OAK RIDGE NATIONAL LABORATORY
MANAGED BY MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE U.S. DEPARTMENT OF ENERGY

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Dear Richard:

In reply to your request that I review the doctoral thesis of Alexander Sich, "The Chernobyl Accident Revisited: Source Term Analysis and Reconstruction of Events During the Active Phase," I found it very interesting but too voluminous to digest fully in a short period of time. I have, however, reviewed the most relevant sections (esp. Chapters VI and VII) and have investigated a few questions. First, the fission product inventory data appear reasonable with one exception: the inventory value for ^{136}Cs appears to be high by at least a factor of 50, as discussed below. Second, considering the uncertainties in much of the data, most of the fission product release values appear to be generally consistent with accepted experimental data for comparable conditions, but the release values for ruthenium and cerium seem very high. Further, the accident scenario hypothesized by Sich in Chapter VII, while requiring considerable stretch of the available data, seems plausible. It does tend to explain the peaks in release rates at day 8.5, a very important point. Perhaps additional information will be forthcoming to clarify his ideas.

1. A review of the calculated core inventories of the most important nuclides (Table VI.4) showed one obvious anomaly. The values in the **Current 1993** (presumably the most reliable) column were compared with values calculated (here at ORNL in 1986) by ORIGEN2. Although the ORIGEN2 calculation was forced to use some "best estimate" input data, because very little real data were available, the agreement with Sich values (Table VI.4, 1993) is reasonably good - generally within 30 to 50%. The Sich values were slightly higher in all except two cases. For ^{110m}Ag, the Sich value was approximately 1/2 of ORIGEN2, but we have found such poor agreement for this nuclide not unusual, and it is not a major contributor to personnel dose. On the other hand, the Sich value for the 13 d ^{136}Cs was approximately 100 times the ORIGEN2 value. Since ^{136}Cs, like ^{134}Cs, is not a direct fission product, but rather a neutron activation product, the dominant factors in its generation must be the neutron absorption cross section and the amount (yield) of ^{135}Cs, the precursor. Inquiries and discussions with several people here at ORNL who work regularly in the areas of nuclear data compilation and ORIGEN2 calculations (O. W. Hermann, R. W. Roussin, and C. W. Alexander) led to the unanimous conclusion that, since no new cross-section data are known to be available, the high Sich value for ^{136}Cs could not be correct. Furthermore, the data shown by Sich in Table VI.13 and Figures VI.5, VI.6, and VI.8 provide the same indication - ^{136}Cs release is quite low compared to the other cesium isotopes, which can be explained by use of the high inventory value. Analysis of ground and water sample data from Finland, Sweden, France, Germany, Belgium, Switzerland,

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Italy, and Greece by Dick Lorenz supports this conclusion also. In fact, Sich recognized and commented on the consistently poor agreement of ^{136}Cs compared to ^{134}Cs and ^{137}Cs (p. 376), but he offered no discussion or explanation of this problem other than to say the ^{136}Cs data are "unreliable."

2. It is difficult to evaluate the fission product release data because the sources of data are not clearly identified and there is a lack of standardization of decay dates. We have no major disagreement with the fractional releases from fuel (FCMs) for iodine, tellurium, and cesium (except for ^{136}Cs) shown in Tables VI.13 and VI.15. However, the data for ruthenium and cerium seem unreasonably high, and there are other significant questions.

   (a) We see no basis for the 95% release for ruthenium shown in Table VI.15; the 75% value in Table VI.13 is more believable, but also seems quite high. While Hunt, Cox, et al at Chalk River have documented very high ruthenium releases from small samples of UO_2 heated in air, both the much smaller surface-to-volume ratios for the Chernobyl core during the active phase and the fact that some 29% of the core had been dispersed, and, presumably, was at relatively low temperature, would indicate significantly lower release. Furthermore, the analysis of 58 fallout samples collected throughout Europe (from Scandinavia to France to Greece) by Lorenz at ORNL showed that the release of ^{103}Ru to windblown particles was ~2% of the release of the major cesium isotopes (see Fig. 1). In addition, the 95% release value is inconsistent with Sich's data. Comparison of ^{103}Ru data from Tables VI.12 and VI.15 (inventory), using ^{137}Cs as a reference, indicates a ^{103}Ru release of 24% — not the 95% shown in VI.15.

   (b) While the release of ^{129}I might be slightly lower than that for ^{131}I (because of the small fraction delayed by the 33 d ^{129}Te, which could be retained in metallic zirconium until it oxidized), I see no reason for the indicated difference — 50% vs 80%. Any difference in release for the two iodine isotopes is likely to be lost in the relatively large uncertainties in the data.

   (c) Although the 65% release of cesium seems low compared to TMI-2 results, there are indications that some cesium form is stable at relatively high temperature. In our test VI-2, a specimen of BR3 fuel was heated in steam at 2300 K for 60 min. At the end of the test, the release rate had declined to a very low level, with a total cesium release of 63% (later revised to 67%), as shown in Fig. 2. If the temperature of the Chernobyl core (that 71% on the lower biological shield) were ~2300 K or less for most of the active phase, such a cesium release fraction seems plausible. In addition, the cesium release from a large part of the 29% of core ejected from the reactor cavity could have been even lower, since temperatures for the smaller masses should have been significantly lower.

   (d) The very high release for cerium (87%) indicated in Table VI.13 seems extremely doubtful. I am not aware of any other data which would suggest a comparable cerium release. Since it is not considered in Table VI.15, I assume that Sich must have omitted it because of a lack in confidence. So far as I know, none of the data from other countries indicated high cerium release. If the 87% release value for cerium is based on measurements of samples from near the reactor, one possible explanation might be that the samples contained small fuel particles containing cerium and other fission products that settled near the reactor. Such count data might have contributed to erroneously high indications of ruthenium (above) and possibly to other low-release elements.

(e) There appears to be no data or discussion to support the extremely high initial release values at 0.5 day plotted in Fig. VI.5. If Sich had access to such data, which indicate first day release rates some seven orders of magnitude higher than any later rates, it should be clearly identified and discussed.

One additional comment with regard to future work: further iodine release data should be obtainable by using the well-known neutron activation analysis method for ^{129}I to determine what fraction of iodine remains in the core material or at other locations. Sich commented on the difficulties in directly measuring ^{129}I, but did not mention this routine method, which we have used for many years.

Please contact me if you have any questions.

Yours truly,

Morris

Morris F. Osborne

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J. T. Bell
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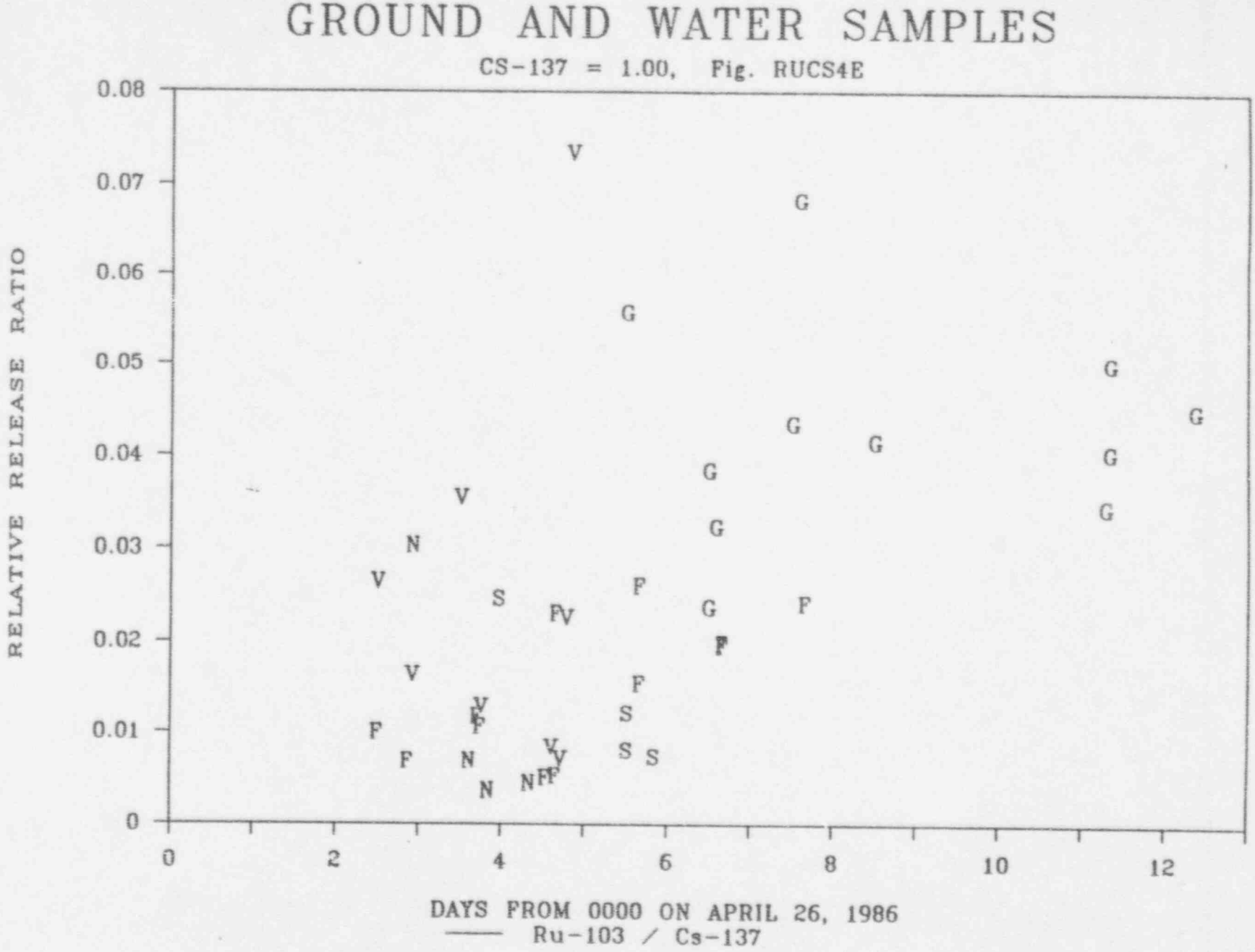

Fig. 1. Comparison of ^{103}Ru and ^{137}Cs release data from fallout samples collected throughout Europe a few days after the Chernobyl accident

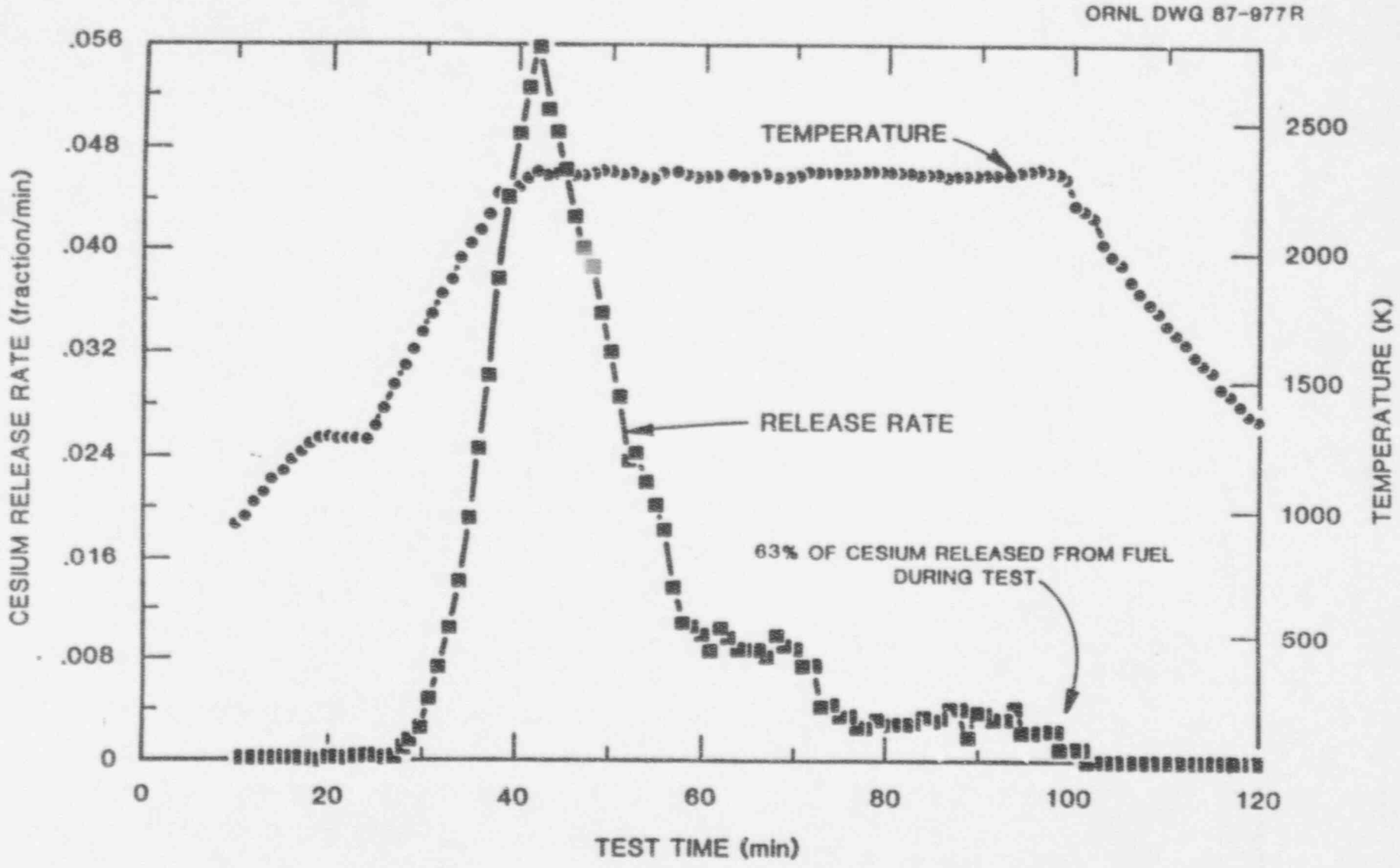

Fig. 2. **Cs RELEASE RATE IN TEST VI-2**