

## A Differential Dosimetry of Pile Radiations Using $\text{Fe}^{2+}$ Oxidation

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Biological material irradiated in the pile is affected by different kinds of radiation<sup>1</sup>: (external)  $\gamma$ -radiation, fast, slow and thermal neutrons. The biological action of the latter is chiefly due to nuclear reactions with H, leading to (internal)  $\gamma$ -radiation through  $\text{H}(n)\text{D}$ , and with N, B and, in certain cases, Li, leading to the emission of heavy ionizing particles formed through the reactions  $\text{N}^{14}(\text{np})\text{C}^{14}$ ,  $\text{B}^{10}(\text{n}\alpha)\text{Li}^7$ , and  $\text{Li}^6(\text{n}\alpha)\text{T}$  respectively. Since these radiations have very different biological effectivities, mainly due to differences in ionization densities<sup>2</sup>, it is necessary to determine their relative importance in the total dose given. In the present communication the authors show the possibility of measuring the doses of  $\gamma$ -radiation (+ non-thermal neutrons — probably a small fraction) and thermal neutrons separately through the determination of the yield of the reaction  $\text{Fe}^{2+} \rightarrow \text{Fe}^{3+}$ , occurring in solutions of  $\text{FeSO}_4$  in 0.1 N  $\text{H}_2\text{SO}_4$ . It is further demonstrated that the relative chemical efficiencies of the different kinds of radiation can easily be determined.

In a preliminary series of experiments pairs of silica tubes containing 3 ml of air-saturated  $\text{FeSO}_4$ - $\text{H}_2\text{SO}_4$  solution, one solution with, and the other without, an added amount of  $\text{H}_3\text{BO}_3$  or  $\text{Li}_2\text{SO}_4 \cdot \text{H}_2\text{O}$  have been irradiated simultaneously and at the same position in the Kjeller heavy water pile. The reaction yield in a solution without B or Li is due to  $\gamma$ -radiation (+ fast neutrons), whereas the yield increment in the solutions containing B or Li is due to the recoil nuclei produced in reactions with thermal neutrons. The yields of  $\text{Fe}^{3+}$  were determined spectro-photometrically at 520  $m\mu$  after

the addition to the irradiated samples of 0.7 ml of 3 N KSCN. The  $\text{Fe}^{3+}$  concentrations corresponding to the extinctions were obtained from a colorimetric determination of  $\text{FeSO}_4$ - $\text{Fe}_2(\text{SO}_4)_3$  mixtures in the different milieus. The fact that  $\text{Li}_2\text{SO}_4$  (concentrations up to 0.22 moles/l were used) lowered the extinction was allowed for. Admixtures of  $\text{H}_3\text{BO}_3$  (concs. up to 0.08 moles/l) caused no change of the colour intensity.

The G values (number of ions oxidized per 100 eV absorbed) for X-rays were controlled for all solutions (roentgen apparatus run at 175 kV, filtration 0.5 mm Cu + 1 mm Al, dose rate, measured with a Victoreen type chamber, 50 r/min). The absorption of X-ray energy per ml was determined regarding absorption coefficients and densities<sup>3</sup>. Influence of eventual pH changes on the reaction yield was negligible. For X-rays a G value of 18.7 ions oxidized per 100 eV absorbed was found. When the pH dependence<sup>6</sup> of the reaction is considered, this is in close agreement with earlier values<sup>4,5</sup> of about 21 obtained in 0.8 N  $\text{H}_2\text{SO}_4$ . Simultaneously with the irradiation of the solutions the neutron flux was determined by activating sodium carbonate, followed by an absolute measurement of  $\text{Na}^{24}$  in a NaI(Tl) scintillating crystal<sup>7</sup>. In more accurate work a correction has to be made for the neutron absorption through H, B or Li in the solutions. Detailed experimental data will be published in a further communication.

From the neutron densities thus determined and from the known values of neutron cross sections of the nuclear reactions and reaction energies (see Table 1), the oxidizing efficiencies of the particle radiations relative to that of X-rays could be determined, *i.e.* the G values for these radiations (Table 1).

*Conclusions:* 1. The densely ionizing radiations give a lower reaction yield than do X- and  $\gamma$ -radiations. This agrees with the scant information from experiments with  $\alpha$ -rays from Po and Rn<sup>8,9</sup>. The relatively high yield (Table 1) of the products from  $\text{Li}^6(\text{n}\alpha)\text{T}$  is probably due to the less densely ionizing triton, the relative efficiency of which is calculated to be not far from 1, the efficiency of the  $\alpha$ -particle being regarded equal to that of the recoil nuclei produced through  $\text{B}^{10}(\text{n}\alpha)\text{Li}^7$ . This finding

Table 1.

Nuclear reaction	Reaction		Ionizing particle			Oxidizing efficiency rel. to X-rays
	energy (MeV)	cross section (cm <sup>-24</sup> )	type	energy (MeV)	spec. ionization * ion pairs/ $\mu$ H <sub>2</sub> O	
B <sup>10</sup> (n $\alpha$ )Li <sup>7</sup>	2.34 (+0.44 $\gamma$ )	3 990	$\alpha$ Li <sup>7</sup>	1.49 0.85	5 500 5 500 **	0.24 { 0.24 0.24
Li <sup>6</sup> (n $\alpha$ )T	4.78	910	$\alpha$ T	2.05 2.73	5 500 1 150	0.59 { 0.24 0.83
			e <sup>-</sup> from * X-rays	$\sim$ 0.015	100	1.00

\* Calcd. from range data given by Lea<sup>10</sup> and Siri<sup>11</sup> and from the assumption of an energy dissipation of 32.5 eV per ionization.

\*\* Calcd. from the range of the Li ion (*i.e.* Li<sup>3+</sup>  $\rightarrow$  Li<sup>2+</sup>  $\rightarrow$  Li<sup>+</sup>) given by Rossi and Staub (Ionization chambers and counters. New York, 1949, p. 187).

confirms the reliability of the fast neutron dosimetry<sup>3</sup> based on the oxidation of Fe<sup>2+</sup>, the mean ionization density produced here being about 700.

2. The oxidation yield produced by the recoil nuclei is independent of the Fe<sup>2+</sup> concentration within the region 0.3–3.0 mmoles/l.

3. The oxidation yields are independent of the thermal neutron intensity within the region examined, 0.27–4.54  $\cdot 10^{11}$  n cm<sup>-2</sup> sec<sup>-1</sup>, and of the simultaneous  $\gamma$  intensities between 800 and 9 000 rep min<sup>-1</sup>. After a standardization of the yield increment due to B or Li, the system described can thus be used for a determination of thermal neutron densities, irrespective of the contaminating  $\gamma$ -radiation.

4. A determination of the internal  $\gamma$ -radiation (and the form factor for absorption) from n $\gamma$  captures in the irradiated object might be determined by a similar method after shielding off the external  $\gamma$ -radiation.

The experiments are continued with ionizing particles giving other ionization den-

sities, *e.g.* protons from N(np)C<sup>14</sup> and fission fragments, and also with other chemical reactions, *e.g.* Ce<sup>4+</sup>  $\rightarrow$  Ce<sup>3+</sup>.

The investigation has been supported by the Knut and Alice Wallenberg Foundation and by the Hierta-Retzius Research Foundation.

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Received October 20, 1952.