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AEC RESEARCH and DEVELOPMENT REPORT

ALPHA AUTORADIOGRAPHY OF IRRADIATED MATERIALS

J. L. HASCALL

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INTRODUCTION

The selective sensitivity of cellulose nitrate plastic to the deformation by alpha particles has been applied to alpha autoradiography of irradiated fuel samples. This report covers preparation, exposure, processing, and photographic reproduction of a pure cellulose nitrate plate.

Studies of the presence and distribution of alpha emitting radioactive isotopes in nonirradiated materials have been made for many years using photographic emulsions, but the presence of greater energies of beta-gamma radiations in irradiated materials makes it impossible to record alpha activity on such a photographic emulsion. When the use of cellulose nitrate plastic as a solid state nuclear track detector was described by R. L. Fleischer[1] and later by Davies and Darmitzel,[2] metallurgists and engineers were able to broaden their scope in postirradiation studies of irradiated fuel specimens.

Samples of all commercially available cellulose nitrate sheet material were secured; and, in all samples, we found impurities and plasticizers which, when exposed and processed, showed as undesirable background. A glass supported, thin film of pure cellulose nitrate was desired. The search for such a material was futile, so we proceeded to prepare our own.

SUMMARY AND CONCLUSIONS

Alpha autoradiography, using cellulose nitrate has many applications in the study of irradiated materials. Such applications are differentiating PuO_2 from UO_2 in mixed oxide fuels,[3] migration studies,[4] alpha radiation energy studies, and, when used in conjunction with beta-gamma autoradiography and optical metallography, providing information that might

have passed undetected. The preparation and use of the pure cellulose nitrate plate enhances these studies because there are no artifacts or background to distract from the desired information.

PREPARATION OF THE PLATE

One ounce of a highly purified form of nonexplosive cellulose nitrate called PARLODION* is dissolved in 550 ml of amyl acetate. The solvent is added over a period of 2 days with stirring, and the solution is allowed to sit for 24 hr to come to equilibrium. The viscosity of this solution should be such that a 5/8 in. diam steel ball will decend through 50 ml solution in a 1 in. diam, 50 ml graduated cylinder (Corning Number 3022) in 3 sec. A standard viscosity is important in order to maintain standard thickness of film on the finished plate.

Unexposed 3 1/4 x 4 in. projection slide plates (Eastman Kodak) are processed through developer to remove the antihallation backing, are cleared and fixed, and are washed and dried in the usual manner in a dust free atmosphere. A 5 ml aliquot of the prepared solution is poured in the center of each plate (level in both directions), allowed to spread and dry in a dust free atmosphere for 48 hr at room temperature. The plates are then annealed for 4 hr at 100 °C to drive out any residual solvent. This produces a thin film of cellulose nitrate which is about 15 μ thick and tenaciously adheres to the glass plates.

The advantages of this glass supported thin film are:

- Dimensional stability
- Ease of handling in and out of the hot cells
- Ease of processing and protection from scratching and abrasions in storage and examination.

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*Registered trade name of Mallinckrodt Chemical Works.

If for any reason a thin sheet is desired without a glass support, the cellulose nitrate solution can be poured on a clean flat smooth surface or onto a water surface and annealed after the solvent has evaporated.

MAKING THE AUTORADIOGRAPH

An autoradiographic image is formed by alpha particles emitted from the surface of the specimen entering the cellulose nitrate at high velocities where they are slowed and finally stopped. Deformation of the molecular structure of the cellulose nitrate is caused by ionization as energy is transferred from the alpha particles. Most of the energy is transferred just before the alpha particle is stopped; and, when the cellulose nitrate is placed in a sodium hydroxide solution, the deteriorated area is attacked and removed leaving cone shaped voids. The base of the cones are a few microns below the surface where the alpha particles stopped. Those particles entering at an angle or nearly parallel to the surface make elongated or tadpole shaped voids.

The exposure is made by introducing the plate in the hot cell and placing a prepared specimen in intimate contact with the cellulose nitrate. Exposure times range from a few seconds to several minutes depending on the radioactivity of the specimen. Autoradiographs intended for microscopic examination should have less exposure than those intended for macro examination and reproduction. Two or more exposures can be made on the same plate depending on the size of the specimen.

After exposure, the plate is removed from the hot cell, surveyed for contamination, and then carried to a hood where it is etched in a concentrated sodium hydroxide solution. In a 6.25N sodium hydroxide solution held at 55 °C, the etching times are from 2 to 4 min. The areas which were in contact

with the specimen appear frosty and the etching time is determined by visual inspection. The plate is then washed in running water for 10 min and dried in a dust free atmosphere. When the plate is dry a microscope cover glass is taped over the autoradiograph for protection against scratches and abrasions.

To date there has been no problem of radioactive contamination. The contamination that usually shows on the plate when it is removed from the hot cell is removed in the etching and washing process. The photgraphic reproductions of the autoradiographs are made in a radiation-free, uncontrolled zone so the plates must read less than 100 counts/min beta-gamma and less than 500 disintegrations/min alpha before they can be removed from the radiometallurgy laboratory.[3]

PHOTOGRAPHIC REPRODUCTION OF THE AUTORADIOGRAPHS

Kodak Gravure copy film is used to make internegatives from the original autoradiographs for both photomicrographs and photomacrographs, using collimated monochromatic transmitted light. Negatives are usually made to match the magnification of the optical photomacrographs and photomicrographs are made of areas of special interest. The negatives are contact printed and the finished prints are used for evaluation and inclusion in reports.

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4. *M. K. Millhollen. Unpublished Data, Battelle-Northwest, Richland, Washington, Private Communication.*

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