Designation: ~~E2059 – 15$^{\varepsilon 1}$~~ E2059 – 20

Standard Practice for Application and Analysis of Nuclear Research Emulsions for Fast Neutron Dosimetry[1]

This standard is issued under the fixed designation E2059; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon (ε) indicates an editorial change since the last revision or reapproval.

~~ε1 NOTE—In paragraph 1.5, "three major limitations" was corrected editorially to "four major limitations" in March 2016.~~

1. Scope

1.1 Nuclear Research Emulsions (NRE) have a long and illustrious history of applications in the physical sciences, earth sciences and biological sciences (**1, 2**)[2]. In the physical sciences, NRE experiments have led to many fundamental discoveries in such diverse disciplines as nuclear physics, cosmic ray physics and high energy physics. In the applied physical sciences, NRE have been used in neutron physics experiments in both fission and fusion reactor environments (**3-6**). Numerous NRE neutron experiments can be found in other applied disciplines, such as nuclear engineering, environmental monitoring and health physics. Given the breadth of NRE applications, there exist many textbooks and handbooks that provide considerable detail on the techniques used in the NRE ~~method.~~method (**1-4, 6**). As a consequence, this practice will be restricted to the application of the NRE method for neutron measurements in reactor physics and nuclear engineering with particular emphasis on neutron dosimetry in benchmark fields (see Matrix E706).

1.2 NRE are passive detectors and provide time integrated reaction rates. As a consequence, NRE provide fluence measurements without the need for time-dependent corrections, such as arise with radiometric (RM) dosimeters (see Test Method E1005). NRE provide permanent records, so that optical microscopy observations can be carried out any time after exposure. If necessary, NRE measurements can be repeated at any time to examine questionable data or to obtain refined results.

1.3 Since NRE measurements are conducted with optical microscopes, high spatial resolution is afforded for fine structure experiments. The attribute of high spatial resolution can also be used to determine information on the angular anisotropy of the *in-situ* neutron field (**4, 5, 7**). It is not possible for active detectors to provide such data because of *in-situ* perturbations and finite-size effects (see Section 11).

1.4 The existence of hydrogen as a major constituent of NRE affords neutron detection through neutron scattering on hydrogen, that is, the well known (n,p) reaction. NRE measurements in low power reactor environments have been predominantly based on this (n,p) reaction. NRE have also been used to measure the ^{6}Li (n,t) ^{4}He and the ^{10}B (n,α) ^{7}Li reactions by including ^{6}Li and ^{10}B in glass specks near the mid-plane of the NRE (**8, 9**). Use of these two reactions does not provide the general advantages of the (n,p) reaction for neutron dosimetry in low power reactor environments (see Section 4). As a consequence, this standard will be restricted to the use of the (n,p) reaction for neutron dosimetry in low power reactor environments.

1.5 *Limitations*—The NRE method possesses four major limitations for applicability in low power reactor environments.

1.5.1 *Gamma-Ray Sensitivity*—Gamma-rays create a significant limitation for NRE measurements. Above a gamma-ray exposure of approximately 0.025 Gy, NRE can become fogged by gamma-ray induced electron events. At this level of gamma-ray exposure, neutron induced proton-recoil tracks can no longer be accurately measured. As a consequence, NRE experiments are limited to low power environments such as found in critical assemblies and benchmark fields. Moreover, applications are only possible in environments where the buildup of radioactivity, for example, fission products, is limited.

1.5.2 *Low Energy Limit*—In the measurement of track length for proton recoil events, track length decreases as proton-recoil energy decreases. Proton-recoil track length below approximately 3µm in NRE ~~can not~~ cannot be adequately measured with optical microscopy techniques. As proton-recoil track length decreases below approximately 3 µm, it becomes very difficult to measure track length accurately. This ~~3 µm~~ 3-µm track length limit corresponds to a low energy limit of applicability in the range of approximately 0.3 to 0.4 MeV for neutron induced proton-recoil measurements in NRE.

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[1] This practice is under the jurisdiction of ASTM Committee E10 on Nuclear Technology and Applications, and is the direct responsibility of Subcommittee E10.05 on Nuclear Radiation Metrology.

Current edition approved ~~Oct. 1, 2015~~July 1, 2020. Published ~~November 2010~~August 2020. Originally approved in 2000. Last previous edition approved in ~~2010~~2015 as ~~E2059 - 06~~E2059 - 15$^{\varepsilon 1}$~~(2010).~~. DOI: ~~10.1520/E2059-15E01.~~10.1520/E2059-20.

[2] The boldface numbers in parentheses refer to the list of references at the end of the text.

1.5.3 *High-Energy Limits*—As a consequence of finite-size limitations, fast-neutron spectrometry measurements are limited to ≤15 MeV. The limit for *in-situ* spectrometry in reactor environments is ≤8MeV.

1.5.4 *Track Density Limit*—The ability to measure proton recoil track length with optical microscopy techniques depends on track density. Above a certain track density, a maze or labyrinth of overlapping tracks is created, which precludes the use of optical microscopy techniques. For manual scanning, this limitation arises above approximately 10^4 tracks/cm^2, whereas interactive ~~computer based~~ computer-based scanning systems can extend this limit up to approximately 10^5 tracks/cm^2. These limits correspond to neutron fluences of $10^6 - 10^7$ cm^{-2}, respectively.

1.6 *Neutron Spectrometry (Differential Measurements)*—For differential neutron spectrometry measurements in ~~low power~~ low-power reactor environments, NRE experiments can be conducted in two different modes. In the more general mode, NRE are irradiated *in-situ* in the low power reactor environment. This mode of NRE experiments is called the 4π mode, since the *in-situ* irradiation creates tracks in all directions (see 3.1.1). In special circumstances, where the direction of the neutron flux is known, NRE are oriented parallel to the direction of the neutron flux. In this orientation, one edge of the NRE faces the incident neutron flux, so that this measurement mode is called the end-on mode. Scanning of proton-recoil tracks is different for these two different modes. Subsequent data analysis is also different for these two modes (see 3.1.1 and 3.1.2).

1.7 *Neutron Dosimetry (Integral Measurements)*—NRE also afford integral neutron dosimetry through use of the (n,p) reaction in low power reactor environments. Two different types of (n,p) integral mode dosimetry reactions are possible, namely the I-integral (see 3.2.1) and the J-integral (see 3.2.2) (**10, 11**). Proton-recoil track scanning for these integral reactions is conducted in a different mode than scanning for differential neutron spectrometry (see 3.2). Integral mode data analysis is also different than the analysis required for differential neutron spectrometry (see 3.2). This practice will emphasize NRE (n,p) integral neutron dosimetry, because of the utility and advantages of integral mode measurements in low power benchmark fields.

1.8 *This international standard was developed in accordance with internationally recognized principles on standardization established in the Decision on Principles for the Development of International Standards, Guides and Recommendations issued by the World Trade Organization Technical Barriers to Trade (TBT) Committee.*

2. Referenced Documents

2.1 *ASTM Standards:*[3]

E706 Master Matrix for Light-Water Reactor Pressure Vessel Surveillance Standards
E854 Test Method for Application and Analysis of Solid State Track Recorder (SSTR) Monitors for Reactor Surveillance
E910 Test Method for Application and Analysis of Helium Accumulation Fluence Monitors for Reactor Vessel Surveillance
E944 Guide for Application of Neutron Spectrum Adjustment Methods in Reactor Surveillance
E1005 Test Method for Application and Analysis of Radiometric Monitors for Reactor Vessel Surveillance

3. Alternate Modes of NRE Neutron Measurements

3.1 *Neutron Spectrum Measurements*—The neutron energy range of interest in reactors environments covers approximately nine orders of magnitude, extending from thermal energies up to approximately 20 MeV. No single high-resolution method of neutron spectrometry exists that can completely cover this energy range of interest (**12**). Work with proton-recoil proportional counters has not been extended beyond a few MeV, due to the escape of more energetic protons from the finite sensitive volume of the counter. In fact, correction of *in-situ* proportional counters for such finite-size effects can be non-negligible above 0.5 MeV (**13**). Finite-size effects are much more manageable in NRE because of the reduced range of recoil protons. As a consequence, NRE fast neutron spectrometry has been applied at energies up to 15 MeV (**3**). For *in-situ* spectrometry in reactor environments, NRE measurements up to 8.0 MeV are possible with very small finite-size corrections (**14-16**).

3.1.1 *4π Mode*—It has been shown (**3-6**) that a neutron fluence-spectrum can be deduced from the integral relationship

$$M(E) = n_p \; V \int_E^\infty \frac{\sigma_{np}(E)\;\Phi(E)}{E}\; dE \qquad (1)$$

where:

$\Phi(E)$ = neutron fluence in n/(cm^2–MeV),
$\sigma_{np}(E)$ = neutron-proton scattering cross section (cm^2) at neutron energy, E,
E = neutron or proton energy (MeV),
n_p = atomic hydrogen density in the NRE (atoms/cm^3),
V = volume of NRE scanned (cm^3), and
$M\,(E)$ = proton spectrum (protons/MeV) observed in the NRE volume V at energy E.

The neutron fluence can be derived from Eq 1 and takes the form:

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[3] For referenced ASTM standards, visit the ASTM website, www.astm.org, or contact ASTM Customer Service at service@astm.org. For *Annual Book of ASTM Standards* volume information, refer to the standard's Document Summary page on the ASTM website.

$$\Phi(E) = \frac{-E}{\sigma_{np}(E) n_p V} \frac{dM}{dE} \tag{2}$$

Eq 2 reveals that the neutron fluence spectrum at energy E depends upon the slope of the proton spectrum at energy E. As a consequence, approximately 10^4 tracks must be measured to give statistical accuracies of the order of 10 % in the neutron fluence spectrum (with a corresponding energy resolution of the order of 10 %). It must be emphasized that spectral measurements determined with NRE in the 4π mode are absolute.

3.1.2 *End-On Mode*—Differential neutron spectrometry with NRE is considerably simplified when the direction of neutron incidence is known, such as for irradiations in collimated or unidirectional neutron beams. In such exposures, the kinematics of (n,p) scattering can be used to determine neutron energy. Observation of proton-recoil direction and proton-recoil track length provide the angle of proton scattering relative to the incident neutron direction, θ, and the proton energy, E_p, respectively. In terms of these observations, the neutron energy, E_n, is simply:

$$E_n = \frac{E_p}{\cos^2 \theta} \tag{3}$$

In collimated or unidirectional neutron irradiations, the emulsion is exposed end-on as depicted in Fig. 1. The end-on mode can be used to advantage in media where neutron scattering is negligible for two types of benchmark field experiments, namely:

3.1.2.1 Benchmark field validation of the NRE method or characterization of point neutron sources, for example, the standard ^{252}Cf neutron field at the National Institute of Standards and Technology (NIST) (**17**).

3.1.2.2 Measurement of leakage neutron spectra at sufficiently large distances from the neutron source, for example, neutron spectrum measurements at the Little Boy Replica (LBR) benchmark field (**18**).

3.2 *Integral Mode*—It is possible to use emulsion data to obtain both differential and integral spectral information. Emulsion work is customarily carried out in the differential mode (**3-6**). In contrast, NRE work in the integral mode is ~~a more recent concept~~ less frequently used and, therefore, a fuller explanation of this approach is included below. In this integral mode, NRE provide absolute integral reaction rates, which can be used in spectral adjustment codes. ~~Before these recent efforts, such codes have not utilized integral reaction rates based on NRE.~~ The significance of NRE integral reaction rates stems from the underlying response, which is based on the elastic scattering cross section of hydrogen. This σ_{np} (E) cross section is universally accepted as a standard cross section and is known to an accuracy of approximately 1 %.

3.2.1 *The I Integral Relation*—The first integral relationship follows directly from Eq 1. The integral in Eq 1 can be defined as:

$$I(E_T) = \int_{E_T}^{\infty} \frac{\sigma(E)}{E} \Phi(E)\, dE \tag{4}$$

Here, $I(E_T)$ possesses units of proton-recoil tracks/MeV per hydrogen atom. Clearly, $I(E_T)$ is a function of the lower proton energy cut-off used for analyzing the emulsion data. Using Eq 4 in Eq 1, one finds the integral relation:

$$I(E_T) = \frac{M(E_T)}{n_p V} \tag{5}$$

$I(E_T)$ is evaluated by using a least squares fit of the scanning data in the neighborhood of $E = E_T$. Alternatively, since:

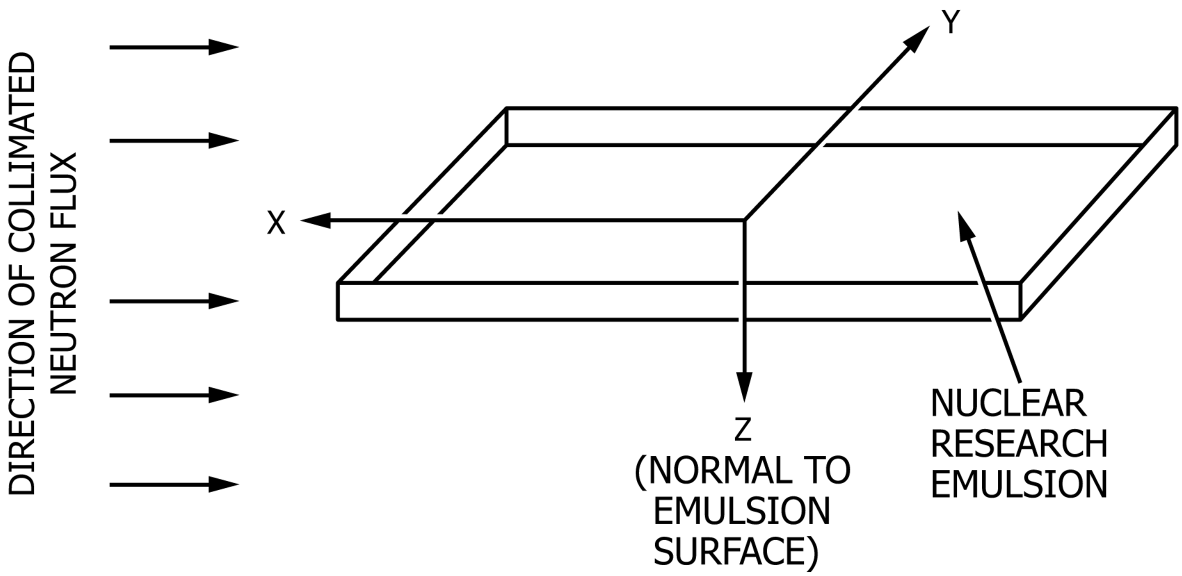

FIG. 1 Geometrical Configuration for End-On Irradiation of NRE

$$M(E_T) = M(R_T)\,\frac{dR(E)}{dE} \tag{6}$$

where: $R(E)$ is the proton-recoil range at energy E in the NRE and dR/dE is known from the proton range-energy relation for the NRE. One need only determine $M(R)$ in the neighborhood of ~~$R = R$~~$R = R_T$. Here M(R) is the number of proton-recoil tracks/µm observed in the NRE. Consequently, scanning efforts can be concentrated in the neighborhood of $R = R_T$ in order to determine $I(E_T)$. In this manner, the accuracy attained in $I(E_T)$ is comparable to the accuracy of the differential determination of $\Phi(E)$, as based on Eq 2, but with a significantly reduced scanning effort.

3.2.2 *The J Integral Relation*—The second integral relation can be obtained by integration of the observed proton spectrum $M(E_T)$. From Eq 1:

$$\int_{E_{\min}}^{\infty} M(E_T)\,dE_T = n_p V \int_{E_{\min}}^{\infty} dE_T \int_{E_T}^{\infty} \frac{\sigma(E)}{E}\,\Phi(E)\,dE \tag{7}$$

where: $E_{\min}$ is the lower proton energy cut-off used in analyzing the NRE data. Introducing into Eq 7 the definitions:

$$\mu(E_{\min}) = \int_{E_{\min}}^{\infty} M(E_T)\,dE_T \tag{8}$$

and:

~~$J(E_{\min}) = \int_{E_{\min}}^{\infty} dE_T \int_{E_T}^{\infty} \frac{\sigma(E)}{E}\,\Phi(E)$ (9)~~

$$J(E_{\min}) = \int_{E_{\min}}^{\infty} dE_T \int_{E_T}^{\infty} \frac{\sigma(E)}{E}\,\Phi(E)\,dE \tag{9}$$

has:

$$J(E_{\min}) = \frac{\mu(E_{\min})}{n_p V} \tag{10}$$

Hence, the second integral relation, namely Eq 10, can be expressed in a form analogous to the first integral relation, namely Eq 5. Here $\mu(E_{\min})$ is the integral number of proton-recoil tracks per hydrogen atom observed above an energy $E_{\min}$ in the NRE. Consequently, the integral $J(E_{\min})$ possesses units of proton-recoil tracks per hydrogen atom. The integral $J(E_{\min})$ can be reduced to the form:

$$J(E_{\min}) = \int_{E_{\min}}^{\infty} \left(1 - \frac{E_{\min}}{E}\right) \sigma(E)\,\Phi(E)\,dE \tag{11}$$

In addition by using Eq 6, the observable $\mu(E_{\min})$ can be expressed in the form:

$$\mu(E_{\min}) = \int_{R_{\min}}^{\infty} M(R)\,dR \tag{12}$$

Hence, to determine the second integral relationship, one need only count proton-recoil tracks above $R = R_{\min}$. Tracks considerably longer than $R_{\min}$ need not be measured, but simply counted. However, for tracks in the neighborhood of $R = R_{\min}$, track length must be measured so that an accurate lower bound $R_{\min}$ can be effectively determined.

4. Significance and Use

4.1 *Integral Mode Dosimetry*—As shown in 3.2, two different integral relationships can be established using proton-recoil emulsion data. These two integral reactions can be obtained with roughly an order of magnitude reduction in scanning effort. Consequently, this integral mode is an important complementary alternative to the customary differential mode of NRE spectrometry. The integral mode can be applied over extended spatial regions, for example, perhaps up to as many as ten *in-situ* locations can be covered for the same scanning effort that is expended for a single differential measurement. Hence the integral mode is especially advantageous for dosimetry applications which require extensive spatial mapping, such as exist in Light Water Reactor-Pressure Vessel (LWR-PV) benchmark fields (see Test Method E1005). In low power benchmark fields, NRE can be used as integral dosimeters in a manner similar to RM, solid state track recorders (SSTR) and helium accumulation monitors (HAFM) neutron dosimeters (see Test Methods E854 and E910). In addition to spatial mapping advantages of these other dosimetry methods, NRE offer fine spatial resolution and can therefore be used *in-situ* for fine structure measurements. In integral mode scanning, both absolute reaction rates, that is $I(E_T)$ and $J(E_{\min})$, are determined simultaneously. Separate software codes need to be used to permit operation of a computer based interactive system in the integral mode (see Section 9). It should be noted that the integrals $I(E_T)$ and $J(E_{\min})$ possess different units, namely proton-recoil tracks/MeV per hydrogen atom and proton-recoil tracks per hydrogen atom, respectively.

4.2 *Applicability for Spectral Adjustment Codes*—In the integral mode, NRE provide absolute integral reaction rates that can be used in neutron spectrum least squares adjustment codes (see Guide E944). In the past, such adjustment codes could not utilize NRE integral reaction rates because of the non-existence of NRE data. NRE integral reaction rates provide unique benchmark data for use in least squares spectral adjustment codes. The unique significance of NRE integral data arises from a number of attributes, which are described separately below. Thus, inclusion of NRE integral reaction rate data in the spectral adjustment calculations can result in a significant improvement in the determination of neutron spectra in low power benchmark fields.

4.3 *The Neutron Scattering Cross Section of Hydrogen*—Integral NRE reaction rates are based on the standard neutron scattering cross section of hydrogen. For fast neutron spectrometry and dosimetry applications, the accuracy of this (n,p) cross section over extended energy regions is essentially unmatched. A semi-empirical representation of the energy-dependence of the (n,p) cross section is given in Eq 13.

$$\sigma_{np}(E) = 3\pi\left[1.206E+\left(-1.860+0.0941491E+0.000130658E^2\right)^2\right]^{-1} + \pi\left[1.206E+\left(0.4223+0.1300E\right)^2\right]^{-1} \quad (13)$$

where: E is in MeV and $\sigma_{np}(E)$ is in barns. This energy-dependent representation of the (n,p) cross section possesses an uncertainty of approximately 1 % at the (1σ) level **(19)**.

4.4 *Threshold Energy Definition*—In contrast with all other fast neutron dosimetry cross sections, the threshold energy of the I and J integral reaction rates can be varied. NRE integral reaction threshold variability extends down to approximately 0.3 to 0.4 MeV, which is the lower limit of applicability of the NRE method. Threshold variation is readily accomplished by using different lower bounds of proton track length to analyze NRE proton-recoil track length distributions. Furthermore, these NRE thresholds are more accurately defined than the corresponding thresholds of all other fast neutron dosimetry cross sections. NRE therefore provide a response with an extremely sharp energy cutoff that is not only unmatched by other cross sections, but an energy threshold that is independent of the *in-situ* neutron spectrum. No other fast neutron dosimetry cross sections possess a threshold response with these significant attributes. The behavior of the I-integral and J-integral response for different threshold energies is shown in Figs. 2 and 3, respectively, in comparison to the threshold ^{237}Np(n,f) reaction used in RM dosimetry.

4.5 *Complimentary Energy Response*—It is of interest to compare the differential energy responses available from these two integral relations. From Eq 4 and 11, one finds responses of the form $\sigma(E)/E$ and $(1 - E_{\min}/E)\sigma(E)$ for the I and J integral relations, respectively. These two responses are compared in Fig. 4 using a common cut-off of 0.5 MeV for both E_T and $E_{\min}$. Since these two responses are substantially different, simultaneous application of these two integral relations would be highly advantageous. As shown in Fig. 4, the energy response of the I and J integral reaction rates complement each other. The J-integral response increases with increasing neutron energy above the threshold value and therefore possesses an energy dependence qualitatively similar to most fast neutron dosimetry cross sections. However, significant quantitative differences exist. As discussed above, the J-integral response is more accurately defined in terms of both the energy-dependent cross section and threshold energy definition. The I-integral possesses a maximum value at the threshold energy and decreases rapidly from this maximum value as neutron energy increases above the threshold value. As can be seen in Fig. 4, the I-integral possesses a much more narrowly defined energy response than the J-integral. While the J-integral response is broadly distributed, most of the I-integral response is concentrated in the neutron energy just above threshold. As a consequence, the I-integral reaction rate data generally provides a more rigorous test of the ability of neutron transport calculations to describe the complex spatial and energy variations that exist in benchmark fields than does the J-integral data. This conclusion is supported by the calculation to experiment ratios (C/E) obtained from NRE experiments in the VENUS-1 LWR-PV benchmark field. For these VENUS-1 NRE experiments, the C/E values for the I integral possessed larger variation and deviated more widely from unity than the corresponding C/E values for the J-integral (**20**).

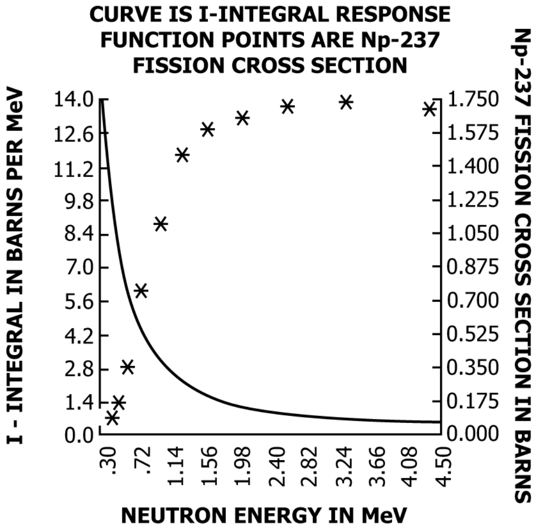

FIG. 2 Comparison of the I-Integral Response with the ^{237}Np (n,f) Threshold Reaction

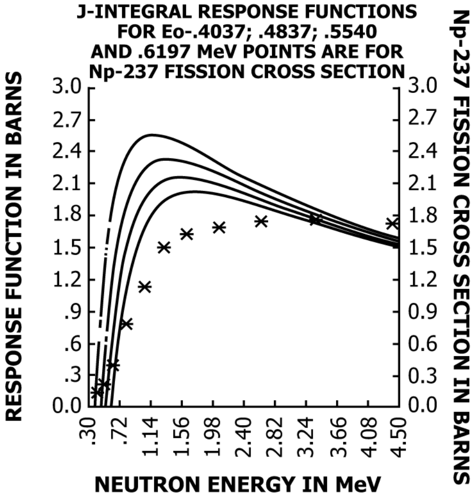

FIG. 3 Comparison of the J-Integral Response for E_T = 0.404, 0.484, 0.554 and 0.620 MeV with the ^{237}Np (*n,f*) Threshold Reaction

5. Apparatus

5.1 *Dark Room*—A dark room equipped with a sink, processing baths and a safe light. There should be adequate bench space in the dark room for pre-irradiation preparation of NRE as well as for the transfer of NRE between processing trays.

5.2 *Constant Temperature Baths*—The constant temperature baths in the dark room should possess temperature control to 0.1°C. One cooling bath should be equipped with a circulating pump so that tap water can be circulated through the coils of the processing bath. One thermostatically controlled processing bath.

5.3 *Refrigerator*—The dark room should be equipped with a refrigerator for storing reagents and chemicals.

5.4 *Stainless Steel Trays*—Stainless steel (SS) trays and cover lids are required, approximately 25 by 15 cm in area by 2.5 cm deep, for NRE processing.

5.5 *Racks*—Racks are required to position and hold the SS trays in the constant temperature baths. These racks hold the SS trays in the constant temperature bath so that the top of the SS trays project above the bath surface by approximately 0.5 cm.

5.6 *Cooling Coil*—A cooling coil is required that is immersed in the constant temperature bath and connected by a suitable tube to the ~~cold water~~ cold-water tap. Another identical tube must serve as a drain line from the cooling coil to the sink. An in-line valve for control of tap water flow should be installed so that a small steady stream of water can be regulated.

5.7 *Optical Microscopes*—Optical microscopes are required for NRE scanning with a magnification of 1000X or higher, utilizing oil immersion techniques. Microscope stages should be graduated with position readout to better than 1 µm and should also possess at least ~~1 µm~~ 1-µm repositioning accuracy. The depth of focus (z-coordinate) should be controlled to the nearest 0.1 µm with similar repositioning accuracy. Calibrated stage micrometers and graduated eyepiece grids (reticles) are also required for track scanning.

5.8 *Filar Micrometer*—A filar micrometer is required for measuring thickness with electronic readout to at least the nearest 0.1 µm.

5.9 *Dial Gages*—Dial thickness ~~gages~~ gauges, preferably with digital readouts, are required with readout scales of at least 2 µm per division.

5.10 *Certified Gage Blocks*—Certified gauge blocks in the anticipated NRE thickness range are required to verify the accuracy of thickness measurements.

5.11 *Scribes*—Diamond point scribes are required for marking NRE glass backing with suitable pre-irradiation identification labels

5.12 *Thermometers*—Thermometers are required for measuring temperature with readout to at least the nearest 0.1°C.

5.13 *Interactive Scanning System*—A computer based interactive scanning system is required for the measurement of proton-recoil track length in NRE. Hardware and software requirements are described in Section 9.

6. Reagents and Materials

6.1 *Purity of Reagents*—Distilled or demineralized water and analytical grade reagents should be used at all times.

6.2 *Reagents*—Tables 1-4 provide detailed specifications for the processing solutions.

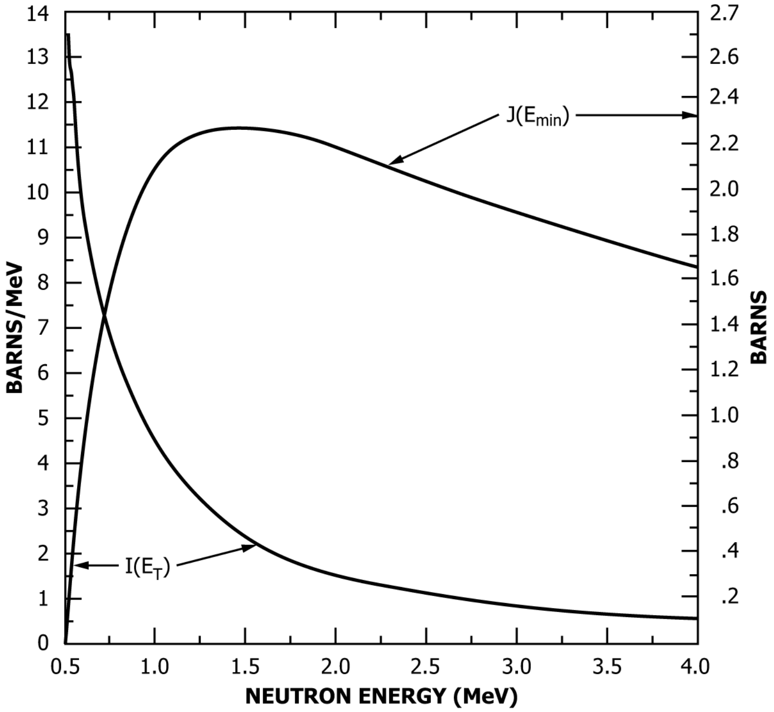

FIG. 4 Energy Dependent Response for the Integral Reactions $I(E_T)$ and $J(E_{min})$

TABLE 1 Developing Solution[A]

Reagent	Volume/Mass
Distilled Water	1.0 L
Boric Acid Crystals	3.0 g
Potassium Bromide	1.0 g
Desiccated Na_2SO_3	50 g
Amidol	2.0 g
Anti-Fog Solution	6.0 cc

[A] ~~Chemicals~~ Chemicals dissolved in order listed at room temperature.

TABLE 2 Anti-Fog Stock Solution

Reagent	Volume/Mass
Ethylene Glycol (50°C)	175 cc
Kodak Anti-Fog #1[A]	41.68 g
Ethylene Glycol	≈ 75 cc[B]

[A] ~~Dissolve~~ Dissolve in warm ((50°C)) Ethylene Glycol

[B] ~~Cool~~ Cool to 24°C and ~~Add~~add cool Ethylene glycol to make 250 cc.

TABLE 3 Fixing Solution[A]

Reagent	Volume/Mass
Distilled Water	1 L
$Na_2S_2O_3$ (Hypo)	400 g
$NaHSO_3$[B]	10 g

[A] ~~Chemicals~~ Chemicals dissolved in order listed at room temperature.

[B] ~~If~~ If $Na_2S_2O_5$ is used, decrease mass by a factor of 0.87.

6.2.1 *Developing Solution*—As specified in Table 1, Amidol, 2,4–Diaminophenol Dihydrochloride is used to develop the NRE (Eastman Organic Chemicals, No. P 614, other commercially prepared amidol developers also work well.) The anti-fog solution specified in Table 2 is used to suppress chemical fog and prevent the development of gamma-ray induced electron tracks and thereby improve proton-recoil track length measurements.

6.2.2 *Stop Bath Solution*—The stop bath solution should be a 1 % glacial acetic acid in distilled water.

6.2.3 *Fixing Solution*—A fixing solution containing sodium thiosulfate (hypo) and sodium bisulfite is required (see Table 3).

6.2.4 *Drying Solutions*—Two drying solutions of glycerine, ethyl alcohol, and distilled water are required (see Table 4).

TABLE 4 Drying Solutions

Reagent	Volume, % Solution 1	Volume, % Solution 2
Distilled Water	35	0.00
Glycerine	30	30
Ethyl Alcohol (95%)[A]	35	70

[A] ~~Absolute~~ Absolute alcohol should not be used, since it contains traces of benzene.

6.3 *Materials:*

6.3.1 *Emulsions*—Ilford type L-4 NRE, 200- and 400-µm thick pellicles, mounted on glass backing. The glass backing is approximately 2.5 by 7.5 cm in area by 1 mm thick.[4]

7. Pre-Irradiation NRE Preparation

7.1 *NRE Preparation*—Care should be taken to handle NRE by the edges to avoid potential damage to surfaces adjacent to measurement locations both during preparation and after the measurements. The NRE should be cut to an acceptable size in the dark room. A safe light with a yellow filter may be used. The diamond point scribe should be used to rule the glass backing undersurface of the NRE and the glass backing can then be snapped along the rule marks to obtain the desired NRE dosimeter size. NRE dosimeters down to approximately 5 mm by 5 mm area can be readily obtained. The diamond point scribe should then be used to mark an ID number on the undersurface of the glass backing. The NRE should then be wrapped in lens paper and then in aluminum foil (~0.002 cm thick) for further handling and to prevent exposure to light. The NRE ID number can then be written on the Al-foil wrapping with an indelible pen. If it is necessary to know the orientation of the NRE in the irradiation field, the undersurface NRE glass backing is marked with an indelible pen to provide a known orientation for the NRE. This marking orientation must then be transcribed to the Al-foil wrapping. The NRE can then be removed from the dark room. However, if the NRE are to be deployed in Al or Cd buckets for the irradiation, this assembly procedure should also be conducted in the dark room if at all possible. It will then be necessary to transcribe the NRE ID number and orientation information to the outer surface of the irradiation bucket. Spacers or other means should be employed to maintain the NRE orientation within the bucket to the desired accuracy. A knowledge of NRE orientation together with a complete record of proton-recoil scanning data (see Section 9 can then be used to determine any anisotropy of the *in-situ* neutron field.

7.2 *NRE Exposure Time*—Neutron fluences of approximately 10^5 cm^{-2} will give optimum track densities for scanning. Fluences greater than 10^6 cm^{-2} for manual scanning and 10^7 cm^{-2} for computer-based scanning will result in unacceptably high track densities.

7.3 *NRE Thickness Measurement*—To measure the original thickness of the emulsion, HO, place the glass undersurface of the NRE on a flat surface in the dark room. Use the dial thickness gauge to measure the thickness of the emulsion and glass backing. Repeat this measurement five to ten times at different locations so that a precise average is obtained. The glass backing thickness is determined after irradiation and post-irradiation processing (see 8.8).

8. Post-Irradiation Processing Procedures

8.1 Processing procedures will depend to some extent on the particular batches of Ilford NRE that are used. Consequently, while the processing procedures recommended below will not necessarily be optimum for any given batch, these procedures can be used as a starting point to attain optimum procedures desired for the specific NRE neutron dosimetry application under consideration. Table 5 summarizes the various steps utilized in the post-irradiation NRE processing procedures.

8.1.1 *Pre-Soaking Step*—Use a mixture of approximately 50 % distilled water and 50 % ethylene glycol in the cooling bath to maintain a temperature of 2°C. Fill a SS tray with distilled water. Pre-cool the distilled water soaking solution to 5°C before inserting the NRE into the distilled water. This will keep the NRE swelling to a minimum. Insert the SS trays into the 2°C bath. The purpose of the pre-soaking step is to facilitate uniform penetration of the Amidol developer throughout the full thickness of the NRE. In this way, development will be uniform, that is, independent of depth (denoted by the z coordinate). Pre-soak 200 µm L-4 NRE for 1 h and 400 µm L-4 NRE for 2 h.

8.2 *Developing Step at 1.2°C*—Prepare a fresh development solution as prescribed in Tables 1 and 2. Place the development solution in a SS tray and insert the tray into the cooling bath at 1.2°C. Transfer the NRE directly from the pre-soaking solution to the development solution. The rate of NRE development is very sensitive to the temperature of the developer. Use of the low 1.2°C temperature provides enhanced developer penetration with very little actual development. The length of time the NRE remain in the 1.2°C developer depends on the NRE thickness. Develop Ilford L-4 200 µm and 400 µm NRE for approximately 1 h and 2.5 h, respectively.

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[4] Details of NRE characteristics and specifications can be found at ~~http://www.polysciences.com/Catalog/Department/81/categoryid-49/.~~ http://www.polysciences.com/default/ilford-emulsions-l4.

TABLE 5 Summary of NRE Processing Steps

Step	Solution	Temperature, °C	Time Duration	
			200 µm^A	400 µm^A
~~Pre-soaking~~	~~Distilled H_2O~~	~~2~~	~~1h~~	~~2h~~
Pre-soaking	Distilled H_2O	2	1 h	2 h
~~Developing-1~~	~~See Tables 1 and 2~~	~~1.2~~	~~1h~~	~~2.5h~~
Developing-1	See Tables 1 and 2	1.2	1 h	2.5 h
Developing-2	See Tables 1 and 2	5	35 to 40 min	35 to 40 min
Stop Bath	1 % Glacial Acetic Acid	5	15 to 20 min	1h
~~Fixing~~	~~See Table 3~~	~~5~~	~~2h to 1 Day~~	~~2 to 3 Days~~
Fixing	See Table 3	5	2 h to 1 day	2 to 3 days
~~Washing~~	~~Tap Water~~	~~6~~	~~1 Day~~	~~1 Day~~
Washing	Tap Water	6	1 day	1 day
~~Drying-1~~	~~See Table 4~~	~~5~~	~~1h~~	~~2.5 h~~
Drying-1	See Table 4	5	1 h	2.5 h
~~Drying-2~~	~~See Table 4~~	~~5~~	~~1h~~	~~2.5 h~~
Drying-2	See Table 4	5	1 h	2.5 h

^{A} ~~Ilford~~ Ilford L-4 NRE thickness in µm.

8.3 *Developing Step at 5°C*—Transfer the tray containing the NRE in the development solution from the cooling bath at 1.2°C to the processing bath which is maintained at 5°C. Here a development time of approximately 35 to 40 min can be used, independent of NRE thickness.

8.4 *Stop-Bath Step*—The stop-bath solution (1 % glacial acetic acid in distilled water) should be pre-mixed and stored in a plastic bottle in the refrigerator. Fill another SS tray with stop-bath solution and place the tray in the processing tank so it cools to the 5°C temperature of the processing bath. Remove both trays from the processing bath and place the trays on a convenient flat surface in the dark room. Rapidly transfer the NRE from the developer tray into the stop-bath tray and place the stop-bath tray back into the processing bath. Care should be exercised to avoid touching the NRE surface. The NRE should be handled by holding the glass backing. The time duration that the NRE remain in the stop-bath solution depends on NRE thickness. For ~~200 µm~~ 200-µm NRE, approximately 15 to 20 min will do, whereas approximately 60 min should be used for ~~400 µm~~ 400-µm NRE. The stop-bath solution changes the pH of the NRE to stop development.

8.5 *Fixing Solution Step at 5°C*—The fixing solution (see Table 3) should be pre-mixed and stored in a plastic bottle in the refrigerator. Remove the fixing solution from the refrigerator and fill a SS tray at least half-way with the fixing solution. Place the SS tray in the processing bath until the fixing solution comes to equilibrium at 5°C. Remove both the stop-bath tray and the fixing solution trays from the processing bath onto a convenient flat surface in the dark room. Rapidly transfer the NRE from the stop-bath tray to the fixing solution tray. Replace the fixing solution tray back into the processing bath. The fixing solution dissolves the undeveloped silver bromide grains in the NRE, so that the NRE become transparent for track scanning purposes. The residence time of the NRE in the fixing solution depends on NRE thickness. For ~~200 µm~~ 200-µm NRE, it takes several hours to a day. For ~~400 µm~~ 400-µm NRE, it can take several days. The NRE should remain in the fixing solution approximately 1.5 times the time duration that it takes for the NRE to clear. When using the fixing solution for several days, as needed for the ~~400 µm~~ 400-µm NRE, replenish the fixing solution at least once a day by pouring off 50 % of the old solution and adding 50 % new fixing solution.

8.6 *NRE Washing*—The fixing solution needs to be thoroughly removed from the NRE. This washing process can be done in daylight. Actually, the darkroom (bright) lights can be turned on as soon as the fixing process is completed. Wash the NRE with a stream of tap water, which runs through coils at the bottom of the 1.2°C cooling bath. Let cold tap water run slowly through a tube to the coils in the cooling bath and then through a tube to an empty SS tray in the sink. A good control valve is needed on the tube carrying the cold tap water so as to ensure a good even flow of water. When the water in the tray has ~~reaches~~reached a temperature of ≈6°C, transfer the fixed emulsions from the SS tray in the 5°C bath to the tray in the sink. The tap water should run into the SS tray slowly so as not to produce a significant stream that might distort the emulsions. The NRE should be washed in this manner for approximately 24 h.

8.7 *Drying Solution Steps*—The NRE obtained from the washing step are swelled with water to two to three times the original thickness. The purpose of the drying solution step is to remove the water and thereby reduce distortion and at the same time provide for more precise thickness (z-coordinate) measurements. Two drying solutions are prepared as prescribed in Table 4. The two drying solutions are placed into SS trays and allowed to come to equilibrium in the 5°C processing bath. Using some convenient flat surface in the dark room, the NRE are transferred from the washing tray to SS tray containing the first drying solution. The SS tray containing the NRE in the first drying solution is placed in the 5°C processing tank for approximately 1 h for ~~200 µm~~ 200-µm NRE and 2.5 h for ~~400 µm~~ 400-µm NRE. This step is repeated with the second drying solution. Upon removal from the second drying solution, the NRE can be placed (emulsion side up) on a flat surface so that the NRE can be gently blotted to remove the excess drying solution. The NRE are then air dried at room temperature for at least 24 h. In this drying process, the water in the NRE is replaced with alcohol which, in turn, evaporates and glycerine replaces the silver bromide that was in the unprocessed emulsions. (Actually, the glycerine fills the holes from which the silver bromide was removed in the fixing process.)

8.8 *Post-Irradiation NRE Thickness Measurements*—After processing is complete, place the NRE on edge under a microscope equipped with a filar micrometer eyepiece. Measure the glass backing and processed emulsion thickness separately. Make 5 to 10 observations of each thickness so that a precise average of both the emulsion thickness and the glass backing thickness can be determined. These data provide the processed NRE thickness, HP, and the glass backing thickness can then be used with the pre-irradiation thickness measurements to determine the original NRE thickness, HO. To obtain the true z-coordinate position in the (irradiated) NRE, z_{tr}, from the observed z-coordinate, z_{obs}, one must use the relation

$$z_{tr} = \frac{HO}{HP} z_{obs} \tag{14}$$

8.9 *NRE Microscope Mounting*—The processed NRE is mounted on a watch glass (microscope cover glass), which is cemented in a rigid frame that can, in turn, be attached to the microscope stage. The rigid frame must not obstruct the portions of the NRE that are to be scanned, and the microscope cover glass is permanently glued or cemented to the frame. The NRE is than placed on the microscope glass and glued or cemented to the glass at several edge locations to ensure that the NRE will maintain a constant position relative to the frame during measurements. When NRE are not being scanned, they should be stored in Petrie dishes under controlled temperature and humidity conditions. Standard room temperature is acceptable, but 50 % relative humidity is preferable, since the glycerine is somewhat hydroscopic. Large changes in humidity during storage should be avoided.

9. Track Scanning

9.1 *Instrumentation*—The principal disadvantage of the NRE method of fast neutron dosimetry is the need to measure proton-recoil track length for many tracks. Accurate differential spectrometry measurements require measurement of approximately 10^4 tracks, so that many hours of scanning are required. To facilitate proton-recoil track length measurements and provide a much more cost-effective measurement system, a computer-based interactive system is indispensable. Such a system can store all the (3D) scanning information, in detail, as well as provide on-line computations for individual track analysis. To conduct all of these operations manually would be impractical. Such a computer-based interactive system was built and used successfully for NRE neutron dosimetry some time ago (**17**). The specifications and scanning procedures described here are based on this system, which was called the Emulsion Scanning Processor (ESP). Since the ESP system was built some time ago, many of the components of the ESP system are outdated because of the rapid development of computer technology that has ensued. Consequently, to fabricate such a system today, one should only use this description as an overall guide and replace all components with state-of-the-art components, which will be invariably faster, more reliable and less expensive.

9.1.1 *Overall Design*—Some of the major considerations in the development of an interactive system for NRE scanning are simplicity, ease of operation, stability, and reliability of performance. In the design of the interactive system, the flexibility and power of computer control should be utilized to the maximum possible extent. A photograph of the ESP system microscope, and joystick control boxes with push buttons is shown in Fig. 5. An operator must interact with the system to obtain the desired results. The joystick and push button controls are used to set parameters and boundaries, focus, locate tracks, measure track lengths, categorize, and store track data. The *(X, Y, Z)* stage motion, including depth, that is, focus, of the microscope is performed by the computer under operator control. The computer receives all operator instructions, moves the stage as directed, and stores positional information on command. Software programs, stored on computer disks, provide the flexibility needed to conveniently tailor operating, storage, and data presentation formats to satisfy different experiments and scanning modes.

9.2 *Scanning Coordinate Systems*—A Cartesian coordinate system (*G, R, S*) is used to describe field locations in the emulsion; whereas, another Cartesian coordinate system (*X, Y, Z*) is used to describe track-ending locations in the emulsion. The perimeters of a reticle located in the eyepiece of the microscope serve as the *G, R* boundaries of the field of view. The *S*-coordinate is the depth or focus coordinate. In using the interactive (ESP) system, the selected emulsion is divided into a number of field volumes. The volume of a field corresponds to the area of a field of view times the preselected depth of the emulsion as shown in Fig. 6. The distance ΔS is prescribed in order that scanning be primarily confined to the interior of the emulsion, where proton-recoil escape probabilities are either negligible or small. Hence, a field volume FV is given by the relation:

$$FV = \Delta G \cdot \Delta R \cdot S \tag{15}$$

To provide orientation for track scanning from day-to-day as well as between different scanners, a zero reference point must be chosen on the emulsion. To this end, a needle having a red dye on its tip is mounted on a microscope objective holder and is used to pierce the emulsion surface thereby leaving a red spot. A color microphotograph is taken of the spot. A proton track escaping from the top surface of the emulsion is selected near the spot. The zero reference point, G = O, R = O, S = O, is then stored as the point of escape of this proton-recoil track.

9.3 *Track Scanning*—The actual measurement of a typical track in an emulsion using the interactive ESP system is described below. Fig. 5 shows the ESP controls in more detail. Operations with the left (L) and right (R) push buttons are summarized in Table 6. The left joystick controls the *Z* (focus) position, whereas, the right joystick controls both *X* and *Y* positions. The design of the interactive system permits performance of all track scanning and recording activities without interrupting the observation of proton-recoil tracks in the NRE.

9.3.1 *Recording the Zero Reference Point*—The emulsion is clamped to the microscope stage, and the operating program for the interactive (ESP) system is initiated in the computer system. The first step is to bring the zero reference point into focus under the

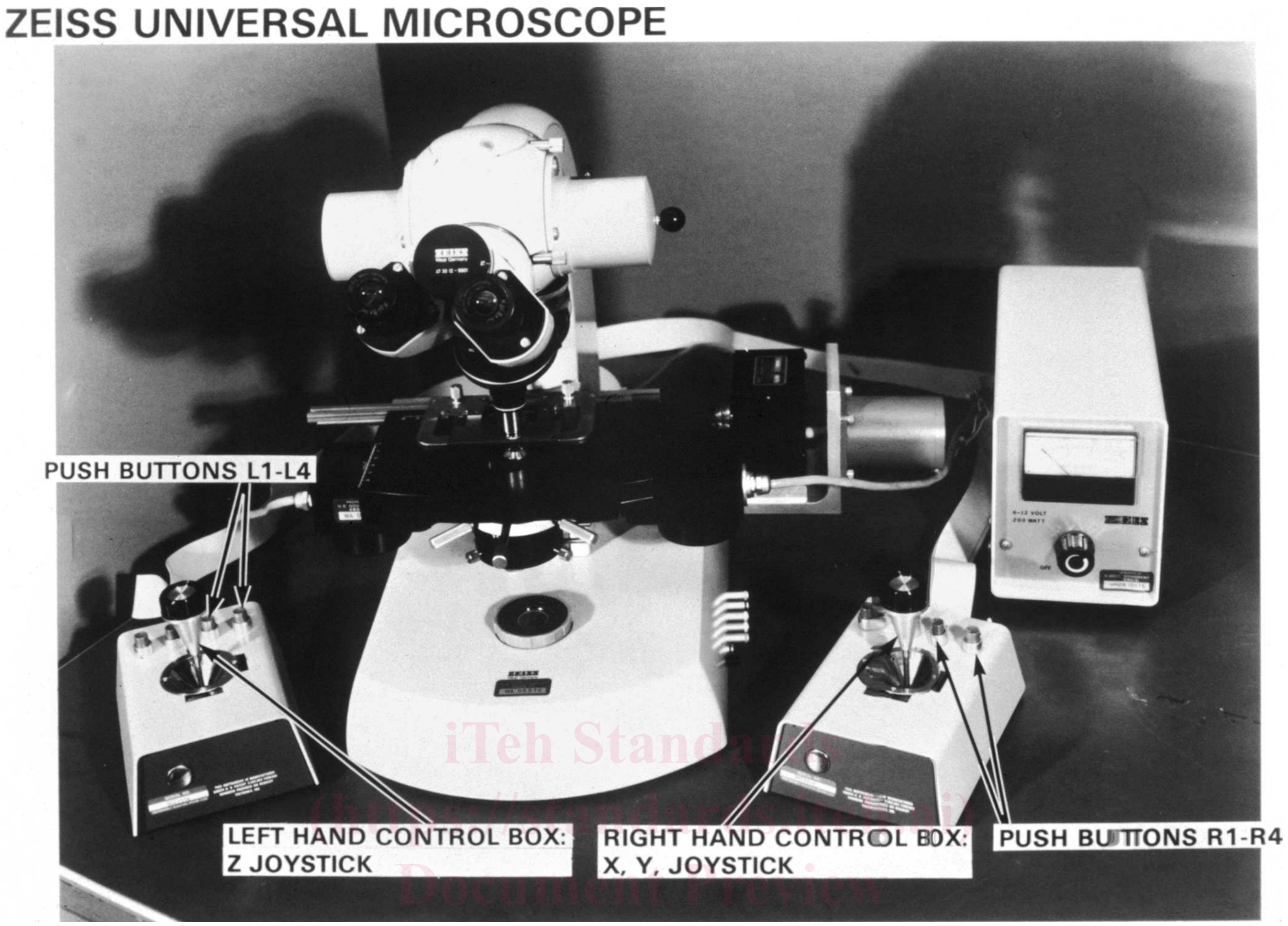

FIG. 5 Close Up of the ESP Microscope Showing Push Buttons and Stage Controls

reticle cross hair, that is, the exact center of the reticle grid. Pressing push button 3R (see Table 6) stores the coordinates of the zero reference point $G = 0$, $R = 0$, $S = 0$ on a (scanning) data disk.

9.3.2 *Field of View Parameters*—The scanner must then measure a number of parameters that are to be used in the analysis of the proton-recoil track scanning data. The field width ΔG and field height ΔR must both be measured. To measure the field width ΔG, a track ending is moved to the left edge of the field reticle and its position is stored. Next, the same track ending is moved to the right edge of the field reticle and its new position is stored, whereupon the computer will automatically retrace and calculate the field width ΔG in µm. The field height ΔR is measured in the same fashion except the track ending is moved to the top and bottom edges of the reticle. These measurements can be repeated to obtain average values for ΔG and ΔR in establishing field-of-view parameters. Other parameters that must be operator-prescribed include the area limits, G_{max} and R_{max}, which define the maximum traverses in the G and R directions, respectively, as shown in Fig. 6. In addition, the operator must enter the distance ΔS (which defines the interior emulsion depth S) as well as the original thickness, HO, of the NRE before processing. With these operations completed, the computer will calculate the number of fields of view contained within the boundary prescribed by G_{max} and R_{max} as depicted in Fig. 6.

9.3.3 *Field of View Selection*—The microscope is then automatically moved to a field of view to be scanned. Fields can be scanned either sequentially by rows, where consecutive fields are ordered according to the matrix (G_kR_j) or by directing the interactive system to a prescribed field (G_kR_j), which is obtained by entering the integers (k,j) at the computer keyboard.

9.3.4 *NRE Thickness Measurement*—When the field of view is reached, the emulsion thickness is measured by first focusing on the top surface, then on the bottom surface, and recording the respective S levels with push button 2R. The computer calculates the NRE thickness and automatically returns the focus to the preselected depth ΔS, as measured from the top of the emulsion (see Fig. 7). In this way, the thickness of the processed NRE is measured for each field that is scanned. The initials of the operator must be entered for each field scanned. This permits comparisons between scanning results of different individuals as well as comparisons between an individual scanner and the mean results obtained by a group of observers. Consequently, the objectivity and accuracy of any given scanner can be examined.

9.3.5 *Track Selection*—The field is scanned by continuously increasing the S coordinate with the left joystick. The microscope focus is moved slowly downward in the NRE until one end of a track comes into focus. This track ending is moved until it is