning solutions, together with close attention to all the factors listed in Appendix E as contributing to fog, would allow autoradiography to provide a useful speed increase even with conventional fast films, but chemical intensifiers could probably be developed to provide the same service more conveniently.

The main hope of applying the technique to produce large increases in speed must rest with the development of films having very low fog levels, yet retaining reasonable sensitivity. As discussed in Appendix D, films are available which will record densely ionising radiation with almost 100 per

cent efficiency and produce zero fog (Bishop & Saha 1968). How closely these results can be approached for radiations of lower ionising power is still not clear and constitutes part of the still evolving art of the emulsion maker (Shepp 1967, Graham 1972).

It is important to remember, however, that detection limits in many systems are not fixed by the inadequacies of the emulsion but by competitive radiation from non-image sources. From experience with astronomical photographs and spectrographic plates, we have found that detection limits are often set more by background radiation such as airglow, galactic background, spectrographic source continuum, *etc.*, than by emulsion fog. Methods for minimising such interferences are suggested in Appendix E.

For visible light recording it may not be possible to improve much on the speed of current fast films (Bird 1971); but in all forms of radiography using ionising radiation or in autoradiography, where silver halide film responds in a completely linear manner to exposure in both cases, there is no theoretical reason why films cannot be developed which function virtually as counters of individual photons or particles. If this becomes possible (and it is already possible for densely ionising radiation such as alpha particles or protons - see Bishop & Saha 1968) methods of intensification will make possible the construction of useful pictures from only a few thousand information grains per cm^2.

Chemical and autoradiographic techniques will be in competition to intensify such photographs. To give a reasonable picture at an image grain density of 10^4 cm^{-2}, each grain must be increased in diameter from about 2 μm to > 50 μm. This represents an increase in grain volume of > 25^3, which is far beyond even multiple applications of conventional chemical intensifiers. With autoradiographic intensification (provided the particle range extends far enough) this increase in apparent grain diameter is achieved merely by controlling the exposure time. Recently, new types of physical developer have been described (Jonker 1969) which can be used to intensify photographs by the continuous deposition of silver on preexisting grains, and it is possible this process will again make chemical intensification competitive with autoradiographic intensification in these applications.

Whichever method of intensification is employed, such photographic systems will be in competition with electronic systems of two-dimensional photon counting such as image intensifiers and gamma ray scintillometers. Recent developments in the technology of the electronic camera may also make the two systems complementary (Decker & Mostwerdt 1969).

In general, the amplification factor of electronic systems cannot be equalled by photographic systems, but the latter systems are much more competitive with regard to resolving power and capital cost.

5.2 Future Applications

The full benefits of autoradiographic intensification (*i.e.* its ability to convert a photographic plate into a two-dimensional counter for particles or photons) can be realised only for densely ionising radiations with emulsions currently available. The most obvious and useful applications are in the fields of neutron radiography and in autoradiography; in each of these applications exposures could be very greatly reduced if suitable films were chosen, and the silver grains liberated by individual particles were intensified to visible dimensions. Several possible applications of the pinhole camera with ionising radiations are discussed in Appendix D, but the field of greatest importance is that of medical radiography.

Diagnostic radiation dose is closely related to the speed of available X-ray films. Corney & Splettstosser (1951) have pointed out that a one-stage chromium deposition process can frequently be of value in medical radiography by removing the necessity for a second radiograph of a seriously ill patient when an error in exposure or development occurs. Krug (1967) has claimed that the use of thinner films and chemical intensification would lead to great economies of silver and processing time in medical radiography. In view of the seriousness with which the radiation dose to the general population from diagnostic X-rays is now viewed (Weeks 1971), it is surprising that these ideas have not been much investigated.

Current technology of medical radiography has been based almost entirely on the use of fluorescent salt intensifying screens in conjunction with very fast blue sensitive film. This system produces high fog levels. Under-exposed radiographs at these fog levels can be adequately intensified by conventional chemical intensifiers, but only very modest exposure reductions are possible for an appreciable expenditure of time and effort. This system is currently under challenge from image intensification systems but still remains highly competitive. It is a matter for conjecture whether new types of colloidal metal screens (Bayol 1972), and low fog films loaded with heavy metals (as discussed in Section 3.3) and intensified by autoradiography will prove competitive in this field with the two established systems.

If the intensification procedure should become established on a large scale in any branch of radiography, it will be necessary to use the safest and most convenient way of radioactivating the image. Although it is

possible to conceive fully enclosed and automatic units for toning the photograph and making the autoradiograph, such units are intrinsically more complex than a neutron irradiation facility. As mentioned previously, conventional photographic materials, irradiated with neutrons, produce high fog (from chlorine-38) when one attempts to use silver-108 (2.5 min. half-life) to prepare the autoradiograph. These conventional materials are also prone to radiation damage when silver-110m (253 days half-life) is used. Recently however, photographic plates have been prepared in which the silver halide is supported in an anodic oxide layer on an aluminium plate (Fayyari 1967). Another process permits diffusion transfer of a photographic image to an aluminium surface while development is occurring (Wainer 1967). Such photographs would be completely resistant to radiation damage and, if the process could be extended to beryllium surfaces, it should be possible to autoradiograph the short-lived silver-108 isotope with very little interference from activated impurities.

5.3 Production of Relief Images and Applications to Photomechanical Reproduction

It is the contention of this report that a photograph toned with californium-252 (and, to a lesser extent, with alpha emitters such as polonium-210) is a completely new type of tool which makes possible a new range of technologies. In contact with such a photograph emitting densely ionising radiation, all surfaces become photoreceptive whereas previously only certain special materials have been photoreceptive, *e.g.* silver halides, blue print paper, diazo type paper *etc.*

In the surface which is contacted, the photograph is exactly reproduced in the form of a radiation damage pattern which will have different electrical, magnetic and chemical properties, possibly leading to applications which have not yet been envisaged. The image is also slightly radioactive by virtue of embedded fission products and, although the surface appears unchanged, it can be used to produce further autoradiographs. This property may have some applications in military or commercial intelligence.

The changed chemical properties of surfaces exposed to the photographs are most easily demonstrated by etching and very decorative etchings on glass and plastics have been produced by the technique. Relief replicas of these etchings in precious metals could be applied in high class jewellery manufacture.

It seems highly probable that the changed chemical properties of the radiation damage image could also be made to manifest themselves in the form

of differential 'wettability'. (Polyethylene sheets are currently irradiated with electrons to make them 'take' printing ink.) This would be the most simple system of lithographic printing which can be envisaged.

It is in the printing industry that the widest applications of the technique can be expected. Gravure printing from etched glass plates has already been demonstrated, and crude images have been formed on trimetallic printing plates used in lithography.

Since exact metal replicas of the intaglio images can be enhanced by electrodeposition techniques (Despic & Popov 1971), off-set printing can also be envisaged, but this has not yet been attempted. A more detailed comparison of current methods of manufacturing printing plates with a hypothetical method using autoradiography is given in Appendix C.

5.4 Interaction of Nuclear Science and Photography

It is clear that the long history of mutual interaction between photography and nuclear technology, which began in 1896 when Becquerel observed the fogging of photographic film stored close to compounds of uranium, is by no means ending with the work described in this report. The author has been continuously surprised at the rate at which these subjects interact to generate new ideas. There has been little time to test most of these experimentally but a few are mentioned here to stimulate further consideration.

5.4.1 Autoradiography without radioactivity

On the simplest possible model of a photographic negative, it can be readily calculated that it requires about 10^6 grains having a diameter of 1 μm in each cm^2 of film to cause a 1 per cent drop in the transmission of a beam of light. It is because light transmission is the usual method of extracting information from photographs that very high grain densities are usually required. Although low grain densities cannot normally be employed in photography, we have shown that autoradiography permits their utilisation. However, any other method which makes the silver image emit radiation should be theoretically equivalent to radioactivation and most such systems would be much less hazardous.

(a) *Image fluorescence.* It was suggested (J.S. Watt, private communication) that conversion of underexposed images to compounds of silver which fluoresced when illuminated with ultra-violet light or X-rays would permit the image to be re-photographed and intensified in a manner analogous to autoradiographic intensification.

Ultra-violet stimulation of fluorescein and several other dyes absorbed on photographic images was attempted but without success. The dyes ceased to

fluoresce as soon as they became adsorbed and time was not available to extend the search.

(b) *Catalysis of chemiluminescence.* The reactions of several organic compounds in solution are accompanied by the emission of visible light. The most outstanding example of such a reaction is the oxidation of luminol (1:4 phthalazinedione 5-amino 2:3 dihydro) in aqueous alkaline solutions which, under certain conditions, produces an emission of blue light which is clearly visible to the naked eye (Courmier & Pritchard 1968, Lee & Salinger 1972). Hydrogen peroxide and halogens are the most common oxidants but the material is also slowly oxidised by atmospheric oxygen. The reactions are intensely catalysed by traces of certain transition elements in solution (Seitz 1972).

There is some evidence that the atmospheric oxidation of luminol can be catalysed by photographic silver images. For example, when a developed photograph and an unexposed film were both soaked in a dilute alkaline solution of luminol, and then pressed together (emulsion to emulsion) for several hours, crude autoradiographs were obtained when the film was developed. It is not suggested that this could ever develop into a method for making acceptable autoradiographs, since the greater spread of visible light under these conditions would always give poor resolution. However, if free-standing photographs in luminol solutions could be rephotographed while producing catalytic light emission, a satisfactory method of photographic intensification which did not employ radioactivity might be developed.

5.4.2 Use of californium-toned photographs to test neutron recording systems in neutron radiography

When making fast neutron radiographs,considerable difficulty arises in recording the neutron image and several systems are currently being evaluated (Berger 1970, Vasilik *et al.* 1972). Since intricate test patterns can be toned with an exactly measurable amount of californium-252, the neutron emission rate can be calculated and autoradiographs prepared from these neutrons should permit a rapid comparison of the resolving power and efficiency of various systems for registering fast neutron images.

5.4.3 Autoradiography and xerography

Almost all the work in this report has been concerned with silver halide systems of photography, but in many fields xerography now provides a competitive system for rendering images permanent. In this system, the incident photons expel electrons from a suitable semiconductor surface and produce an electrostatic image which is then rendered visible by its attraction for very fine grains of pigment. It is obvious that the use of a radioactive pigment

(*e.g.* radioactive carbon black [^{14}C]) would produce immediately and very easily a radioactive image which could be autoradiographed if underexposed. The autoradiography could also be performed on xerographic surfaces.

The use of this system, or pigments activated with californium, might also prove more rapid and convenient in any commercial system using fission track autoradiography to prepare intaglio images (see Section 4).

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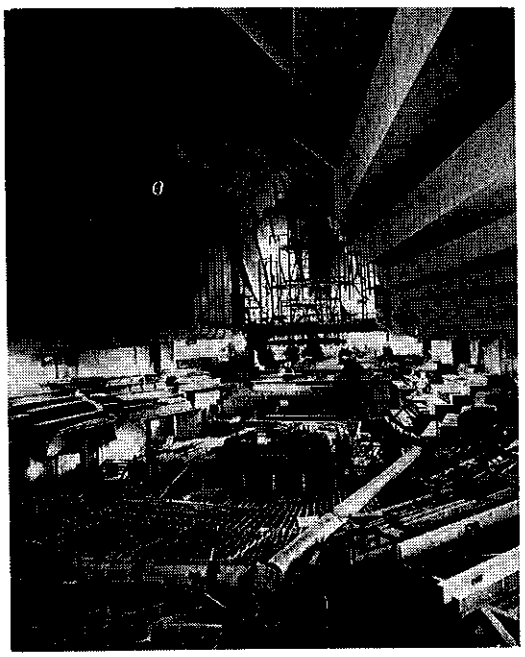
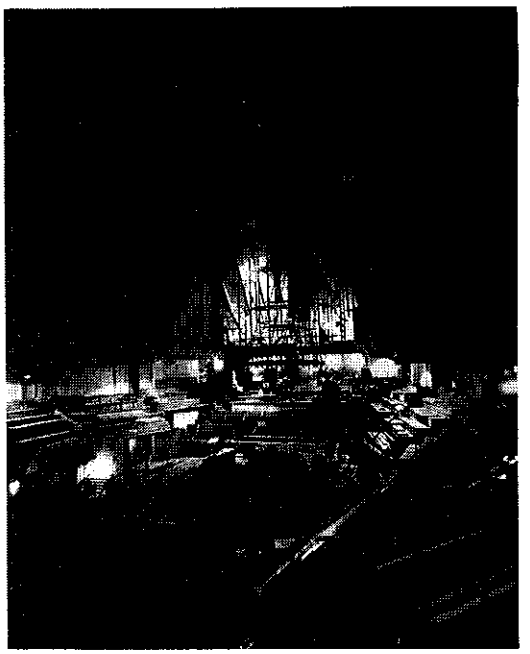
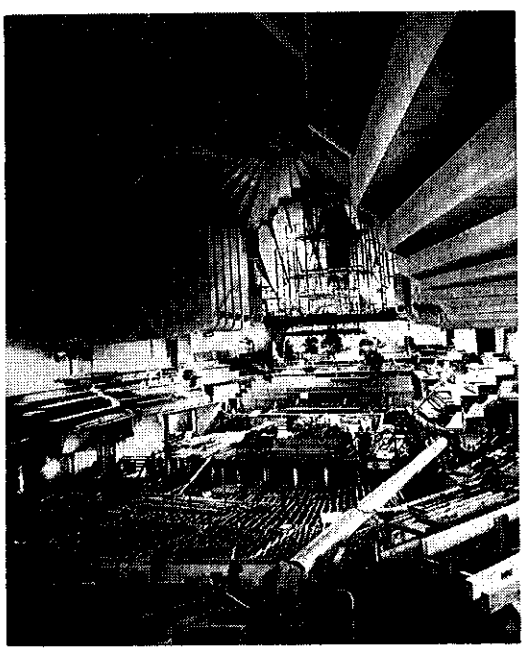


(a)



(b)

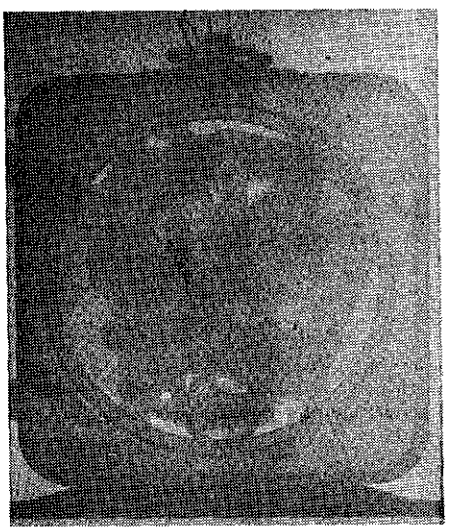


(c)

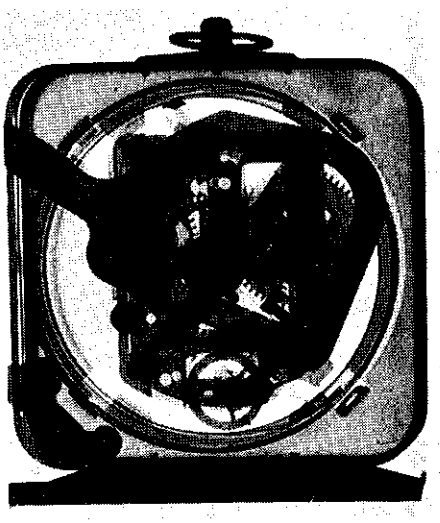
THE CONCERT HALL OF THE SYDNEY OPERA HOUSE PHOTOGRAPHED IN LATE 1971.
FIGURE 1(a). NORMAL EXPOSURE USING ILFORD HP4,400ASA FILM, *f* 8 AT 10 s.
FIGURE 1(b). TAKEN ON THE SAME FILM, EXPOSURE *f* 8 AT 1/5 s.
FIGURE 1(c). AN AUTORADIOGRAPH PREPARED WITH ^{35}S FROM UNDEREXPOSED NEGATIVE 1(b).

FIGURE 1. INTENSIFICATION BY AUTORADIOGRAPHY USING AN AUTORADIOGRAPH PREPARED FROM AN UNDEREXPOSED NEGATIVE

(a)



(b)



(c)

FIGURE 2(a). RADIOGRAPH PREPARED WITH OPTIMUM EXPOSURE ON ILFORD F TYPE X-RAY FILM.
FIGURE 2(b). RADIOGRAPH PREPARED WITH 2% OF OPTIMUM EXPOSURE ON SAME FILM.
FIGURE 2(c). AUTORADIOGRAPH PREPARED FROM RADIOGRAPH WITH 2% EXPOSURE (ILFORD F TYPE X-RAY FIIM) EXCHANGED ON ONE SIDE WITH ^{110}Ag AS AgBr, TRANSFERRED SAME FILM TYPE.

FIGURE 2. INTENSIFICATION BY AUTORADIOGRAPHY USING AN AUTORADIOGRAPH PREPARED FROM AN UNDEREXPOSED RADIOGRAPH

APPENDIX A

PROPERTIES OF ISOTOPES COMMONLY USED FOR RADIOTONING AND ILLUSTRATIVE TONING PROCEDURES

A1. SULPHUR-35

E_β = 0.167 MeV $t_{½}$ = 87 days

Range = 130 µm

ADVANTAGES

- A well-known toning element that can be applied directly to silver images as a polysulphide ion in a one-stage process.
- Cheaply available as a by-product from the production of phosphorus-32.
- No gamma radiation.

DISADVANTAGES

- Sulphides and polysulphides are subject to atmospheric oxidation. Radiolytic oxidation of solutions of high specific activity is rapid. Fresh reagent must be prepared frequently and wastage is high.
- Solutions smell of hydrogen sulphide and close attention to pH is necessary to prevent evolution of radioactive gas.

TONING

Irradiated sulphur (approximately 1 g) is freed from any residual phosphorus-32 by melting several times under fresh lots of phosphoric acid. The solidified sulphur is washed with boiling water and dried. The sulphur is then powdered and mixed in 1:3 ratio with anhydrous potassium carbonate. The mixture is heated to about 280°C in a paraffin bath under a stream of nitrogen. After cooling, the residue is taken up in a 2 per cent solution of K_2CO_3 and stored under nitrogen. This stock solution should be free from turbidity when used and may require filtration after long standing.

A volume of 2 per cent K_2CO_3 solution sufficient to cover the film is mixed with polysulphide-S35 solution. About 1 µCi of sulphur-35 is used for each square cm of film to be toned. The solution is stirred in contact with the film for about one hour. The film is then washed with 2 per cent K_2CO_3 then with an inactive polysulphide solution of similar concentration to the one used for toning for about 5 min., and again with 2 per cent K_2CO_3 solution. Exposure to the atmosphere,or solutions of lower pH before washing is complete,

may give rise to precipitation of active colloidal sulphur in the gelatine. Prolonged exposure to inactive polysulphide solutions will remove activity from the image. The film is finally washed in running water and dried.

A2. POLONIUM-210

E_α = 5.3 MeV $t_{½}$ = 138 days

Range = 33 μm

ADVANTAGES

- High activities are cheaply available.
- Very high specific activity.
- Autoradiographs can be obtained very rapidly from the faintest images.
- No gamma radiation.
- Strong affinity for silver permits single stage toning.

DISADVANTAGES

- High biological toxicity.
- Any silver fog is greatly intensified.
- Alpha energy is too high for high resolution.

TONING

Polonium deposits onto silver images from dilute nitric acid solutions at ~ pH 2. About 0.1 μCi for each square cm of film is usually adequate. After 2 hours' stirring, the film is washed three times with dilute nitric acid and then with running water. Owing to the high specific activity of polonium and its strong affinity for silver, it is necessary to wash the film further with a solution capable of complexing polonium and having some solvent action on the silver fog. A 2 per cent solution of thiourea adjusted to ~ pH 2, usually with oxalic acid, is one of the very few solutions we have found to be satisfactory for this purpose.

The removal of activity from the gelatine and silver fog can be followed at intervals by counting a disc of unexposed film which has been developed and processed in parallel. Several washes with thiourea solution may be necessary to reduce the activity of the control disc to an acceptable level. The film is then washed in running water and dried. At this stage it is possible to remove all trace of the visible silver image with cyanide solution without diminishing the activity of the polonium image.

A3. NICKEL-63

E_β = 0.062 MeV $t_{½}$ = 85 years

Range = 25 μm

ADVANTAGES

. Soft beta emitter capable of fairly good resolution.

. No gamma radiation.

. Known toning reactions.

. Sensitive test for gelatine residue (dimethylglyoxime).

DISADVANTAGES

. Required specific activity is expensive.

. Long half-life and uncountable beta emission creates a hazard.

TONING

An exchange procedure proved most convenient for the high specific activity nickel-63 used (16.6 Ci g^{-1}). The photographs were first toned with inactive nickel.

The following stock solutions are prepared:

Solution A 10 g of potassium ferricyanide and 20 g of sodium citrate are dissolved in 1 litre of water.

Solution B 10 g of nickel sulphate (hydrated) are dissolved in 1 litre of water.

One part of solution B is added slowly with stirring to four parts of solution A until a clear green solution is obtained. The photograph is treated with this solution until the image acquires the characteristic yellowish-green colour of nickel ferrocyanide. The photograph is then washed briefly in running water and then twice for periods of 5 min. in a 2 per cent solution of sodium citrate brought to a pH of 2 with sulphuric acid. The picture is then transferred to a further portion of the acidified citrate to which nickel-63 solution has been added at the rate of about 1 μCi cm^{-2} of film. The solution is then stirred for several hours to allow exchange to occur. The photograph is then again washed in acidified citrate solution and in running water before drying.

A4. CALIFORNIUM-252

E_α = 6.1 MeV $t_{½}$ = 2.65 years

Alpha emission 96.9%

Spontaneous fission 3.1%

ADVANTAGES

. Fission fragment emission allows intaglio autoradiographs to be prepared on a large variety of surfaces.

. Fast neutron emission is useful for testing neutron imaging and recording systems.

. Concurrent alpha emission allows sensitive measurement of quantities

by simple counting.

- Fission fragments embedded in an adjacent surface permit reproduction of the image in a β - γ active form.

DISADVANTAGES

- Highly toxic alpha emitter.
- Fast neutron shielding is required.

TONING

The negative is first hardened with formaldehyde and then shaken with a 5 per cent solution of potassium ferricyanide to convert the image to silver ferrocyanide. The negative is thoroughly washed in running water and then allowed to stand in a solution containing californium chloride for about 4 hours. The pH of the californium solution should be adjusted to a value of 2.0 with nitric acid and it should contain about 1 microcurie of californium-252 for each cm^2 of film which has to be treated. As californium deposits from solution, the photographic image slowly turns blue and this is a useful indication of the extent to which the reaction has occurred. The photograph is then rinsed with demineralised water and washed for about 30 min. in running water. Negatives having high fog levels are much improved by a few minutes' treatment in a 0.1 per cent solution of thiourea adjusted to pH 3 with oxalic acid before the final water wash. The photograph is then dried.

A5. SILVER-110m

E_β = 0.087 MeV (59%)	Range = 48 µm	$t_{½}$ = 270 days
E_β = 0.53 MeV (35%)	Range = 700 µm	
E_β = 2.86 MeV (3%)	Range = 5 mm	

ADVANTAGES

- Reaction is by exchange rather than toning and this is sometimes useful.
- Conversion of the image to AgBr can proceed concurrently with silver-110m exchange as an efficient one-stage process which also permits some fog reduction.
- High specific activity is fairly cheap.
- Lightly exposed thin emulsions can give fairly good resolution *via* 0.087 MeV beta emission.

DISADVANTAGES

- Hard gamma emitter.
- Solutions are degraded rapidly by exchange and wastage is high.

. High beta energies give poor resolution on heavily exposed autoradiographs especially with double-coated X-ray emulsions.

TONING (Exchange Procedure)

A photographic bleach solution is prepared to convert the image to silver bromide. A typical composition is:

Potassium ferricyanide	10 g
Potassium bromide	5 g
Potassium carbonate	10 g
Water to make	1 litre

High specific activity silver-110m is added to the bleach solution (sufficient to cover the photograph) in the form of silver nitrate solution. The solution is stirred until it becomes completely clear before immersing the photograph. Stirring is then continued for a further hour. If the photograph shows high fog levels it is improved by stirring for several hours. The photograph is rinsed with demineralised water and washed in running water for about half an hour. The picture can be reduced back to metallic silver at this stage to restore its original appearance by re-development after which it is again washed and dried.

APPENDIX B

EXPONENTIAL RADIOACTIVE DEVELOPMENT

(CONCURRENT AUTORADIOGRAPHY AND DEVELOPMENT)

If the silver image can be toned with a radioactive material while development is proceeding (Section 2.5), the radiation from the deposited toner will immediately begin to affect the neighbouring silver halide grains making them developable. A situation arises whereby:

- The more silver develops, the more activity deposits.
- The more activity deposits, the more silver develops.

Under such conditions, development should show an exponential increase with time, and high intensification of underexposed photographs may be possible. Radioisotopes having very soft radiations would be necessary for the application of this process so that radiation from the bulk of the solution did not produce appreciable fogging of the film. The use of tritiated dye-couplers would be the most likely method to succeed, but these are rather expensive.

In cases where a photograph is known to be underexposed, the application of this intensification technique could save a great deal of time, although it is difficult to see how exponential development could be controlled for panchromatic films which must be developed in total darkness. Some sort of infrared viewing system would seem to be required for panchromatic films.

The following systems which have been employed to colour photographic images may be adaptable to activating the developing image:

(a) Precipitation of insoluble developer oxidation products

When the silver halide grains which contain latent image specks are developed, silver ions are reduced to metallic silver. The developer molecules which bring this about are themselves oxidised and if the oxidation product has lower solubility, it may precipitate in the silver image. Several organic developers, *e.g.* pyrogallol, are known to produce brown organic images concurrently with the production of silver images.

Ferrous oxalate and titanous oxalate developers can also be applied under conditions of pH where ferric hydroxide or titanium dioxide precipitate in the image during development (Glafkides 1960 p.585).

(b) Coupling of developer oxidation products with other compounds

This is the basis of all modern systems of colour photography. The developer oxidation product (usually a derivative of p-phenylene-diamine) will

react with a number of phenols and amines, also present in the film or developer, to precipitate an insoluble coloured compound (Glafkides 1960 p.535).

(c) Adsorption of other materials onto insoluble developer oxidation products

The insoluble image materials deposited as in (a) or (b) can also react with or adsorb further constituents of the developer. The ferric hydroxide and titanium oxide used in (a) can act as mordants for many organic dyes. These materials can also act as carry-down precipitates for many carrier-free radioactive ions in solution as well as precipitants for some ions such as phosphate present at higher concentrations (Glafkides 1960 p.540).

APPENDIX C

COMPARISON OF CURRENT PRINTING PROCESSES WITH AUTORADIOGRAPHIC PLATE MANUFACTURE

The two principal methods currently employed for the reproduction of photographs in the printing industry are rotogravure and lithography. The high speed of the rotogravure presses makes it necessary that the printed image should dry almost instantaneously and, to this end, a 'weak' ink consisting largely of petroleum ether is used. Because the ink is weak, quite large etch pits are required in the copper cylinders (up to 35 μm deep).

It is difficult to produce such large pits by the fission track etching technique, and to do so fails to use the high resolution of which the process is inherently capable. However, local gravure managers consider that the current technology for producing etched copper cylinders is not very satisfactory, and they are continuously seeking new processes which would make it more controllable and reproducible. In addition, they seem to be expecting legislation in the next few years to prohibit the high pollution and fire hazard caused by large quantities of petroleum. Such legislation would lead to the use of stronger, waterbased inks and a complete change in technology. Although the gravure managers were intrigued by the much greater simplicity of the autoradiographic process for producing etched cylinders, it seemed unlikely that they would change their current methods until compelled to do so by legislation.

In lithography, the printing ink is not retained in etch pits but on areas of the surface, which can be wetted by a greasy ink but not by water. Because the presses run more slowly and because the image is transferred from the plate to the paper by means of a rubber roller, 'weak' quick drying inks are not required. Stronger inks are compatible with smaller printing areas and with high fidelity images. It is, therefore, to the field of lithography that the new process could be more directly applied.

C1. PREPARATION OF A TRIMETALLIC LITHOGRAPHIC PRINTING PLATE

C1.1 Conventional Process

The plate usually consists of a thin layer of chromium on a thicker layer of copper supported by a steel or aluminium base plate. The chromium surface is cleaned with 2 per cent phosphoric acid, washed and dried. It is then coated with an even layer of etch resist which is baked on. A gridded screen having 150 lines per inch is placed in contact with the etch resist

and a photographic transparency is placed on top of the screen in a special frame to which vacuum can be applied. The photograph is irradiated with strong ultra-violet light for about 10 minutes to render parts of the etch resist insoluble. The etch resist is then developed by means of a solvent to give a series of fine holes corresponding to areas of light and shade. The plate is dried and transferred to a special etchant which removes the top layer of chromium where it is not protected by the etch resist. The remainder of the etch resist is then removed and the plate is ready for inking. Greasy inks adhere to the copper areas and water to the chromium.

C1.2 Envisaged New Process

The trimetallic plate is cleaned with acid, washed and dried. It is coated by dipping with a thin layer of nitrocellulose or polycarbonate. The photograph toned with a suitable radioisotope such as californium-252 is pressed against the plate and exposed for a suitable period. (The period depends upon the amount of californium on the plate, the thickness of the etch resist and the amount of resolution required on the plate. For example, a mean value of 10^6 ink spots of 10 μm diameter per cm^2 will give a much better (x 100) reproduction than existing commercial processes. This level of exposure can be obtained in one hour with a californium loading of 2×10^{-8} g cm^{-2} and a barrier transmission factor of 10 per cent.) The fission tracks are then etched for about 15 minutes in a hot solution of sodium hydroxide. The plate is then transferred to a second etchant which removes the chromium only in those places where the fission tracks penetrate the etch resist. The etch resist is then removed and the plate is ready for inking.

The two processes as here envisaged are very similar and would differ little in cost. Any extra cost of the isotopic process would arise from the toning procedure and from radiation safety aspects. A possible saving in the isotopic process would be the development of plates which did not need coating with an etch resist if an oxide layer on chromium could be made to fulfil this function.

C2. THE TONING PROCEDURE

C2.1 Californium

The photographs are first bleached to silver ferrocyanide in a 5 per cent solution of potassium ferrocyanide. They are then thoroughly washed and transferred to a solution of californium acidified with nitric acid to pH 2. The photographs are agitated with the californium solution for about an hour and again thoroughly washed and dried.

It is envisaged that the californium would be contained in some form of

hot cell with remote and possibly automatic handling. Only the printing plates would need to pass in and out of the cell continuously. They would be automatically monitored for alpha activity on leaving the cell. When a satisfactory plate had been produced, the californium could be extracted from the photograph and recycled. The californium wash solutions would also be recycled through an ion exchange column (see Figure C1).

Assuming a throughput of 50 plates per week with a printing area of 4 ft^2 ($\sim$ 3,720 cm^2), a toning quantity of 7.4×10^{-5} g of californium would be required. The actual californium inventory would depend on the speed with which the californium could be extracted from the films and recycled but, even with fairly slow recycle, 1 mg of californium would probably be adequate. This could be reduced by a factor of ten if longer (overnight) exposures were permissible, by a further factor of ten if 10^5 ink spots per cm^2 were considered to give adequate resolution, and by a further appreciable factor if the californium was recycled immediately after exposing the plate.

The small quantity of californium-252 (somewhere between 1 µg and 1 mg) and the extreme simplicity of the operations (agitation of photographs with toning solution, washing and drying, contacting with lithographic plates followed by agitation with extracting solvent) should permit the design of a small compact and fully automatic hot cell.

C3. SHIELDING

The maximum envisaged quantity of californium-252 (1 mg) emits 2.34×10^9 of fast neutrons per second, but these would be somewhat moderated by the solutions in which they were dissolved.

Judging from californium neutron sources which have previously been described, about 30 cm of borated plastic and 80 cm of concrete would be required as shielding. Alternatively, the cell might be built underground.

C4. COSTS

The projected costs for californium issued by the USAEC are as follows:

Year	mid 1970s	late 1970s	early 1980s
$ per µg	5 - 7	2 - 3	0.5 - 1.5

It is expected that an initial cost of several thousand dollars for californium would be incurred but only small makeup quantities would be required after that.

The capital and operating costs of a fully automatic hot cell would also be costed to the process if the full inventory of 1 mg of californium was required. A recently completed investigation (R. Bolling, AAEC private

communication) places the total cost of such a cell facility at about $A400,000.

The current manufacturing cost of this type of lithographic plate is about $35 per plate and this would be further increased in the new process by the capital costs of the californium and the hot cell. This cannot be accurately estimated at the moment. Against this extra cost may be set the following advantages and savings:

- Much improved reproduction of full-tone photographs (*i.e.* 10^6 ink spots per cm^2 instead of less than 10^4) possibly leading to an increased share of this large market.
- Possibility of using oxide barrier layers on the metal for fission track etching, thus eliminating the tedious and difficult procedure of organic etch resists.
- Possibility of a fully automated plate production process.

C5. ALTERNATIVE PROCEDURES

Although the project has been described throughout using californium, this is not the only possible method for producing etch tracks. The following methods are also available:

- Tone the photograph with uranium and irradiate with neutrons.
- Tone the photograph with boron and irradiate with neutrons.
- Tone the photograph with alpha emitters, *e.g.* polonium-210, uranium-233 *etc.*

Whether or not these processes will become competitive with the use of californium will depend on the availability and cost of portable neutron sources and the development of materials in which alpha tracks can be easily etched.

Although health physics protection increases the cost of any process using radioactivity, the additional cost per plate, which can produce more than a million prints, may be quite trivial and could be more than offset by the much improved reproduction of full-tone photographs, and by the possibility of using entirely new and more controllable materials as etch resists (thin films of metallic oxides or silica instead of the much thicker organic compositions such as gum-dichromate mixtures).

Since bulk printing ranks as about the fourth largest industry in most industrial countries, this would be the most profitable field in which these techniques could be employed. Additional applications might lead to potentially large markets in the high quality reproduction of works of art, jewellery manufacture, the production of decorative etching on glassware, pottery and other ceramics and the reproduction of durable scales on measuring

instruments. There is also a slight possibility that the technique will find application in the field of microcircuits.

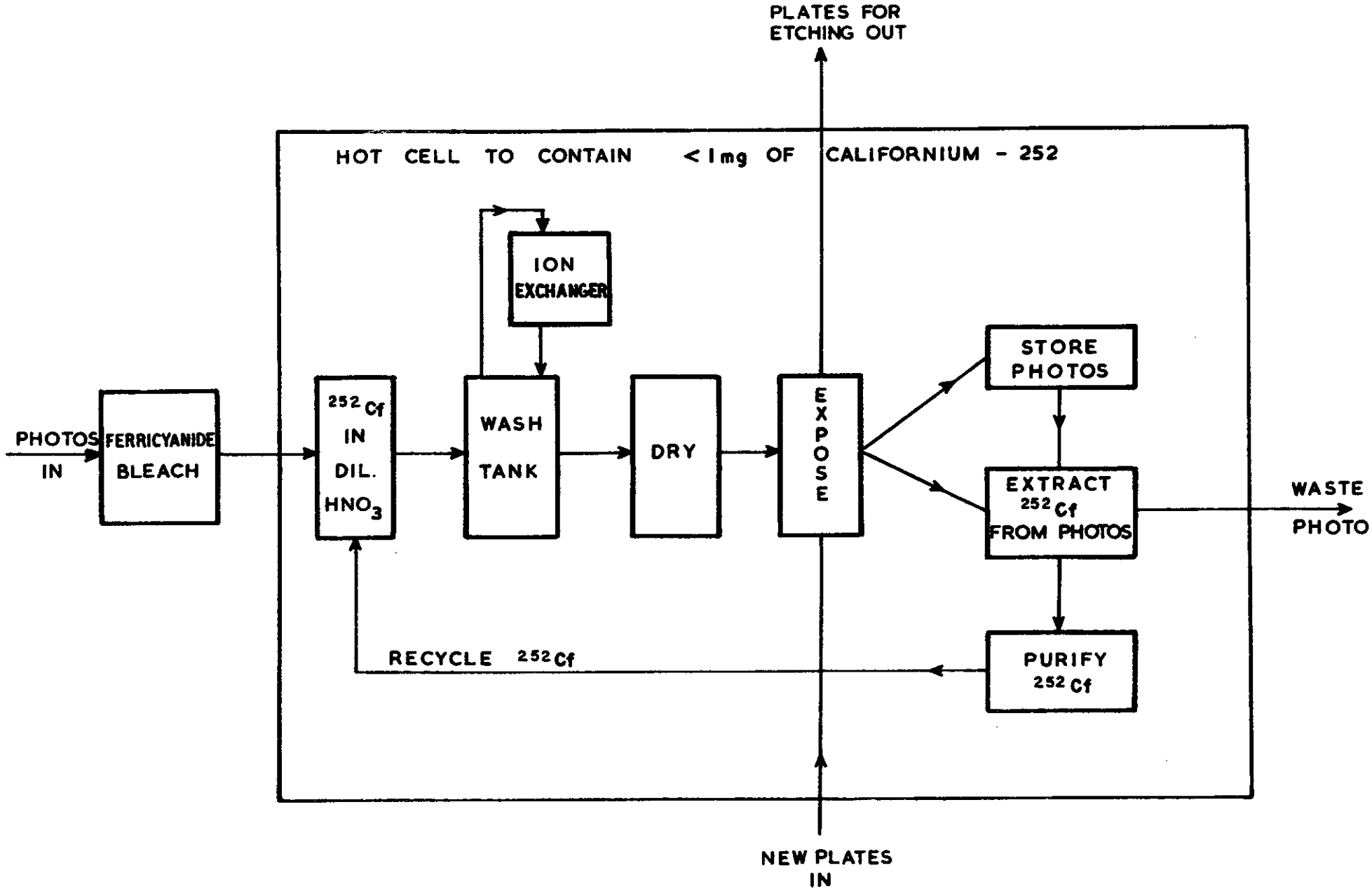


FIGURE C1. PRODUCTION OF LITHOGRAPHIC PLATES USING CALIFORNIUM-TONED PHOTOGRAPHS

APPENDIX D

POSSIBLE USES OF PINHOLE CAMERAS WITH IONISING RADIATION

Although it has been known for many years that silver halide film is sensitive to very many types of radiation, including all charged particles and a large range of the electromagnetic spectrum, the possibility of using these other radiations to take photographs with anything like the facility and sensitivity with which they can be taken with visible light is still remote.

The wide range of film sensitivity has not been fully exploited in the past for the following main reasons:

- The Earth's atmosphere is opaque to most of the radiations to which the film can respond.
- Lens and mirror imaging systems can operate over only a small range of these radiations.
- The pinhole camera, the only other practical imaging system, gives such a reduced flux at the image that long exposures are often necessary to build up a visible photographic image.

As laboratories begin operating in space and on the Moon, and as medical science becomes more skilled at marking tumours or body organ systems with gamma-emitting isotopes, the need is arising to push the pinhole camera to the absolute limit of its sensitivity.

Anyone doubting the great importance of extending the spectral range of photography would be well advised to study pseudocolour aerial photographs taken in the near infrared spectrum which are proving invaluable in the study of the thermal and chemical pollution of inland waterways, the spread of plant diseases and several other difficult problems (Grimes & Hubbard 1972). Even the crude pictures obtained by the gamma scintillation camera of internal organs labelled with radioisotopes have proved of the greatest value in medical diagnosis (Peterson *et al.* 1970).

Owing to the low flux achieved by the pinhole camera, it cannot be considered of any value until it can be operated virtually as a particle or photon counter with high spatial resolution. Each particle or photon stopped by the film must produce a mark which can be recognised with good contrast against the general background when viewed under a microscope of medium power. This problem of single particle recognition is a familiar one in the health physics monitoring of personnel exposed to ionising radiations (Agard *et al.*

1971), and also to physicists studying the dynamics and modes of interaction of nuclear particles (Barkas 1963). For all densely ionising radiations varying from protons to fission fragments, and including primary cosmic radiation (and indirectly, neutrons), the problem has been solved.

For these radiations, there is now some controversy as to whether they can be more efficiently counted using special silver halide films and developers (Barkas 1963) or by using dielectric track detectors (Somogyi & Srivastara 1971) such as nitrocellulose which are subsequently etched. The choice usually depends on the level of gamma ray background and the method by which the tracks are viewed. For neutron radiography, there is also a third method of image registration which uses a 'convertor', usually a thin metal foil which is activated by the neutrons and then autoradiographed (Mullner & Jex 1972).

Under the microscope, the tracks of densely ionising particles in silver halide films usually appear as very opaque rows of silver grains about 2 μm wide and about 10 μm long (varying with particle energy). The same tracks etched in dielectric films usually appear as holes or cones of low opacity and around 10 μm diameter. The method of autoradiographic intensification can amplify an individual silver grain until it appears as a black disc having a radius equal to the range of the beta particle used to prepare the autoradiograph (130 μm for ^{35}S). Several methods have also been described for increasing the opacity or visibility of etch tracks (Alter 1968, Sherwood 1970, Berger 1972).

The use of the two methods for recording pinhole camera images taken with densely ionising particles and neutrons is likely to remain competitive for some considerable time depending on individual circumstances.

Particles of lower ionising power, such as electrons and mesons, are not registered by dielectric track detectors, but they can be recognised as individual events in an appropriate emulsion of silver halide. The image grain to fog grain ratios are not nearly as good as those for densely ionising radiation but, using these radiations and the appropriate films, a pinhole camera would still operate almost as an individual particle counter showing spatial resolution.

For high energy photons of X-rays or gamma rays the problem is more complex. Although each absorbed photon is thought to produce more than a thousand atoms of latent image silver and render a cluster of photographic grains developable (Günther & Tittel 1933), the probability of interaction between a photon and emulsion grain is so low (see Table 1) that the film

has to be very heavily loaded with silver halide. All the silver halide can contribute to fog, but only a small fraction of it will contribute to the image. Research is still in progress to prepare films which, while retaining the same sensitivity to X-rays, resist all other fogging mechanisms (Dostes 1972). Further research is aimed at producing intensifying screens which give high amplification factors while producing low fog (Bayol 1972). As pointed out in Section 3.3, autoradiography permits heavy inert materials to be added to the emulsion which can greatly increase its opacity to X-rays without necessarily increasing fog.

While all these improvements will probably find application in conventional radiography, it seems doubtful that they will increase the signal to noise ratio of film sufficiently to allow the pinhole camera to compete with the gamma scintillometer for organ visualisation.

Any method of rendering visible pinhole camera images prepared from alpha particles, beta particles or gamma rays by means of track etching or conventional or autoradiographic intensification must, of course, be equally applicable to any autoradiographs prepared using these radiations, since the same two-dimensional patterns of radiation are involved.

As the use of the pinhole camera as a particle counter with spatial resolution obviously functions best for densely ionising radiation, and as such densely ionising radiation has very limited range in the Earth's atmosphere, most applications of the device will occur in space or on planets such as the Moon with very low density atmospheres.

Since the rapid location of indigenous ore bodies and hydrogenous materials for conversion to water would have a very large effect on the economics of manned exploration of the Moon and Mars, it is possible that the use of pinhole cameras, designed to record thermal neutrons and employing special loaded films suitable for autoradiographic intensification, could assist the location of such mineral deposits. Such cameras in orbit could survey very large areas of the planetary surface if nuclear explosions could be used as a source of neutrons.

APPENDIX E

PHOTOGRAPHIC PROCEDURES TO OBTAIN MAXIMUM SENSITIVITY WHEN APPLYING AUTORADIOGRAPHIC INTENSIFICATION TO NEGATIVES

The lower limit of exposure which can be successfully intensified is fixed by the level of silver fog on the negative. Accidental exposures to extraneous light or to darkroom safelights may completely obscure the intended exposure. The use of old films or old solutions and bad techniques for processing can also obscure images which are grossly underexposed. To prevent patchy development which would be completely invisible on correctly exposed films, developing and fixing solutions must be continuously agitated.

The following steps should be carefully followed:

1. Check all darkrooms, cameras, cassettes, *etc.*, for light leaks.
2. Conduct all film handling operations in total darkness.
3. Exclude all sources of ionising radiation from the vicinity of film store and darkroom.
4. Exclude all chemicals known to cause fogging from the vicinity of darkroom and store - volatile sulphur compounds are particularly important.
5. Use fresh film (or treat film to remove all latent image centres produced by time and cosmic rays). (Barkas 1963 p.94, Laban & Nicolae 1966.)
6. Use film designed for the job (*e.g.* 1. Do not use panchromatic film if the same information can be obtained at the violet end of the spectrum. 2. Films thicker than 2 μm will add only fog to autoradiographs prepared with tritium, owing to the short range radiation).
7. Film sensitivity can sometimes be increased just before exposure by treatment with ammonia or other chemicals (Barker 1968, Hoegner 1968).
8. Avoid abrasion, finger prints *etc.*, when loading film in clean, dry cassettes and cameras.
9. Reduce all non-image radiation when making exposure, *i.e.*
   (a) use a lens hood and appropriate filters;
   (b) minimise scatter of X-rays by air and adjacent objects;
   (c) use the correct X-ray energy for the radiograph (err on the high energy side not the low).

(d) take astronomical photographs from above the atmosphere whenever possible; and

(e) take all necessary steps to reduce inter-line continuum in spectrography (Clegg 1972).

10. Develop the film immediately after exposure.
11. Develop at the lowest convenient temperature.
12. Use a non-fogging developer and continuous agitation.
13. Use the same optimum development time for an underexposed film as for a correctly exposed one.
14. Develop an identical unexposed film as a measure of developer fog.
15. Use a stop bath and wash well before fixation.
16. Use a fixer which cannot produce silver sulphide with AgBr emulsion (decomposing acid hypo can!) Use continuous agitation. Double fixation is beneficial.
17. Fix an identical unexposed film as a measure of fixer fog.
18. Treatment with a hypoeliminator will improve results from some methods of radioactivation.
19. Prolonged washing in running water is necessary to remove all processing residues which can increase fog on the autoradiograph.