UNITED STATES DEPARTMENT OF THE INTERIOR

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NEUTRON INDUCED AUTORADIOGRAPHY, NEUTRON RADIOGRAPHY & NEUTRON INDUCED LUMINESCENCE OF LITHIUM AND BORON IN GEOLOGIC SPECIMENS

By

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Copies of this report can be made at the USGS libraries of the National Center, Reston, VA; Denver Federal Center, Colorado; and the Western Region Office, Menlo Park, California. The Books and Open-File Report Section, P. O. Box 25425, Denver, CO 80225 can also provide color-Xerox print-replicas of Figures 1, 2, 3, 10-A, -B, -C, -D and -E in addition to a text copy. The U.S. Geological Surveys Photographic Library; MS 914, Denver, CO 80225 can provide color slides of these or true-color 4x5" photographic reprints of Figures 10-A, -B, -C, -D, -E and duplicate electron micrographs shown in Fig. 4-A, 4-B.

ABSTRACT

Neutron induced-particle-autoradiography can be used to indicate the abundance, localization and distribution of lithium and boron in geologic hand-size samples. Neutron radiography is also used to graphically illustrate neutron absorption and scattering in these and other elements as they occur in thick, sawed sections of rocks and mineral specimens. Black and white photographic and x-ray type film, Polaroid and color films are used in addition to the new plastic polymer CR-39 that has recently become available for particle track recording. Neutron-induced autoradiographs are produced both from particle exposure and from luminescence of lithium-bearing phenocrysts in the spodumene specimen. Use of color film with various particle absorption techniques including foil, screens, and film-backing plastic have helped resolve and illustrate the luminescence of the phenocrysts as a separate phenomenon in addition to particle autoradiography. Collimated neutrons from a reactor beam-port and uncollimated fast and thermal neutrons from a portable californium-252 neutron source have been used to produce lithium and boron autoradiographs, neutron radiographs and neutron induced luminescence in these studies.

INTRODUCTION

The future need of lithium for new energy related uses has prompted a reappraisal of the geologic occurrence of lithium (Vine, 1976), its resource potential, and research for new methods of detection and analysis. The need and use for boron are expected to rise in the future although present resources are believed to be adequate for several centuries (Smith et al., 1973). Although the chemical and physical properties of these two elements do not provide a simple means to determine their elemental distribution, neutron induced autoradiographs and radiographs can be used to illustrate their abundance and localization in geologic specimens. Applications of induced-particle autoradiography to lithium distribution in geologic specimens have been reported by Dooley (1979) and Dooley, et al. (1979).

Neutrons, because of their lack of electric charge readily pass through thick solid objects. After moderation by scattering, some neutrons are captured by absorption in the rocks and minerals. From the prompt nuclear reactions $^{6}Li(n,\alpha)^{3}H$ in lithium-bearing minerals and $^{10}B(n,\alpha)^{7}Li$ in boron-bearing minerals, the low energy alpha and triton particles along with the ^{7}Li recoil nuclei can be used to produce photographic darkening and a particle track image for autoradiography. Unlike most radioactive decay that takes place after neutron activation, these particular prompt nuclear reactions for lithium and boron have to be recorded during the neutron irradiation in order to obtain a permanent autoradiographic record. The photographically exposed areas in the autoradiographs show the lithium and boron distribution in the geologic specimens. Such neutron-induced autoradiographs are analogous to autoradiographs produced by the natural radioactive decay in uranium and thorium bearing minerals. The alpha particles produced by radioactive decay are simply recorded by a photographic film in contact with the surface of the specimen.

In contrast, a neutron radiograph is similar to an x-ray photograph. Neutrons pass through a thick object similar to x-rays, but their attenuation is a function of the neutron absorption and scattering characteristics of the nuclei rather than atomic density of the object. However, neutrons will show little reaction with a photographic film. To detect the neutrons emerging from the object, they must be converted into some type of radiation which will be recorded by the film. For this reason a thin metallic foil, such as gadolinium which has a high neutron cross section and an electron capture mode of rapid radioactive decay, is placed next to the film detector. If the foil and film are placed in close contact there is little loss in resolution and a good neutron radiograph can be obtained.

Neutron radiographs of a relatively thick geologic specimens containing lithium and boron will differ in detail from neutron-induced particle autoradiographs. Particle autoradiographs will record only that lithium and boron which is situated close to the surface and in contact with the film. A neutron radiograph will be affected by the presence of these elements throughout the depth of the samples and to various degrees by other nuclei in the specimen.

Neutron-induced luminescence as it occurs during irradiation, can be recorded by contact color photography and can be resolved from particle produced autoradiography. Depending upon the situation and the intention of the illustration, such luminescence as fluorescence and phosphorescence may or may not be considered as interference with lithium and boron neutron-induced autoradiography. The luminescence of minerals and other material has been photographed for many years but we feel that this method of recording luminescence as it is actually taking place in a neutron beam is unique and exclusive. The usual means of exiting luminescence in minerals, rocks and other material is by irradiation with ultra violet light, x-rays, cathode rays, and other copuscular irradiation which also exposes photographic film. Once the electromagnetic radiation or particle beam has been shut off only the phosphorescent component can be photographed. In contrast neutron-induced techniques used with color film and coupled with the proper reflector-foil, intensifying-screen and particle absorbing foil or screens may be used to differentiate the several sources of film darkening used in illustrating luminescence and autoradiography.

In summary, there are four distinct ways one can use neutrons to produce an autoradiographic or radiographic image of a rock or mineral specimen. They are from (1) radioactivation produced radioactivity; (2) neutron-induced particle emission; (3) neutron-induced luminescence; and (4) in addition to autoradiographic images, a radiographic image can be formed by neutron absorption (neutron radiography) in the sample, similar to an x-ray radiograph. This study is primarily concerned with neutron-induced particle autoradiography, neutron radiography and neutron-induced phosphorescence and fluorescence produced in selected geologic specimens containing lithium and boron.

Historical Background

Shortly after the discovery of the neutron by Chadwick (1932), laboratories throughout the world noted that the interaction of these new-found particles with stable elements resulted in induced radioactivity. Because of the great penetrating power of neutrons, Kallman and Kuhn (1937) found that neutrons could be used in neutron radiography. Neutron radiography has found popularity as a method of inspection similar to the radiographic method developed for x-rays during the preceding half century. Early techniques used a special phosphor screen to convert the neutron interactions to light in order to make a neutron radiograph on the slow speed photographic film available at that time. Generally, lithium and boron were mixed with zinc sulfide phosphor and the scintillations of visible light produced by alpha particles in the phosphor produced an intensified photographic image. Later methods with reactor-produced neutrons made use of metallic foils, such as gold, silver or gadolinium, which all have a high neutron capture cross section. After transmission through the object the neutrons produced local areas of radioactivity (e.g. electron capture in gadolinium) which varied in intensity in accord with the total neutron absorption of the object. Upon termination of the neutron exposure, the film was developed if a gadolinium foil was used. If the radioactivated foil emits beta and gamma radiation with measurable decay times (e.g. gold or silver), the foil was transferred to a photographic film for a short radiographic exposure. The result was a relatively high resolution neutron radiograph. Further descriptions of the early research and recent developments in neutron radiography are given by Peter (1946), Kallman (1947), Thewlis (1956), Berger and McGonnagle (1962), and Berger (1971).

Another early application to the study of lithium by neutron-induced particle tracks was made by Picciotto and Van Styvendael (1951). By microscopically determining particle-track lengths in special photographic emulsions, they could distinguish the alpha and triton tracks of lithium by their short length from the alpha tracks of longer length produced from natural radioactive decay and transuranic elements. They were able to determine minute amounts (10^{-10}gm) of lithium in liquid samples and geologic minerals. More recent geologic applications have been described for the microscopic localization of particle tracks from boron and lithium in tourmaline by Berman and Stolyarova, (1977), boron distribution in scarn minerals (Berzina, et al., 1974), the distribution of boron in mollusc shells (Furst, et al., 1976), boron in mineral specimens from Mendip Hills in southwest England (Din and Henderson, 1982) and boron in chert (Truscott and Shaw, 1984). Potts (1984) reports platinum-group metals and other long-lived beta emitting radioisotopes have been selectively autoradiographed in granitic rocks six months to two years after radioactivation.

Method

This investigation has been mainly directed to a photographic method using commercially available film, and large hand-sized geologic specimens. The large size commercial film was used to make multiple radiographs of several specimens simultaneously when the large beam port (20 x 25 cm) of the

quarter-megawatt TRIGA reactor at the General Atomic Corporation was used for the irradiation. This reactor provided a well collimated thermal neutron beam. In addition to the reactor neutron experiments, californium-252 also was used as a point source of neutrons. In the latter case, the flux consisted of fast and thermal neutrons which were not well collimated. No neutron radiography experiments were performed with the californium-252 source because, after collimation, the number of neutrons available for radiography would be too small for practical application.

Depending on the experiment, Kodak Commercial 6127, DuPont x-ray NDT-75, Vericolor II, Ektachrome and Polaroid film were used. In addition to the use of photographic film, we have used a new plastic polymer (CR-39) that has recently become available for particle track recording (Cartwright, et al., 1978; Ahlen, et al., 1981). The polyester (allyl diglyco carbonate) commercially marketed under the trade name Homelite* has unique qualities of sensitivity and resolution for almost the complete charged particle energy spectrum as well as excellent optical properties.

Typical neutron reactor exposures vary from 10 to 40 minutes depending upon the experiment. Reactor power adjustments were made so the total neutron fluence was within the range of 10^7 to 10^9 nyt for various experiments. About 60 μgm (35 mCi) of californium-252 which has a 2.6 year half-life also was used for the experiments. The useful thermal neutron fluence from this source for induced-particle autoradiography is within the range of 10^5 to 10^9 nvt (calculated at the source) and thus required exposure times of hours to several days. Larger californium-252 sources are available but the shielding required for personnel protection reduces the mobility and therefore the usefulness of the equipment for portable application.

Four hand-size geologic specimens were selected for this study. Two were lithium bearing; one containing the mineral spodumene, and the other the mineral amblygonite. The other two specimens contained boron; one was a specimen of colmanite, and the other contained borax and tincalconite. The samples were sawed, and lightly polished to obtain a flat surface required for good resolution and high quality photographs. A parallel double cut at a thickness of about 1 cm was made on those specimens that were to be used for neutron radiography.

EXPERIMENTAL CONFIGURATION

The different configurations we have used for neutron irradiation from a reactor beam port and the use of a portable californium-252 source are shown diagramatically in Figures 1, 2, and 3. The arrangement for neutron-induced particle autoradiography (Figs. 1 and 2) show the film or plastic particle detector being placed between the source of neutrons and the geologic specimen. This prevents "shadowing" the detector and reducing the intensity of the neutron beam by absorption from the high cross-section elements in the geologic sample itself; especially in the boron and lithium minerals.

*Use of commercial names is for identification purposes only and does not imply government endorsement.

The configuration for neutron radiography (Fig. 3) allows the neutron beam to first pass through a geologic specimen about 1 cm thick. By the shadowing from lithium, boron, and other high cross-section elements in addition to neutron scattering from the lighter elements, a neutron radiographic image is produced upon the adjacent film. Using a converter foil to produce detectable radioactivity gives a picture analogous to an X-ray radiograph which uses atomic density and bulk density as the shadowing mechanism. The direct effect of neutrons upon the film and the detector package itself can be shown to be slight in experiments where the geologic specimen is removed from the detector package.

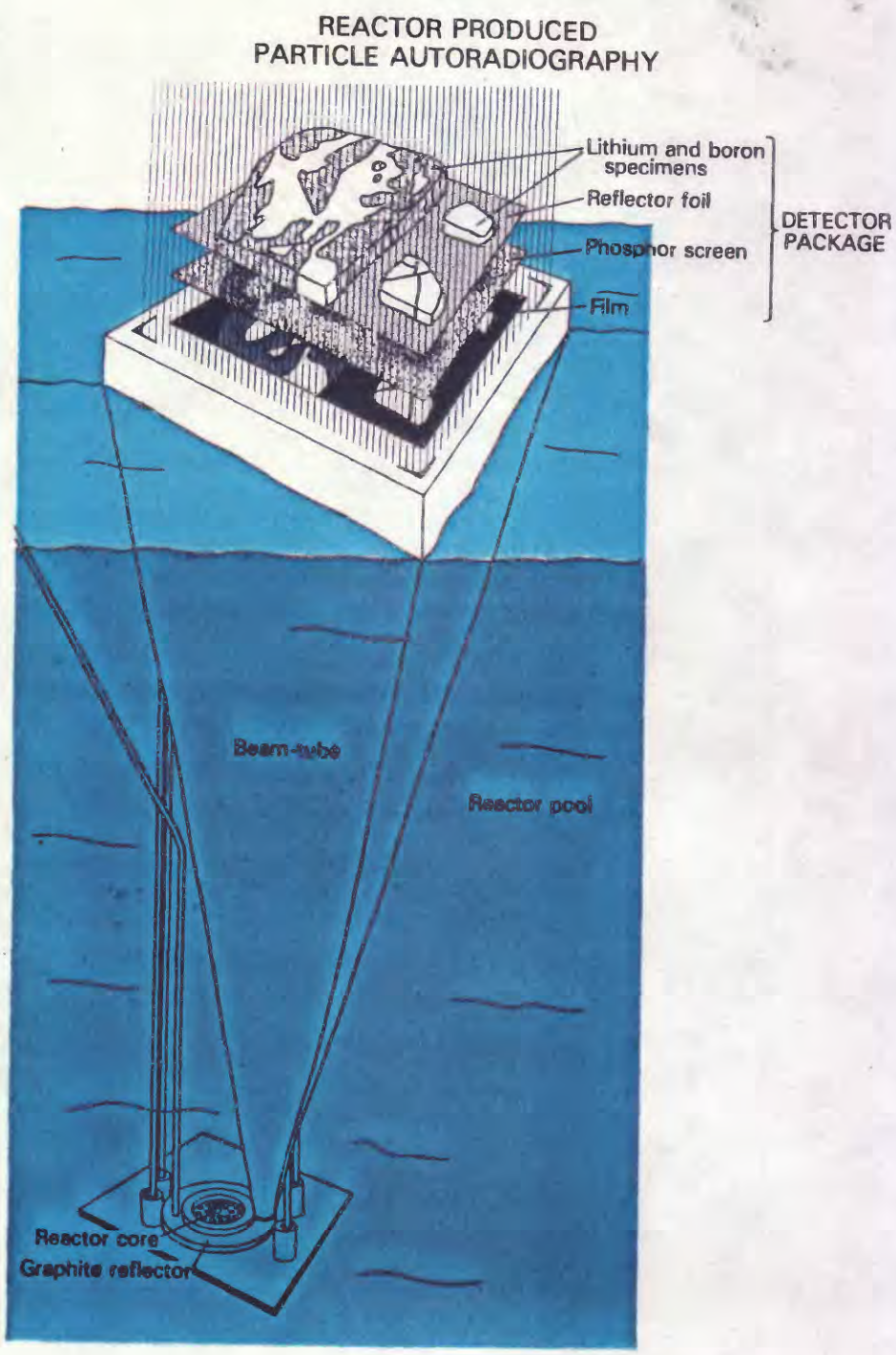


Figure 1. Perspective view of a neutron-induced particle autoradiograph being produced by the TRIGA reactor. The neutron beam is collimated through the beam tube which comes up through the reactor pool from the graphite reflector around the reactor core. The expanded view of the detector package shows the position of the geologic specimen above the film as it is being activated by the neutron beam. This causes the prompt particle production from the bottom surface of the specimen to produce an image on the photographic film or plastic particle detector. A thin foil protects the film from any neutron induced luminescence from the specimen and enhances the particle image from the phosphor screen by back reflection.

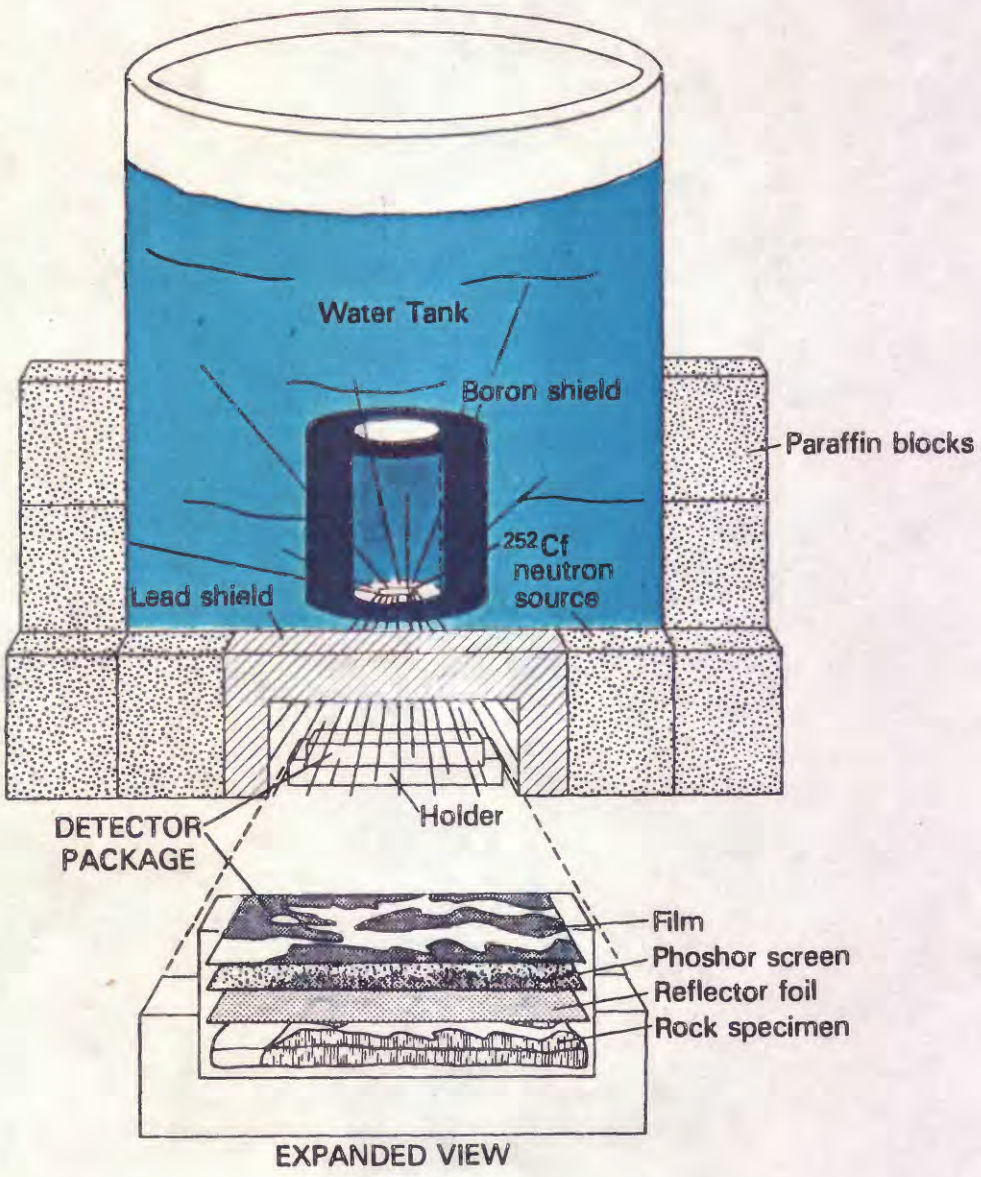


Figure 2. A perspective front view of the water tank shield containing the portable californium-252 portable neutron source and an expanded view of the detector package. The boron shield and paraffin moderator are shown with the lead-shielded cave where the detector package is placed during neutron irradiation. Multiple samples have been stacked in detector packages during one irradiation and the detector package has been inverted in some experiments; neither of these rearrangements have shown a signifciant shadowing effect upon induced-particle autoradiographs.

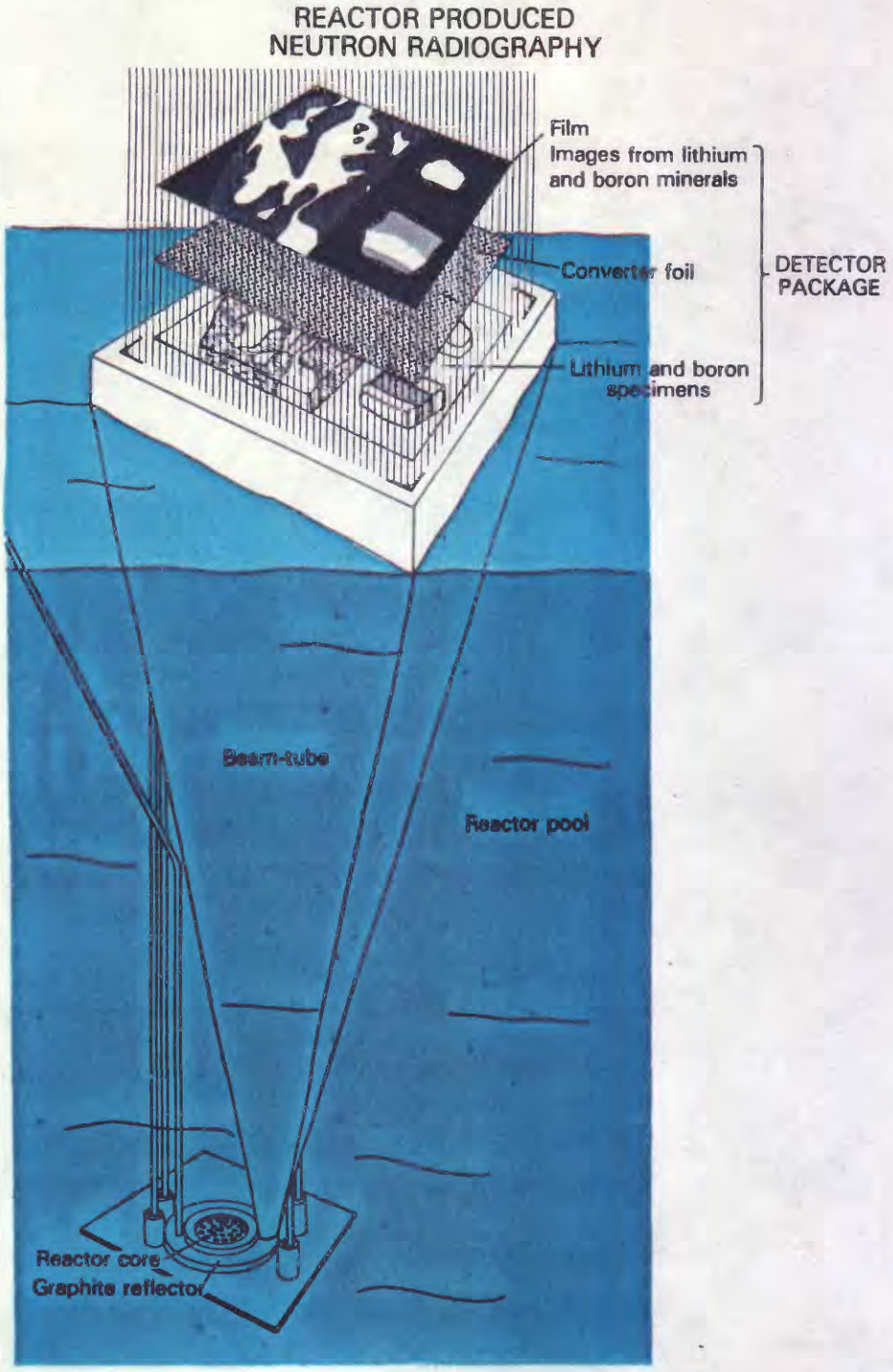


Figure 3. Perspective view of a neutron radiograph being produced by the collimated neutron beam from the TRIGA reactor. The geologic specimens are placed in front of the photographic film and the converter foil. A shadow image is produced from high cross-section elements and neutron scattering from the lighter elements. This is shown as a photographic negative image on the film upon which boron bearing specimens of colemanite and borax along with a lithium bearing specimen containing spodumene are illustrated. A gadolinium-converter foil can be used on either side of the film and is sometimes held in a vacuum cassette in a position above the film.

of radioisotopes (Buckaloo and Cohn, 1956); (Shindo, et al., 1968) and (Bodemann and Von Erichsen, 1973) but we have not been able to find reference to the use of color film as a means of resolving various sources of neutron-induced fluroescence and phosphorescene.

Neutron-Induced Particle Autoradiography

Different types and arrangements of absorber foils, phosphor screens and converter foils have been used in our application of neutron-induced particle autoradiography. In most of the experiments, a silver activated zinc sulfide intensifying screen, adapted from earlier work with the radioluxograph (Dooley, 1958), was used to amplify the photographic detection of the alpha particles. The phosphor coated side of the screen is carefully placed in contact with the flat geologic specimen and the film so as to avoid tribolumnescence. Depending on the experiments, very thin aluminum-on-mylar reflector foils, 1.5 μm in thickness, were used. The first objective of this multi-purpose foil was to use the opaque metal coating to prevent the luminescent light proudced in the mineral phenocrysts from reaching the photographic film. Next, the very thin nature of the foil should allow most alpha particles, tritons and some recoil nuclei to penetrate the foil; this in turn, allows only the induced-particles from just the lithium and boron in the geologic specimen to produce scintillations on the adjacent phosphor screen and cause film darkening. In addition, the mirror-like quality of the aluminum coating intensifies the particle-produced scintillations by back reflection. The reflector foil should be kept as thin as possible because of the energy-attenuation loss of the alpha and recoil particles passing through the foil. This loss is somewhat compensated for by scintillations enhanced by back reflection and from increased ionization at the end of the alpha particle path within the phosphor screen itself caused by range straggling at the end of the Bragg alpha particle ionization curve.

Activation of the specimen, the film, intensifier screen, reflector foil and other parts of the detector package during the neutron exposure does produce some beta particle emission due to induced ratioactivity which in turn produced a low level fogging of the film. The relector foil and phosphor screen tend to shield some of the beta emission from the sample itself, but they do not completely eliminate beta fogging. Tests with minerals containing known intense beta-emitting isotopes show that the effect of beta fogging is minimal for the relatively short neutron irradiations.

Neutron-Induced Luminescence

The more troublesome source of interference with induced particle autoradiography is fluorescence from the minerals in the geologic specimen. To some extent this problem can be anticipated by inspection of the geologic specimen with an ultra-violet lamp prior to making an autoradiograph in order to localize the fluorescent minerals (Gleason, 1972). Although a foil will essentially block all the fluorescence interference from the sample, it will not eliminate the fluorescence originating in the intensifier screen or the

The reactor beam port, as shown in Figures 1 and 3, has a resolution factor of about 40 defined as the L/D ratio; where L is the length of the neutron beam from the reactor core to the specimen (about 10 m), and D is the neutron density distribution across the image plane or the focal spot size. The focal spot is essentially equal to the exit tube diameter (about 25 cm) which gives a L/D ratio of 40 for the reactor arrangement. The reactor power levels and the length of exposure times we used are in agreement with the recommendations of Hawkesworth and Walker (1983, p. 6) for optimum total neutron fluence versus L/D ratios for thermal neutron radiography.

The californium-252 source was used at a distance of about 20 cm from the film and does not give a comparabale resolution over a 12 cm image plane. Because of the poor collimation and the relatively low intensity of the source, neutron radiography was not attempted with the californium-252 source. Others have reported (Ruddy, et al., 1984) use of a much larger californium-252 neutron source using fast neutrons for pinhole radiography.

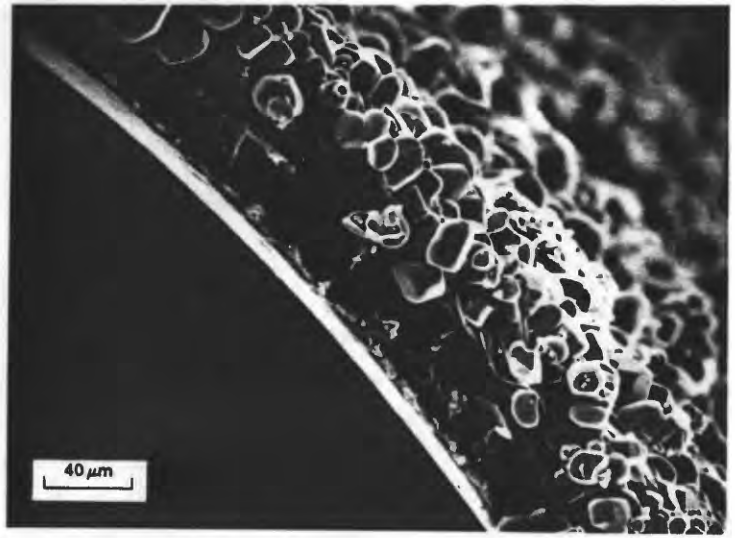
The amount of gamma and x-ray radiation reaching the photographic film in the detector package during the neutron reactor irradiation is considered a source of interference because it causes film darkening and fogging in competition with the production of the particle-induced image. A distance of 10 meters from the reactor core in our case diminishes the intensity of this unwanted radiation. The californium-252 neutron source has a very low intensity of gamma and x-rays and therefore can be used with some lead shielding (5 cm) at much closer distances to the film without causing appreciable fogging. A small amount of gamma and x-radiation does build up in the californium-252 neutron source with ageing of the source due to the daughter products formed and the build up of fission products. This is minor in comparison to the gamma radiation produced from neutron interaction with the water in the tank, which is used for thermalizing the fast neutrons and shielding personnel from the source. Our use of the californium-252 source as close as 12 centimeters from the film within the lead cave shield has not produced any serious film fogging problems. Roll film should be avoided other than for single exposures because it can accumulate all of the gamma and x-ray radiation from successive exposure use until the last exposure is made on the roll. Single sheet film of Polaroid and other films can be used when this becomes a problem. The new particle-track detecting plastic CR-39 is not significantly photon sensitive at these relative low neutron irradiation levels, and does not require the protective procedures which are necessary to protect film from light, x-rays and gamma radiation.

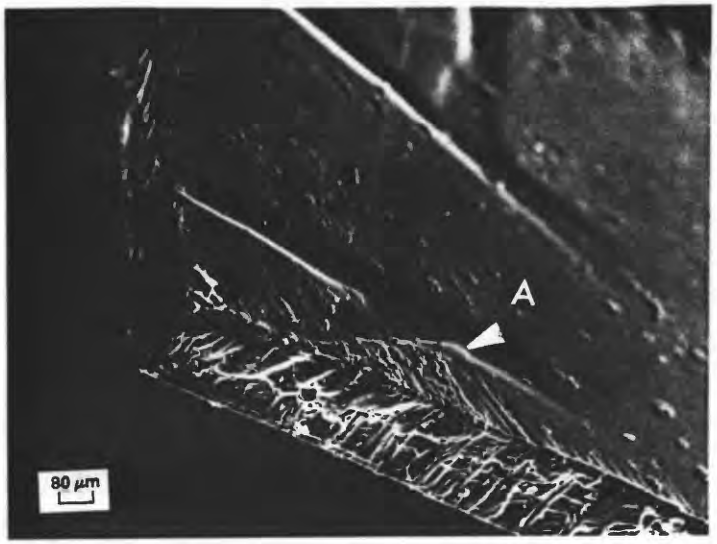
TYPES OF NEUTRON IMAGING

Of the four or more different means by which neutrons can produce photographic images, usually two or more autoradiographic processes can be recorded concurrently with either film or other track detecting methods. It has been our objective to attempt to resolve the prompt neutron-produced emissions of lithium and boron from other prompt and delayed radioactivation and luminescent produced interferences. By adding the extra dimension of "color" to the use of black-and-white film for autoradiography we believe we have been able to resolve the fluorescence of the spodumene phenocrysts during neutron irradiation. Color film has been used in the past for autoradiography

film itself. Generally, these latter sources of fluorescence can be reduced to a workable level by keeping the neutron irradiation at a low to moderate level.

Although induced luminescence is an interference problem in induced-particle autoradiography, it can also be used in its own right as an advantage to make a neutron induced image. This can be accomplished with black and white film, although without using a reflector foil or a particle absorbing screen of the proper thickness, the lumninescent component is not obviously evident. The particle image and the luminescent component both expose the film to give a black and white image. Color film adds a special dimension to the illustrations. The color-shades of the luminescence are revealed when the color autoradiograph is properly made and interpreted. The transparent acetate plastic base of both the photographic film and the phosphor screen can be used to absorb most alpha, triton and recoil nuclei particles when the plastic base is reversed so the acetate backing is next to the plane-sawed mineral specimen. An exposure through the acetate base and antihalation layer of the film (a special layer used to prevent back-scattering of light); or an exposure through the acetate backing of the phosphor screen allows the visible luminescence to produce a color image. Figures 4-A and 4-B show magnified images of both the phosphor screen and the acetate-base color film showing the various components. By making a neutron-induced luminescent photograph in this manner with the color film reversed, the colors are shifted to the red by the color filtering action of the antihalation layer, just the luminescence is shown but in a false red color-shifted image. If the neutron-induced luminescent photograph is made in another manner with just the acetate backing of the phosphor screen reversed and next to the spodumene specimen, a slight blue enhancement of the luminescent light is observed because of the neutron induced blue phosphorscence of the zinc sulfide grains of the phosphor screen itself. Similar experiments using isotopically enriched ^{6}Li and ^{10}B inorganic chemical salts without a zinc sulfide screen show a white induced particle image on the color film. Therefore it was found that when using the film emulsion in direct contact with the spodumene specimen, the color photograph is white-color brightened (by the induced-particles penetrating all three color-layers) and a near-true but white-brightened color of the luminescent spodumene is produced.





ELECTRON MICROGRAPHS

Electron micrograph of an oblique view of both the 1101-D phosphor screen and the acetate-base film. The acetate layers were used to suppress particle autoradiography and radioactivation autoradiography while allowing neutron induced luminescence to produce a color photographic image.

Figure 4-A

At 3000-X magnification the largest zinc sulfide phosphor grains are shown to be about 35 μm and are upon a transparent adhesive that is supported on a 5 μm (0.2 mil) thick acetate base. The backing has been found to absorb most natural radioactive alpha particles of 4 to 6 Mev energy.

Figure 4-B

At 1400-X magnification the acetate base of the film ∿ 160 μ (6.3 mils) thick is shown with the emulsion surface to the top. Region "A" shows where a portion of the developed emulsion was peeled away next to the manufacturers embossing. The bottom shows a very thin surface where the antihalation layer remins. This layer and the acetate base are of a sufficient thickness to adsorb most charged particles and beta radiation ≦ 0.81 Mev energy, according to Bodemann and von Erichsen, 1973.

Neutron Radiography

Many methods have been used for converting the neutron beam intensity as it emerges from the specimen into a photographic image for neutron radiography. As mentioned above Kallman and Kuhn used a phosphor screen impregnated with boron and lithium to produce a photographic image (Berger and McGonnagle, 1962). Hawkeswroth (1971) used a ^{6}Li-ZnS system along with Polaroid film for neutron radiographic studies. Later methods of neutron radiography have used transfer techniques with converter foils, e.g. silver, gold and others (Berger, 1971). Today gadolinium foil (about 0.05 mm thickness) is used to produce high resolution detection from prompt electron capture (Feigle and Rauch, 1968). The gadolinium foil can also act as a light reflector to opaque out any induced luminescence from fluorescent minerals in the sample when these are included in the detector package. Generally, in practice, the samples being radiographed are outside the film cassette containing the gadolinium converter and x-ray film; this protects the film from extraneous light exposure including any sample luminescence.

RESULTS

Illustrations are shown in Figures 5-10 for the geologic samples we have studied using black-and-white and color films along with the CR-39 particle-track detecting plastic. The first (Fig. 5-A) shows a photograph of the plane-sawed and polished side of the lithium bearing spodumene specimen at a 1::1 scale, which is also the scale for all the other specimen photographs and illustrations. Figure 5-B shows the neutron-induced particle autoradiograph of the spodumene using reactor neutrons; Figure 5-C shows a companion particle autoradiograph using californium-252 produced neutrons. Figure 5-D shows a neutron radiograph of the specimen. Figure 6 shows a supplementary photograph of the opposite side of the spodumene specimen shown in Figure 5-A. This picture allows a better visulization of the lithium bearing phenocrysts to be made as they occur in depth in the 0.7 cm thickness of the specimen and as they appear in the neutron radiograph shown in Fig. 5-D. Figure 7 shows a similar sequence of photographs illustrating a lithium-bearing amblygonite specimen.

Boron is illustrated in a specimen containing the boron mineral colemanite in the Figure 8 sequence of photographs. An evaporite specimen containing the boron minerals, borax and tincalconite, is shown in the Figure 9 sequence. Use of the C-39 particle-track detector is illustrated with each boron-containing specimen Figures 8-D and 9-D.

The Figure 10 sequence shows color photographs of neutron-induced luminescence from the lithium-bearing spodumene specimen. Both sides of the spodumene specimen (Fig. 5-A and Fig. 6) are illustrated in the sequence of color photographs. These color photographs demonstrate that neutron-induced luminescence can be equally or more important than particle emission as an exposure agent especially with photographic color film.

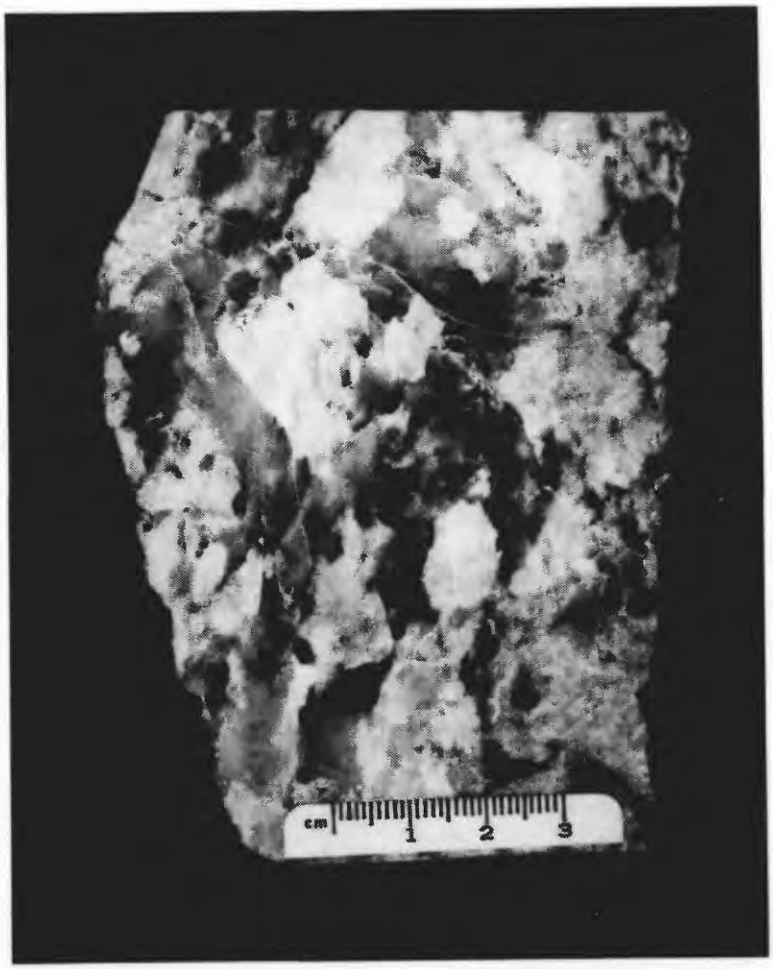


Figure 5-A. Photograph of the polished surface of a spodumene-bearing pegmatite about 0.7 cm thick from Kings Mountain, North Carolina; area described by (Kesler, 1961). The lithium concentration of spodumene is 3.7% by weight and the grade of the ore specimen is about 0.7% lithium. The scale shown (approximately 1::1) applies to all of the photographs and specimen figures.

Figure 5-B. Neutron-induced particle autoradiograph of the specimen shown in Fig. 5-A. This photographic positive illustrates the prompt $^{6}Li(n,\alpha)^{3}H$ particle reaction as white on the radioluxograph from the lithium bearing spodumene, occuring only on the surface of the specimen; the image is somewhat enchanced with neutron induced luminescence. Only a zinc sulfide phosphor screen was used between the geologic specimen and the photographic film to produce the radioluxographic image (reactor produced neutrons ~ 10^{9} n/cm^{2}).

Film used was Dupont type ND-75 x-ray film, normally processed and printed.

Figure 5-C. Neutron-induced particle autoradiograph, the same as shown in Fig. 5-B but produced with a thin aluminized mylar reflector screen in front of the phosphor screen blocking the neutron induced luminescence and reducing somewhat the alpha (^4He) and triton (^3H) radioluxographic particle image. A photographic positive shows the image as white. (californium-252 source produced ~10^9 n/cm^2). The white scattering beyond the edge of the specimen indicates the asymmetry of the divergent neutron beam, additional particle production as interference, and possibly a gamma component due to an unknown inadequacy in shielding.

Film used was Kodak Commerical 6127 normally processed and printed.

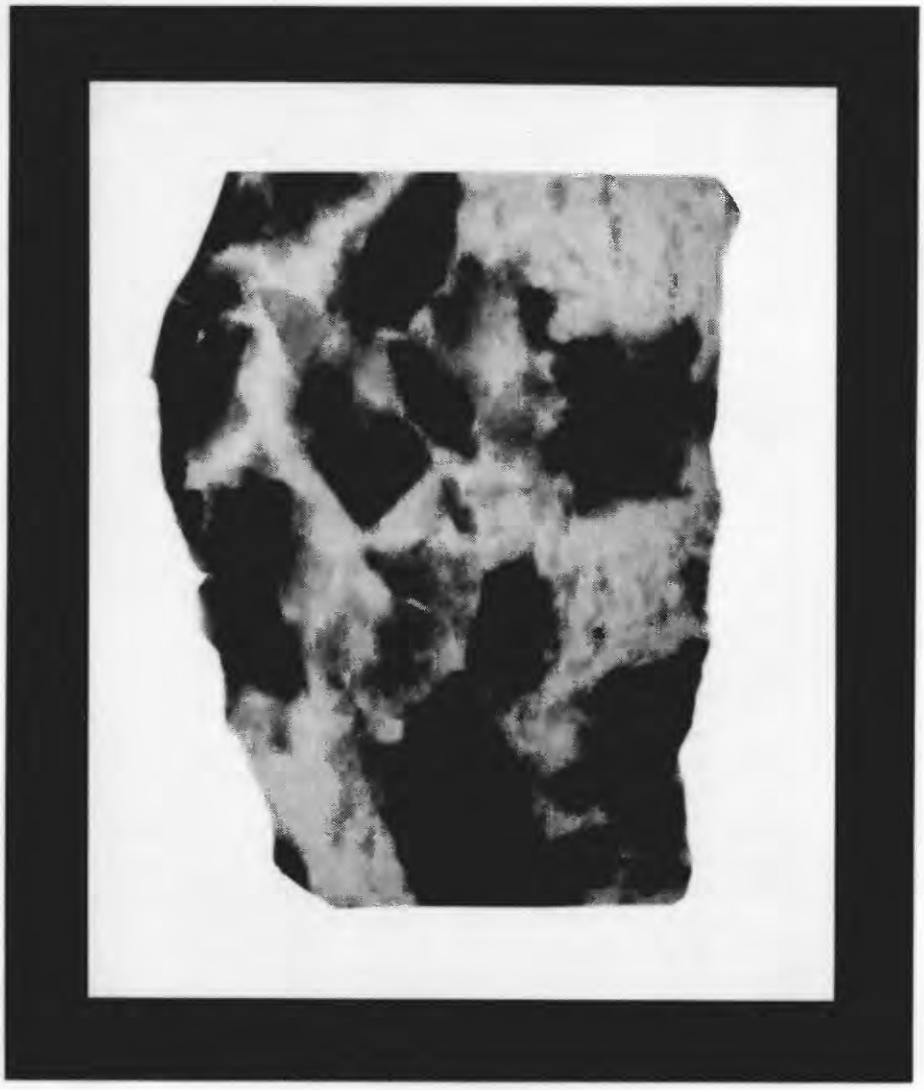


Figure 5-D. Neutron radiograph of the 0.7 cm thick, plane-sawed spodumene bearing specimen shown in Fig. 5-A. This is a photographic negative print illustrating the highest cross-section and greatest depth of neutron absorption and scattering by the darkest color; it is analogous to a photographic print made of a x-ray negative. Most spodumene phenocrysts appear throughout the thickness of the specimen, but generally do not maintain their surface configuration. This neutron radiograph made with a 0.05 mm (2 mils) thick gadolinium converter foil (reactor produced neutrons ~10^9n/cm^2).

Film used was DuPont type ND-75 x-ray film, normally processed and printed.

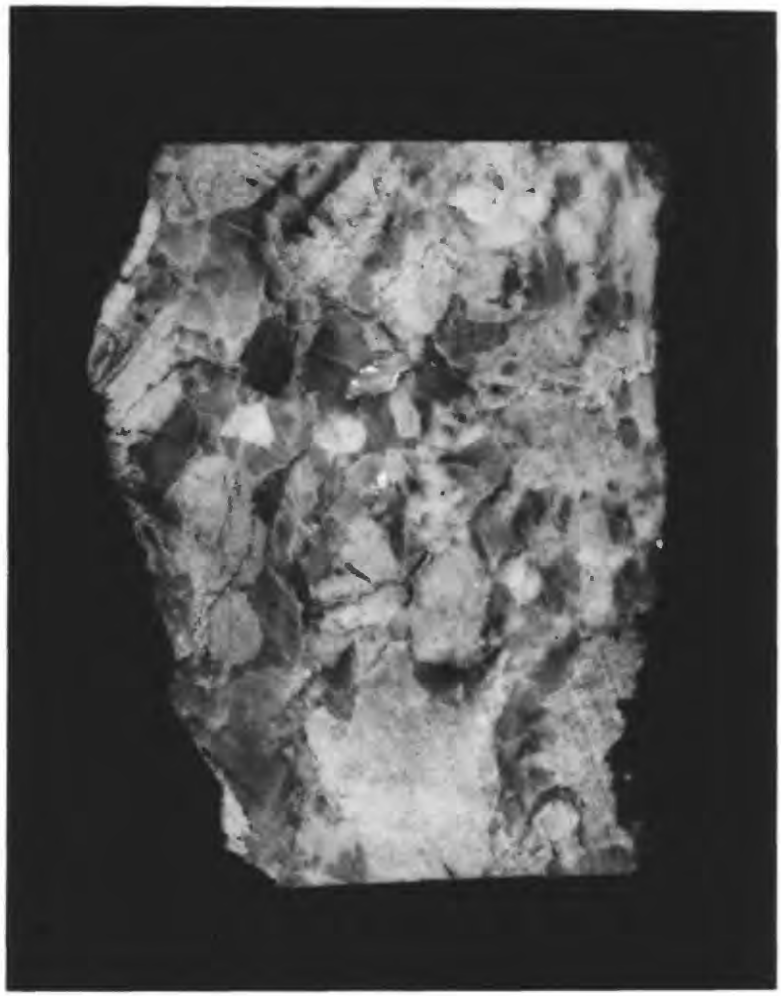


Figure 6. Supplementary photographic view of the opposite side of the spodumene bearing specimen from Kings Mountain, North Carolina. The other side, the plane-sawed and polished side, is shown in Fig. 5-A. This print was inverted so that the image shown here could be directly viewed and compared to the neutron radiograph in Fig. 5-D which shows the dimensional in-depth view of the lithium-bearing phenocrysts as they occur throughout the thickness of the specimen. Later illustrations (Figs. 10-A, 10-B, 10-E) also show this side of the plane-sawed spodumene specimen.

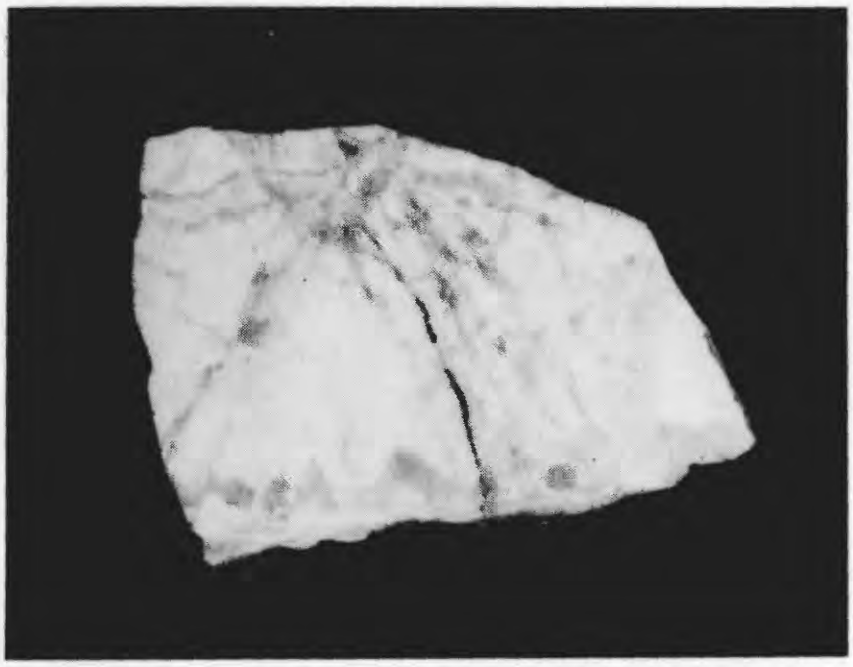


Figure 7-A. Photograph of the polished flat surface of a amblygonite-bearing pegmatite specimen about 1 cm thick from the White Picacho District of Arizona; the area is described by Jahns (1952, p. 81-83).

Figure 7-B. Neutron-induced particle autoradiograph of the lithium-bearing amblygonite specimen shown in Fig. 7-A. A reflector foil was used to block neutron induced luminescence in front of the zinc sulfide phosphor screen, which in turn reduces the $^{6}Li(n,\alpha)^{3}H$ particle reaction on the radioluxograph (californium-252 source produced neutrons ~10^{8} n/cm^{2}). A slight asymmetry of the divergent neutron beam is indicated at the right edge of the specimen by a diminished image intensity.

Film used was Kodak Commerical 6127 normally processed and printed.

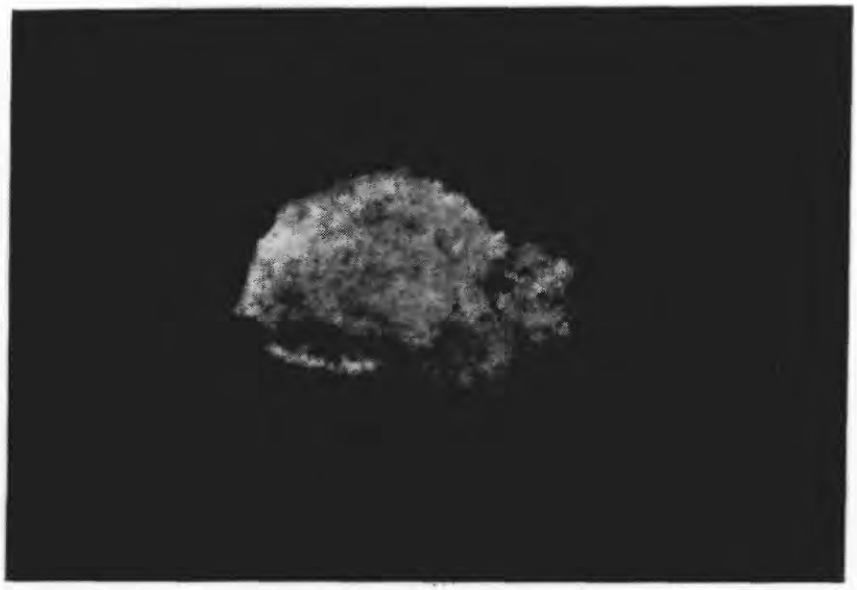


Figure 8-B. Neutron-induced particle-autoradiograph of the boron specimen of colemanite shown in Fig. 8-A. No reflector particle-absorber foil was used to block any luminescence or attenuate the $^{10}B(n,\alpha)^{7}Li$ particle interaction but a zinc sulfide radioluxographic amplifying screen was used that could allow some transmitted luminescence through the phosphor screen to the film (californium-252 produced neutrons ~10^{7} n/cm^{2}). No asymmetric beam enhancement of the image is evident at this low intensity neutron irradiation.

Film used was Kodak commerical 6127 normally processed and printed.

Figure 7-C. Neutron radiograph of the lithium bearing amblygonite specimen (~1 cm thick) shown in Fig. 7-A. This is a photographic-negative print, analogous to a x-ray negative (reactor produced neutrons ~10^9 n/cm^2). Non-lithium bearing phenocrysts and fracture-vein fillings are shown as they occur throughout the thickness of the specimen rather than just on the surface as shown in the previous induced-particle autoradiograph, (Fig. 7-B). This neutron radiograph was made with a 0.05 mm (2 mils) thick gadolinium converter.

Film used was DuPont type ND-75 x-ray film, normally processed and printed.

Figure 8-A. Photograph of the surface of a colemanite mineral specimen; the plane-sawed and polished nodule is approximately 1 cm in thickness. Collected near Shoshone, California by McAllister (formerly with the USGS but now retired), this local area and the adjoining Furnace Creek areas are described by McAllister (1970). The boron concentration of colemanite is 15.8% by weight and the ore-grade of this nodule is about 10% boron.

Figure 8-C. Neutron radiograph of the colemanite nodule shown in Fig. 8-A. The dense black color of this photographic-negative print is analogous to an x-ray negative and illustrates the almost complete absorption of neutrons by the ~1 cm thick boron bearing specimen. The dense black or high photographic density of this figure contrasts with the corresponding densities of the previous neutron radiographs (Figs. 5-D and 7-C) which show a grey to black color for lithium. This comparison directly illustrates the very high neutron cross section of boron as compared to the high neutron cross section for lithium (reactor produced neutrons $\sim 10^8$ n/cm^2). This neutron radiograph was made with a 0.05 mm (2 mils) thick gadolinium converter foil which also blocks luminescence from the film.

Film used was DuPont type ND-75 x-ray film, normally processed and printed.

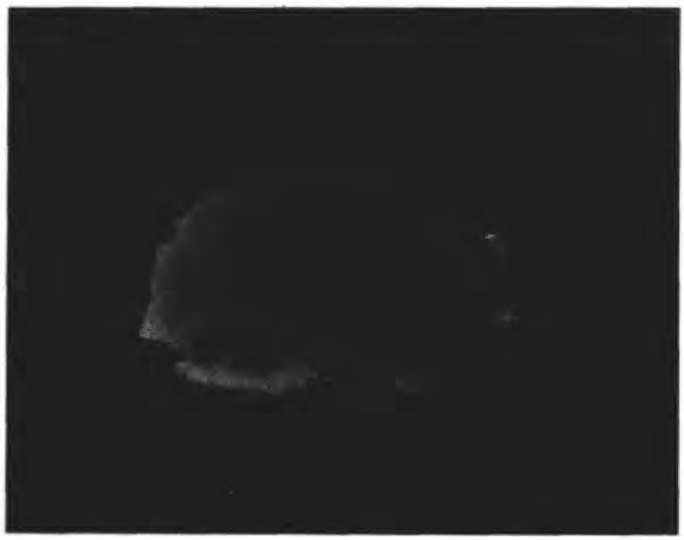


Figure 8-D. Neutron-induced particle autoradiograph of the boron containing colemanite nodule. The image is shown on the CR-39 track detecting allyl diglyco carbonate plastic, photographed in "light field" incident-light illumination. This Homelite plastic is 1.6 mm (1/16") thick and was alkli-etch developed for 10 min. at 50°C. Other neutron induced tracks are visible under megascopic and microscopic viewing, but are not apparent in this photograph (irradiated with californium-252 produced neutrons ~ 10^8 n/cm^2). No foil or screen was used between specimen and plastic detector.

Photographed with 35 mm Kodak Tec Pan film, normally processed and high contrast printed.

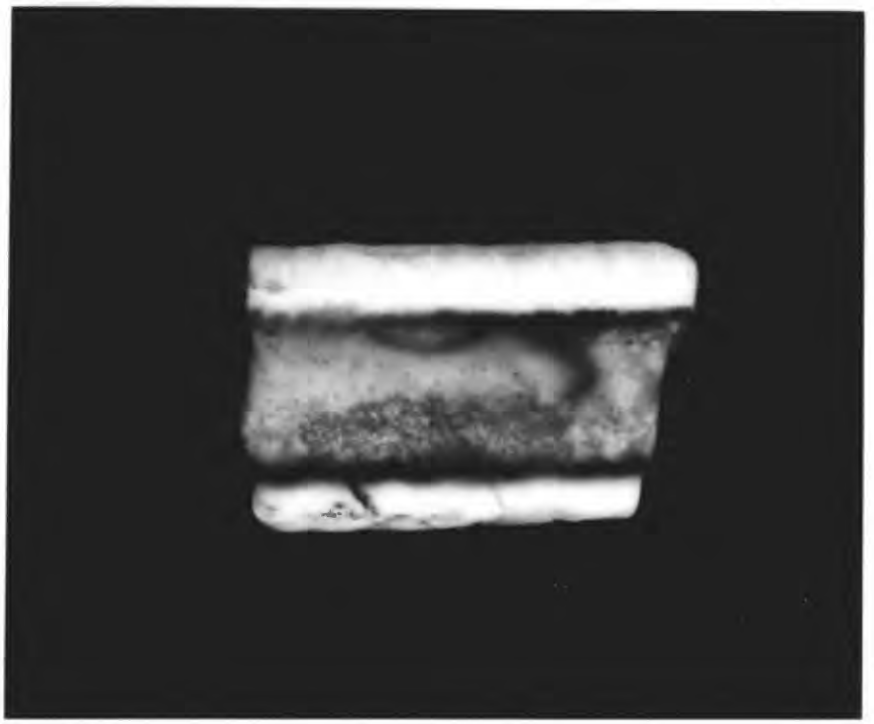


Figure 9-A. Photograph of the plane-sawed and polished surface of an evaporite specimen, about 1 cm thick, consisting of trona and containing boron in the minerals borax and tincalconite. Collected near Trona, California by G. I. Smith of the U.S. Geological Survey. Occurrences of these evaporites are described by Smith (1976).

Figure 9-B. Neutron-induced particle autoradiography of the plane-sawed and polished surface of the boron bearing evaporite specimen shown in Figure 9-A. A zinc sulfide screen was used next to the specimen to utilize the amplification of the radioluxograph (reactor produced neutrons ~ 10^8 n/cm^2). The particle autoradiograph could be slightly enhanced with luminescence.

Film used was DuPont type ND-75 x-ray film, normally processed and printed.

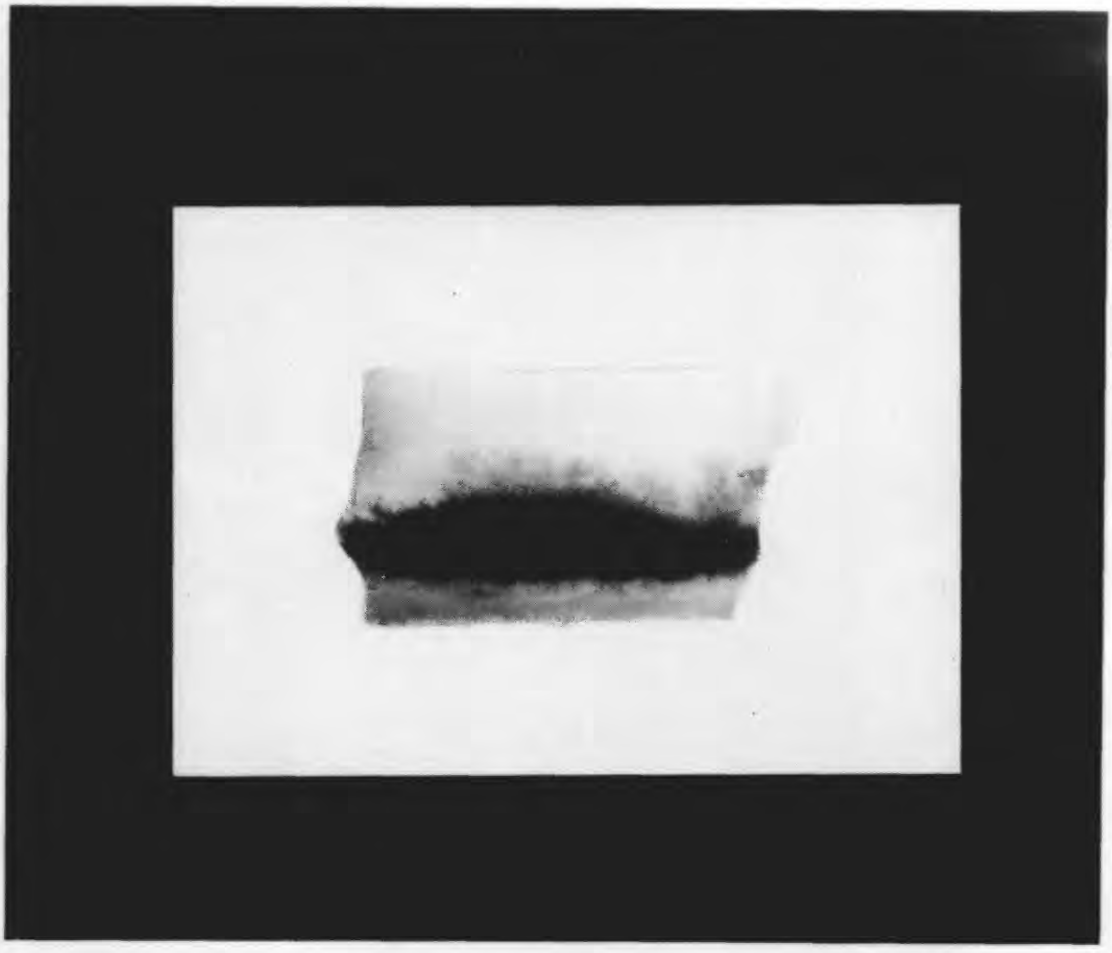


Figure 9-C. Neutron radiograph of the ~1 cm thick plane-sawed evaporite specimen containing the boron minerals borax and tincalconite (reactor produced neutrons ~10^9 n/cm^2). A gadolinium converter foil, 0.05 mm (2 mils) thick, was used in making this neutron radiograph.

Film used was DuPont type ND-75 x-ray film, normally processed and printed.

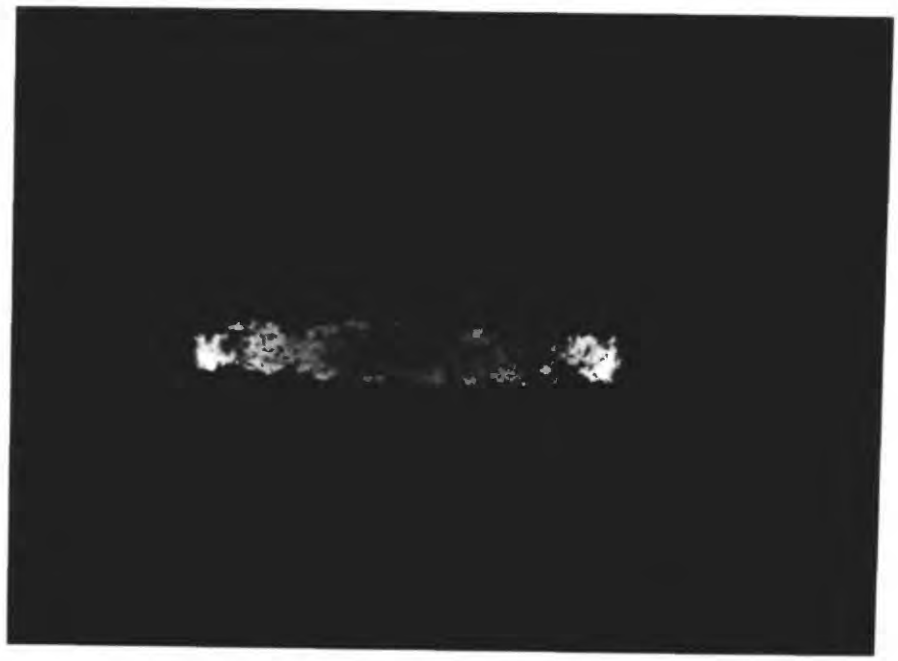


Figure 9-D. Neutron-induced particle autoradiograph of the evaporite specimen containing boron in the minerals borax and tincalconite. The image is shown on the "light field" incident light-illuminated photograph of the CR-39 new track-detecting allyl diglyco carbonate plastic. This specimen was neutron irradiated in the same detector package and developed in exactly the same manner as the colemanite specimen shown in Figure 8-D. (californium-252 produced neutrons -10^8 n/cm^2). No foil or screen used between specimen and plastic detector.

Photographed with 35 mm Kodak Tec Pan film, normally processed and high contrast printed.

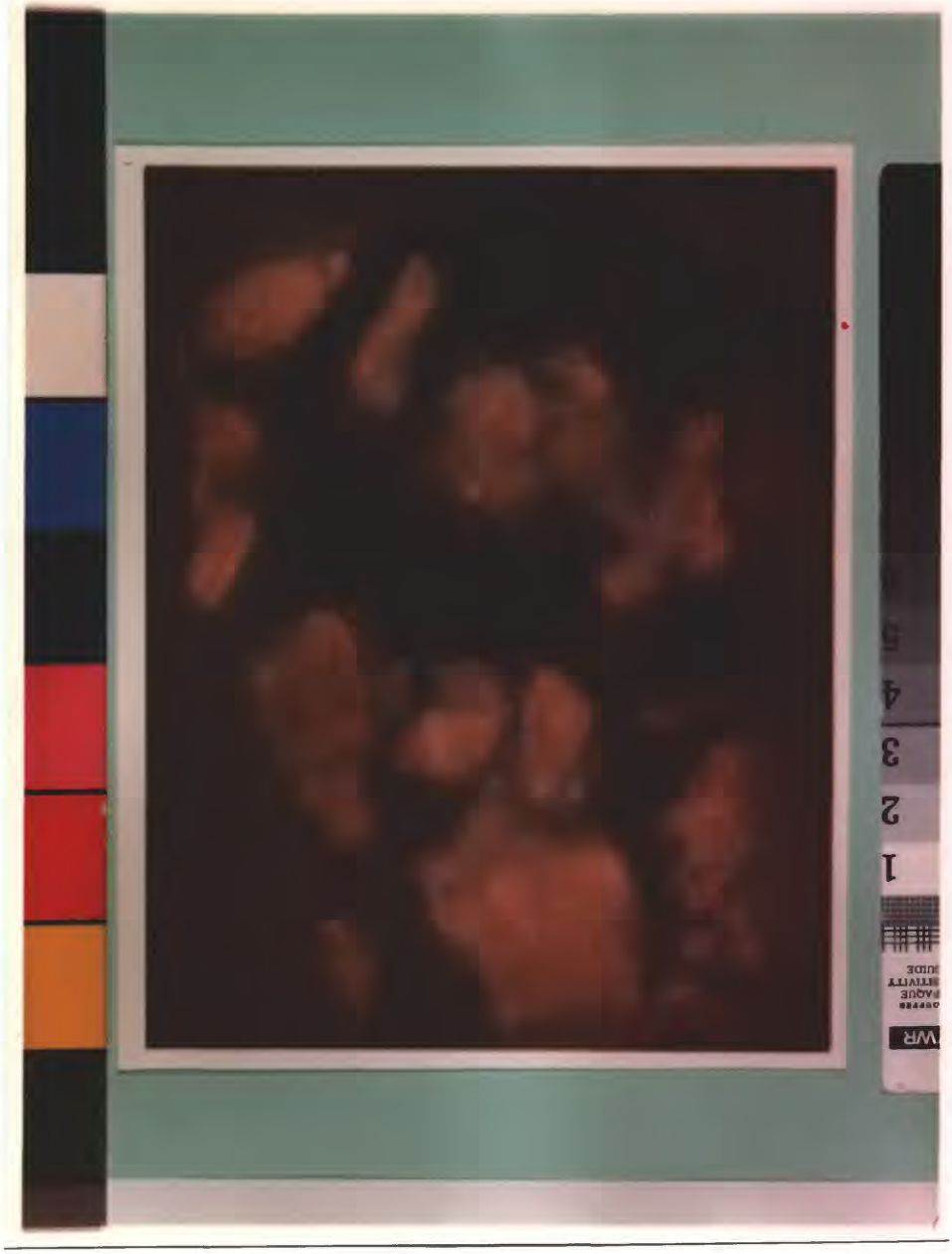


Figure 10-A. Color photograph of neutron-induced luminescence (charged-particle enhanced) from the spodumene specimen from Kings Mountain, North Carolina. Neutrons were reactor produced $\geq 10^{8}$ n/cm^{2} from a 10 minute irradiation. Plane-sawed side of specimen was in direct contact with emulsion side of film (side of specimen shown in Fig. 6). The spodumene phenocrysts appear to be luminescing on the surface and at some depth through the thickness of the specimen in a color we see as burnt orange to beige. There are regions of a few fluorescent crystals that show shades of pink to lavender, characteristic of zones of minor trace impurities that could act as luminescent color emission centers for all these colors.

Vericolor II type L, 100 ASA speed film; C-41 processed and printed using a Macbeth color chart balancing reference.

Figure 10-B. Color photograph of neutron-induced luminescence (charged-particle enhanced) from the spodumene specimen from Kings Mountain, North Carolina. Neutrons were reactor produced at ~ 10^9 n/cm^2 from a 40 minute irradiation. Plane-sawed side of specimen (side shown in Fig. 6) in direct contact with emulsion side of film. A longer irradiation appears to brighten the colors somewhat and emphasize the shape of the geologic specimen with a dark background. We interpretate the luminescence as being induced during prompt-particle production from lithium and the neutron activation of the host aluminum, silicon, and other elements of the spodumene phenocrysts. This allows the prompt particles and the very short-lived and intense beta emission to load the fluorescent and phosphorescent molecular-level color emission centers with electrons which, in turn, produce the luminescence.

Vericolor II type S 100 ASA speed film; C-41 processed and printed using a Macbeth color chart balancing reference.

Figure 10-C. Color photograph of neutron-induced luminescence (charged-particle enhanced) using an 1101-D phosphor screen with the zinc sulfide side in direct contact with the plane-sawed and polished surface of the spodumene specimen from Kings Mountain, North Carolina (side shown in photograph Fig. 5-A). Neutrons produced from californium-252 were used for irradiation at ~ 10^8 n/cm^2 that extended for several days. The rectangular shape of the zinc sulfide phosphor screen is evident, almost completely covering the geologic specimen. The phosphor screens fluorescence is also apparent but the blue color from particle production and induced luminescence of the zinc sulfide is shifted to a purple color probably because of the special developing.

Ektachrome transparency daylight-type film, 200 ASA "push-process" E-6 developed to film speed 800 ISO. Printed on Ektaflex paper using a Macbeth color chart balancing reference. Color shift to the violet probably caused from push-processing and recipocity effect.

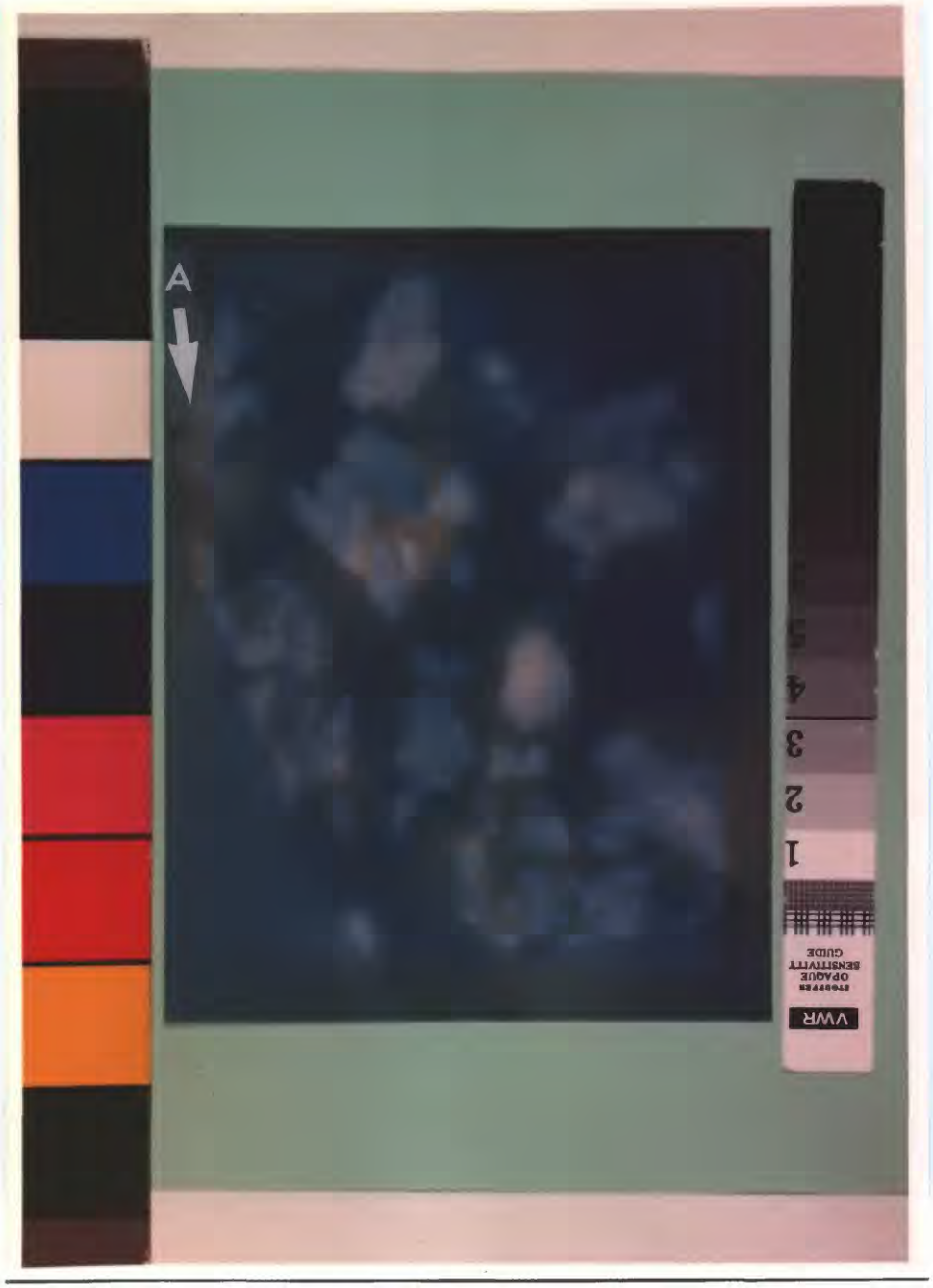


Figure 10-D. Color photograph of neutron-induced luminescence of the plane-sawed and polished side (side shown in photograph Fig. 5-A) of the spodumene specimen from Kings Mountain, North Carolina. Neutrons produced from californium-252 were used for irradiation at ~ 10^9 n/cm^2 irradiation that extended for several days. The phosphor screen was reversed to supress charged particle interaction through the acetate base side (5 μm, 0.2 mil thick) of the screen which was in direct contact with the specimen; the zinc sulfide was not in direct contact with the specimen. The blue fluorescence of the rectangular phosphor screen is near the true color as compared to an ultra-violet or x-ray irradiated screen. Of particular interest is the burnt orange to beige color of the specimen at point 'A' where the screen does not completely cover the specimen.

Vericolor II type S film 100 ASA speed; C-41 processed and printed using a Macbeth color chart balancing reference.

Figure 10-E. Color photograph of neutron-induced luminescence from the spodumene specimen from Kings Mountain, North Carolina. This experimental configuration illustrates the spodumene phenocrysts fluorescing while the prompt-particle interaction is being suppressed. By reversing the film the luminescent light is color shifted to the red by first passing through the antihalation layer and acetate film backing (shown in Fig., 4-B). This red-shift can be demonstated by making an ordinary photograph of a Macbeth color-reference chart with the film reversed. Most of the neutron-induced particles are absorbed by the antihalation layer and the acetate backing (160 μm, 6.3 mils thick) while the fluorescent light goes on through to the emulsion. The plane-sawed side of the spodumene specimen (shown in photograph in Fig. 6) was in direct contact with the back-side of the film. This illustration shows striking similarities but with some differences to the neurton radiograph shown previously in Fig. 5-D. Reactor produced neturons ~10^9 n/cm^2 were used for a 30 minute irradiation.

Ektachrome transparency, daylight-type, ASA 64 speed film. Normal E-6 developed and printed on Ektaflex paper using a Macbeth color balancing reference.

SUMMARY

The photographs in the preceeding figures illustrate the applications we have described using neutrons to study the occurrence and distribution of lithium and boron in geologic material. The techniques of neutron-induced particle autoradiography and neutron-induced luminescence are unique and new. We hope these will find continued application and lead to development of other methods in the study of geologic material. In addition we have described a use of neutron radiography for the study rocks, minerals, and other geologic specimens, and hope that this application will find continued use.

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REFERENCES

Ahlen, Stephen P., Price, P. Buford, and Tarlé, Gregory 1981, Track-record in solids: Physics Today, v. 34, no. 9, p. 32-39.

Berger, Harold, 1971, Neutron radiography: in Annual Review of Nuclear Science, (Serge, Emilio, Grover, J. Robb, and Noyes, H. Pierre, editors), v. 21, p. 335-364.

Berger, H., and McGonnagle, W. J., 1962, Progress Report on Neutron Radiography: Argonne National Laboratory, ANL-6279, 32 p.

Berman, I. F., Stolyarova, A. N., 1977, Razdel'noe Opredelenie Bora i Litiya Metodom (n,α) - Radiografii, (Separate determination of boron and lithium by the method of (n,α) - radiography): Akad. Nauk SSSR Doklady, ton 237, no. 4, p. 932-922.

Berzina, I. G., Berman, I. F., Malinko, S. V., and Stolyarova, A. N., 1974, O Raspredelenii Bora v Porodoobrazuyushchikh Mineralakh Skarnov po Dannym (n,α) - Radiografii, (Distribution of boron in rock-forming skarn minerals from (n,α) radiography data): AN SSSR Izvestia, Ser. Geol., no. 8, p. 75-80, translated in; Internat. Geology Rev., 1976, v. 18, no. 4, p. 425-429.

Bodemann, M., and von Erichsen, L., 1973, Untersuchungen zur autoradiographie mit farbfilmen (Investigation of autoradiography with color film): Messtechnik, v. 81, no. 4, p. 93-103.

Buckaloo, G. W., and Cohn, David V., 1956, Color autoradiography: Science, v. 123, no. 3191, p. 333.

Chadwick, J., 1932, The existence of a neutron: Proc. Royal Soc. (London), A 136, p. 692.

Cartwright, B. G., Shirk, E. E., and Price, P. B., 1978, A Nuclear-track-recording polymer of unique sensitivity and resolution: Nuclear Instruments and Methods, v. 153, nos. 2,3, p. 457-460.

Din, V. K., and Henderson, P., 1982, Applications of CR-39 to the mapping of B in minerals and rocks; in Proceedings of the 11th International Conference on Nuclear Track Detectors, 7-12 September 1981, (editors, Fowler, P. H., and Clapham, V. M.), Nuclear Tracks and Radiation Measurements, v. 6, supplement no. 3, p. 597-600.

Dooley, J. R., Jr., 1958, The radioluxograph: a fast, simple type of autoradiography: United Nations International Conference on the Peaceful Use of Atomic Energy, v. 3, p. 550-553.

Dooley, J. R., Jr., 1979, Autoradiographic method of lithium determination: in Geological Survey Research, U.S. Geological Survey Professional Paper 1150, p. 16-17.

Dooley, J. R., Jr., Vine, J. D., Kraker, G. P., Shoptaugh, J. R., and Enemaerke, Jan., 1979, Lithium-autoradiography of geologic specimens: Abstrats with Programs, Geological Society of America, 1979 Annual Meeting, v. 11, no. 7, p. 414.

Feigl, B., and Rauch, H., 1968, Der Gd-neutronenzähler, (On the 70 Kev internal conversion electron from gadolinum for high resolution neutron radiographie): Nuclear Instruments and Methods, v. 61, no. 3, p. 349-56.

Furst, Marian, Lowenstam, H. A., and Burnet, D. S., 1976, Radiographic study of the distribution of boron in recent mollusc shells: Geochimica et Cosmochimica Acta, v. 40, no. 11, p. 1381-1386.

Gleason, Sterling, 1972, Ultraviolet guide to minerals: Ultra-Violet Products, Inc., San Grabriel, California, 244 p.

Hawkesworth, M. R., 1971, The use of polaroid positive film in radiography with beams of thermal neutrons: Non-destruct. Test., v. 4, no.1, p. 45-48.

Hawkesworth, M. R., and Walker, J., 1983, Basic principles of thermal neutron radiography: in Neutron Radiography, Proceedings of the First World Conference, San Diego, California U.S.A., Dec. 7-10, 1981 (editors, Barton, John P. and von der Hardt, Peter), p. 5-22.

Jahns, R. H., 1952, Pegmatite deposits of the White Picacho District, Maricopa and Yavapai Counties, Arizona: University of Arizona Bulletin, Bureau of Mines, v. 23, 105 p.

Kallmann, H., 1947, Neutron radiography: Research, v. 1, no. 6, p. 254-260.

Kallmann, H., and Kuhn, E., D. R. Patent No. 694,634; U.S. Patent No. 2,186,757 (1937); U.S. Patent Nos. [1,230,618; corrected no. 2,188,115] and 2,246,443; D. R. Patent No. 753,326; French Patent Nos. 886,513, and 886,516 (1938) and U.S. Patent No. 2,245,787 (1939).

Kesler, T. L, 1961, Exploration of the Kings Mountain pegmatites: Mining Eng., v. 13, no. 9, p. 1062-1068.

McAllister, J. F., 1970, Geology of the Furance Creek borate area, Death Valley, Inyo County, California: California Div. Mines and Geology, Map Sheet 14, 9 p.

Peter, Von Otto, 1946, Neutronen - durchleuchtung, (Neutron radiography): Zeitschift fur Naturforschung, v. 1, no. 10, p. 557-559.

Picciotto, Edgard, and Van Styvendael, Marcel, 1951, Dosage et localisation du lithium par reaction nucleaire dans les mineraux, (The localization and concentration of lithium in minerals by the use of nuclear reactions): Comptes Rendus, v. 232, p. 855-857.