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

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. 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 anytime 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 three 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 3R, 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 3μ in NRE can not be adequately measured with optical microscopy techniques. As proton-recoil track length decreases below approximately 3μ, it becomes very difficult to measure track length accurately. This 3μ 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.

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 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 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 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 and the J-integral (**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.

2. Referenced Documents

2.1 *ASTM Standards:*[3]

E706 Master Matrix for Light-Water Reactor Pressure Vessel Surveillance Standards, E 706(0) (Withdrawn 2011)[4]

E854 Test Method for Application and Analysis of Solid State Track Recorder (SSTR) Monitors for Reactor Surveillance, E706(IIIB)

E910 Test Method for Application and Analysis of Helium Accumulation Fluence Monitors for Reactor Vessel Surveillance, E706 (IIIC)

E944 Guide for Application of Neutron Spectrum Adjust-

ment Methods in Reactor Surveillance, E 706 (IIA)

E1005 Test Method for Application and Analysis of Radiometric Monitors for Reactor Vessel Surveillance, E 706 (IIIA)

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:

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

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

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energy, E_p, respectively. In terms of these observations, the neutron energy, E_n, is simply:

$$E_n = \frac{E_p}{\cos^2 \theta} \quad (3)$$

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 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:

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

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:

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

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

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

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_T. Here M(R) is the number of proton-recoil tracks/micron 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:

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

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

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

and:

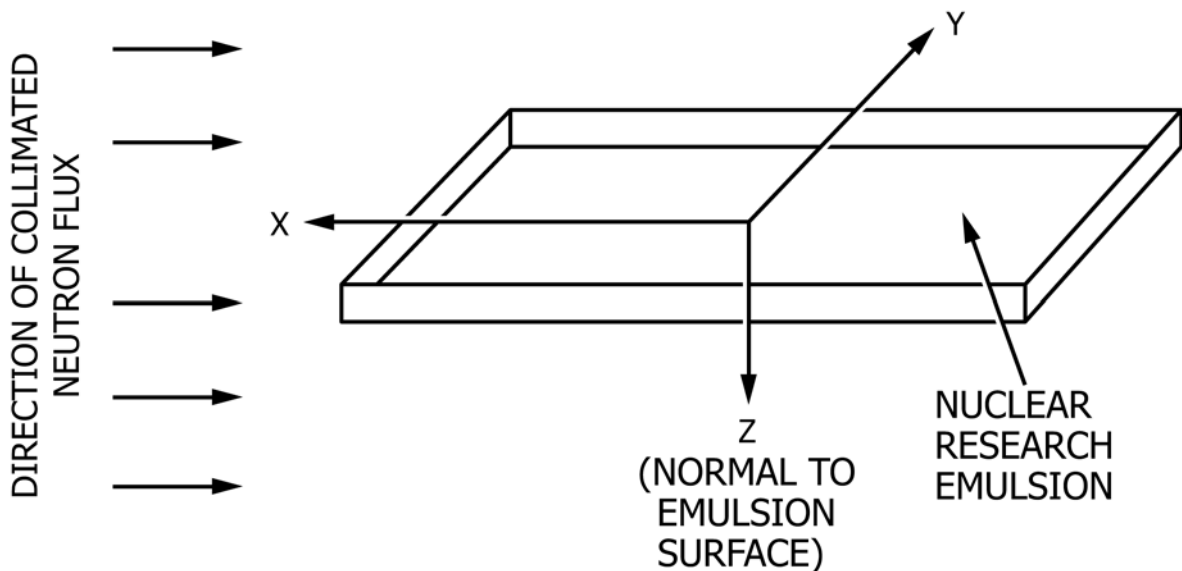$$J(E_{min}) = \int_{E_{min}}^{\infty} dE_T \int_{E_T}^{\infty} \frac{\sigma(E)}{E} \Phi(E) dE \quad (9)$$

has:

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

DIRECTION OF COLLIMATED NEUTRON FLUX

X

Y

Z (NORMAL TO EMULSION SURFACE)

NUCLEAR RESEARCH EMULSION

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

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:

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

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

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

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.

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

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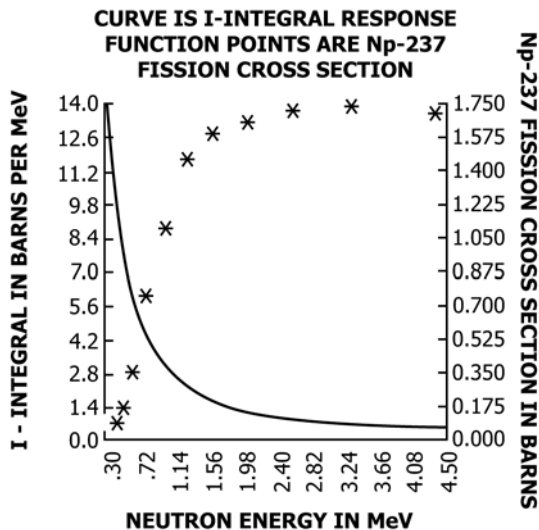
FIG. 2 Comparison of the I-Integral Response with the ^{237}Np (n,f) Threshold Reaction
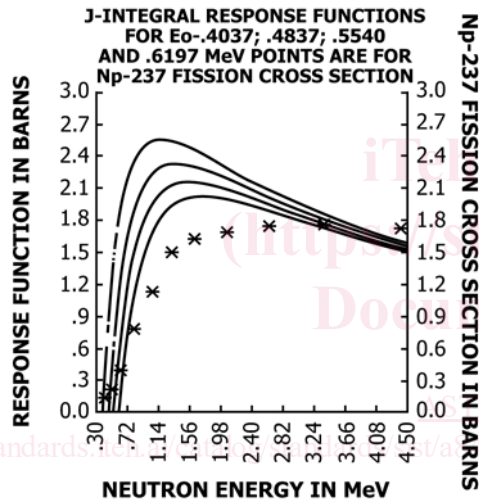

FIG. 3 Comparison of the J-Integral Response for E_T = 0.404, 0.484, 0.554 and 0.620 MeV with the ^{237}Np (n,f) Threshold Reaction

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).

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 posses 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 which and connected by a suitable tube to the 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 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 are required with readout scales of at least 2 μm per division.

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

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

5.12 *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.
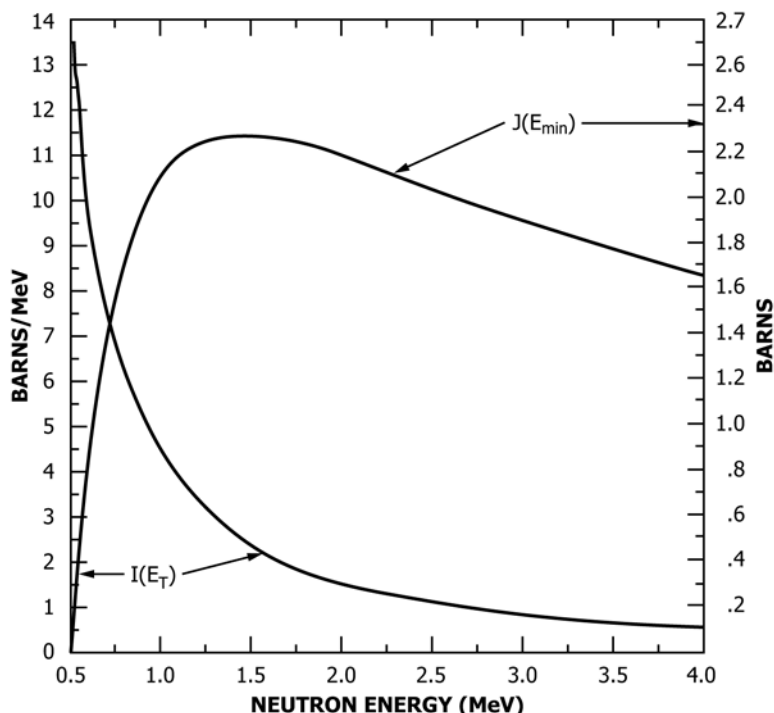
FIG. 4 Energy Dependent Response for the Integral Reactions $I(E_T)$ and $J(E_{min})$

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

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 acedic 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).

6.3 *Materials:*

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 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 in warm ((50°C)) Ethylene Glycol
[B]Cool to 24°C and 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 dissolved in order listed at room temperature.
[B]If $Na_2S_2O_5$ is used, decrease mass by a factor of 0.87.

TABLE 4 Drying Solutions

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

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

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.[5]

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[5] Details of NRE characteristics and specifications can be found at http://www.polysciences.com/shop/assets/datasheets/271B.pdf.