

in error. Using the variational values for  $A$  through  $D$ , Eq. (16), in Eq. (27) yields  $d = 0.7118 \lambda_{tr}$ , which is 0.20 percent in error. Thus the use of the variational boundary conditions has reduced the error by almost a factor of four. For completeness we mention the  $P_3$  Mark value for  $d$  which is  $0.6940 \lambda_{tr}$ , 2.31 percent in error. Although the Milne problem is primarily of academic interest, it is suggested that the variational boundary conditions derived here will yield better results than the Marshak conditions for practical problems involving free surfaces, such as criticality calculations, thermal-control-worth calculations, blackness-coefficient calculations, etc.

At a right-hand free surface, assumed to be at  $z = b$ , the variational boundary conditions, in the format of Eqs. (4) and (5) (with the argument  $z = a$  replaced by  $z = b$ ) are easily found from symmetry considerations to be

$$\begin{aligned} A &= + 0.532591, \\ B &= - 0.868925, \\ C &= + 0.744949, \\ D &= - 1.522003. \end{aligned} \quad (28)$$

A more detailed account of this work, including generalization to even-order and higher odd-order expansions, and treatment of entrant boundary conditions, will be published in the future.

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Received September 3, 1963

## Differences in the Removal of Activity from an Irradiated Thoria/Urania Slurry and from a Slurry Containing Adsorbed Activity

The method suggested for chemical reprocessing and purification of Th/U/O slurry type reactor fuels by Gardner<sup>1</sup> was tested with thoria/uranium slurry used in an in-pile loop experiment. Less than 1% of the gross gamma activity due to fission products was removed from the slurry by leaching with 6 M HNO<sub>3</sub> (100 C) for 6 hours. A comparison of the gamma activity in the slurry before and af-

ter treatment showed no detectable selective removal of activity. Under his conditions, Gardner reports removal of 60 to 74.3% of the activity from an unirradiated slurry containing Ce<sup>144</sup> tracer as a stand-in for fission products. Our study demonstrates that simple acid leaching can not be used to decontaminate irradiated thoria slurry for recycle.

The slurry used in our study was from an experiment of the Thorium Utilization Program of the Reactor Chemistry Division, Oak Ridge National Laboratory.<sup>2</sup> The slurry contained 1350g of Th/0.4% U<sup>235</sup> oxide per kg of D<sub>2</sub>O, to which was added a sufficient amount of palladium-treated thoria to bring the palladium concentration to 0.019 M. (The palladium was used as a catalyst for hydrogen/oxygen recombination.) The slurry had been prepumped for 900h prior to irradiation, and then irradiated to a total of  $7 \times 10^{16}$  fissions per gram of ThO<sub>2</sub>. The operating temperature of the in-pile loop was 280 C, and pressurized oxygen was kept over the slurry. Initially the slurry particles averaged 2  $\mu$ m in size and had a surface area of 2.7 m<sup>2</sup>/g. Due to the combined effect of pumping and irradiation, the particles averaged about 0.75  $\mu$ m in size and had a surface area of 32.9 m<sup>2</sup>/g when discharged from the irradiation loop. At the time of the decontamination study, the slurry had been out of the reactor about 18 months and typical fission-product analyses were  $1.5 \times 10^7$  dis min<sup>-1</sup> g<sup>-1</sup> of cesium,  $1.3 \times 10^9$  of cerium, and  $4.1 \times 10^9$  of zirconium.

The slurry used by Gardner as the basis for the originally suggested procedure was prepared by fixing Ce(NO<sub>3</sub>)<sub>3</sub> containing Ce<sup>144</sup> tracer on prepumped thoria/uranium by autoclaving at about 600 F. The difference between the results obtained with this material and the irradiated slurry may be attributed to the following: 1) The activity present in our study came from in situ fission of uranium rather than adsorbed tracer 2) Although fission recoil will result in the removal of some of the fission fragments from the particle in which the fission takes place, the stopping point of many of these fragments will be in another thoria particle<sup>3</sup> 3) Fission within the thoria particles results in the ejection of some of the thoria into

<sup>2</sup>E. L. COMPERE, H. C. SAVAGE, A. J. SHOR and E. G. BOHLMANN, "In-Pile Testing of Circulating Thoria Suspensions," (TID-7650) *Proceedings of the Thorium Fuel Cycle Symposium*, Gatlinburg, Tennessee, (Dec. 5-7, 1962).

<sup>3</sup>D. G. GARDNER, "Fission Fragment Impact Trapping by ThO<sub>2</sub> Slurry Products," *Nucl. Sci. and Eng.* 6, 487-492 (1959).

<sup>1</sup>D. G. GARDNER, "A Suggested Method for Chemical Processing and Purification of Th/U/O Slurry Type Reactor Fuels," *Nucl. Sci. and Eng.* 10, 228-234 (1961).

<sup>4</sup>M. E. A. HERMANS, R. G. SOWDEN, and H. S. G. SLOOTEN, "Irradiation Experiments with Fuel-Suspensions," *Symp. on Radiation Damage in Solids and Reactor Materials*, Paper CM-25/82, Venice (May 1962).

the D<sub>2</sub>O; ejected thoria redeposits on the surface of the particles, trapping many of the fission products that had absorbed on the surface<sup>4</sup> 4) Our irradiated slurry had an overpressure of oxygen, contained a palladium catalyst, and was in other ways different from the slurry used by Gardner. However, our slurry is more representative of that expected from an aqueous homogeneous reactor.

To date no method for leaching of fission products from irradiated thoria or urania slurry has been found which does not completely change the properties of the slurry solids or dissolve a prohibitive fraction of the oxide.

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Received April 11, 1963  
Revised October 7, 1963

### Comment on Doppler Broadened Absorption

Since the paper "Accurate Doppler Broadened Absorption"<sup>1</sup> was printed in the June 1963 issue of Nuclear Science and Engineering, Dr. Joseph J. Devaney of the Los Alamos Scientific Laboratory has written to us about his extensive earlier work<sup>2-8</sup> on this subject. Using essentially the same expression for the Doppler broadening as Eq. (7) of the above paper, Devaney and his collaborators have applied a coded digital computation to U<sup>238</sup>, Th, Pu<sup>239</sup>, Pu<sup>240</sup>, Mo, Hf, W.

We regret our oversight in not having given

<sup>1</sup>G. W. HINMAN, G. F. KUNCIR, J. B. SAMPSON, and G. B. WEST, *Nucl. Sci. Eng.* 16, 202 (1963).

<sup>2</sup>J. DEVANEY, M. GOLDSTEIN, and B. FAGAN, "Pu<sup>239</sup> Cross Sections and Their Temperature Dependence," LA-2127, Los Alamos Scientific Laboratory (1957).

<sup>3</sup>J. J. DEVANEY and B. G. FAGAN, U<sup>238</sup> Cross Sections and Their Temperature Dependence," LA-2144, Los Alamos Scientific Laboratory (1958).

<sup>4</sup>J. J. DEVANEY, M. A. DEVANEY, and D. COWARD, "Tungsten Cross Sections and Their Temperature Dependence," LA-2289 Los Alamos Scientific Laboratory (1959).

<sup>5</sup>J. J. DEVANEY, D. COWARD, and R. E. ANDERSON, "Molybdenum Cross Sections and Their Temperature Dependence," LA-2373, Los Alamos Scientific Laboratory (1960).

<sup>6</sup>J. J. DEVANEY, L. O. BORDWELL, and R. E. ANDERSON, "Thorium Cross Sections and Their Temperature Dependence," LA-2525, Los Alamos Scientific Laboratory (1961).

<sup>7</sup>J. J. DEVANEY and L. BORDWELL, "Plutonium 240 Cross Sections and Their Temperature Dependence," LA-2574, Los Alamos Scientific Laboratory (1961).

<sup>8</sup>J. J. DEVANEY, L. O. BORDWELL, and M. J. DEVANEY, "Hafnium Cross Sections and Their Temperature Dependence," LA-2763, Los Alamos Scientific Laboratory (1962).

credit to this very useful work. On the other hand, these reports have not compared the accurate treatment with the more approximate  $\psi$ -function method, and, in this sense, our work can be considered to supplement his.

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Received July 22, 1963

### Effect of Neutron Spectrum on the Branching Ratio of the Ni<sup>58</sup>(n,p) Co<sup>58</sup> Reaction

The effect of branching ratio on the thermal neutron absorption corrections for nickel activation measurements has been discussed fully by Martin and Clare<sup>1</sup>. These authors show that an error of  $\pm 10\%$  in the branching ratio can produce an error of  $\pm 5\%$  in measured fast-neutron doses using nickel activation monitors in a thermal neutron flux of  $\sim 10^{14}$  n.cm<sup>-2</sup>.sec<sup>-1</sup> and that the error is greater in higher thermal-neutron fluxes. It is, therefore, important to know the effect of neutron-spectrum variation on the branching ratio if nickel is to be used as an accurate fast-neutron dose monitor in different reactor facilities.

The mean branching ratio  $\bar{k}$  for a given neutron spectrum is defined by Martin and Clare<sup>1</sup> in terms of the mean neutron activation cross-section of the Ni<sup>58</sup>(n,p) reaction,  $\bar{\sigma}_a$ . The product  $\bar{k}\bar{\sigma}_a$  is the cross section for activation of Ni<sup>58</sup> to the ground state Co<sup>58</sup>, and  $(1 - \bar{k})\bar{\sigma}_a$  is the cross section for activation of Ni<sup>58</sup> to the isomeric state Co<sup>58</sup>. Barry<sup>2</sup> measured the activation cross-section for the Ni<sup>58</sup>(n,p) reaction as a function of neutron energy  $E$ , and obtained a value of  $\bar{\sigma}_a = (111 \pm 12)mb$  by averaging the results over a 'fission spectrum'. This result is in good agreement with the value obtained by Wright (private communication) of 107 mb relative to 65 mb for the S<sup>32</sup>(n,p) reaction by comparison of nickel and sulphur monitors in a hollow fuel element in PLUTO.

Cross<sup>3</sup> has determined the branching ratio as a

<sup>1</sup>W. H. MARTIN and D. M. CLARE, Determination of fast neutron dose by nickel activation. *Nucl. Sci. Eng.*, this issue.

<sup>2</sup>J. F. BARRY, The cross-section of the Ni<sup>58</sup>(n,p) Co<sup>58</sup> reaction for neutrons in the energy range 1.6 to 14.7 MeV. *Reactor Sci. & Technology*, Vol. 16, pp. 467-472 (1962).

<sup>3</sup>W. G. CROSS, Isomeric ratios in the reactions Ni<sup>58</sup>(n,p) Co<sup>58</sup> and Co<sup>59</sup>(n,2n) Co<sup>58</sup>. *Bull. Amer. Phys. Soc. II*, 8, No. 4 p. 368 (1963).