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Assistance Report

INTERIM REPORT

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August 3, 1979

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Dear Don:

I am writing you to deliver my long overdue progress report. I will try to be more prompt in the future, though less detailed. Much of this information was of course discussed during our June program review, however, I'm sure you would like it reiterated in writing. In addition we have made some further progress since then. As our program breaks down rather cleanly into three major tasks I will discuss them separately in that form.

I. Chemical Analysis Of Reactor Construction Materials:

Samples which we now have in hand include the following:

- a. Concrete and rebar from the Rancho Seco reactor building.
- b. Aggregate from WNP 1/4--four different coarse size fractions plus two of fines.
- c. Four test pourings of concrete from WNP 1/4.
- d. One sample of Type 2 cement from WNP 1/4--QA sample of 1200 ton batch.
- e. Four rebar samples from different heats from WNP 1/4.
- f. Four samples of structural steel from the WNP 1/4 bio-shield.

The analysis of the above samples is essentially complete. Much of the preliminary data which I showed in June has been replaced with better quality results and several additional elements have been added. A good deal more tabulation is necessary to get this data in digestible form. Rather than delay the report any further I would prefer to forward that at a later time. I am sending a data table on the concrete samples to serve as an example. The table is still somewhat incomplete. Some special techniques will be required for Li and N for example. This table is quite expanded over the ones you already have since the long decay period data is now in on INAA. I also added many elements not of direct interest to this problem. Multielement techniques give them automatically and may be of some interest from a geochemical standpoint. I have done a preliminary multicomponent mixing calculation on the concrete

component data. A reasonably good material balance is obtained on the $\pm 20\%$ level for three of the samples. The high Zn found in the concrete does in fact come from the cement for example. One of the samples - SP4158 does have large excesses of As, Cu, Sb, Mo, and Pb. While this does not represent any particularly serious problem from an activation standpoint, it does indicate the type of trace element variability which can be encountered due to sampling problems.

Another interesting observation which emerged was that all four rebar samples were relatively high in silver, ≈ 5 ppm. Both silver peaks on INAA were visible so it appears to be real, however, we would like to verify this by XRF. XRF analysis of the metal samples is at present somewhat incomplete. A dissolution technique was used which was somewhat unsatisfactory. We would prefer to run the samples in disk form. The disks have been machined and we are awaiting receipt of standards from NBS.

In addition to the above mentioned samples we also have received the following samples which are currently being analyzed.

- g. Three samples of Type 304 stainless steel from a major supplier of reactor internals.
- h. Three samples of Type 316 stainless from the same source.
- i. One sample of SA503 BWR pressure vessel steel from a utility; supplied by BCL.

II. Activation Calculations:

During the past month I have been devoting some extra effort to preparation for activation calculations. I have written a program of my own (ACTIV) to calculate neutron capture and fission product isotopes. The program includes multiple capture and burnup reaction. Also included are multiple capture fission products from ^{238}U and ^{232}Th . A nuclear data library has been compiled simply using the GE chart information. Resonance integrals are included when known or a $1/V$ cross-section assumed if not. Additional input includes thermal flux, resonance flux and temperature. The resonance flux is calculated assuming a $1/E$ spectral shape. This program is now fully debugged and runs interactively on the department PDP-11/35 computer. Some additional review of the cross-section data is necessary before doing any lengthy calculation work. I have contacted some cross-section experts at the National Nuclear Data Center at Brookhaven and they promised assistance. I plan a two day visit to BNL in mid-August in connection with another trip to pursue this further. I feel that ACTIV will be a useful working tool which I can use easily (and for free) in my own laboratory, however, it involves some approximations which I would like to check. The PNL Energy System group has been routinely using the ORNL program ORIGEN which is presumably more mathematically sophisticated. This program has among other things been used for the

activation calculations in the PWR Decommissioning Study. The program is currently being maintained on the DOE UNIVAC 1100 system. I have set up an account and obtained all of the necessary information to use this program. This will be particularly useful in preparing calculations for my upcoming talk at the ANS meeting at Sun Valley. There appears to be a number of problems with ORIGEN, however, which prompted me to develop my own program. The program is very large and must run on a large computer. This computer is very inconvenient to use and rather expensive. In addition, the data being used appears to be badly out of date. In particular many important resonance cross-sections and burnup cross-sections shown on the GE chart are missing. I have been told by several people that a new ORIGEN library is available, but have so far failed to connect with the appropriate person at ORNL. This problem should be solvable in the near future. If necessary we can always modify the existing library where necessary.

Neutron flux information will be taken from ANISN calculations. These calculations have already been carried out for the PWR and BWR decommissioning reports. Dick Smith has provided me with a copy of their computer output. This information can be used directly without any further work. For reactor internals and pressure vessel fluxes I also have available in addition a report provided by Frank Rahn of EPRI on very detailed two-dimensional neutron transport calculations for both a PWR and a BWR. That part of the problem is well in hand.

III. Radiochemical Analysis Of Activated Samples Of Reactor Materials:

At this point in the program the main effort in radiochemistry has been in planning a usable procedure. Since irradiated steel samples seem to be the most readily obtainable at this time, the procedure is being designed around steel. A multielement separation procedure is being worked out for Co^{60} , Ni^{59} , Ni^{63} , Zn^{65} , $\text{Ag}^{110\text{m}}$, $\text{Ag}^{108\text{m}}$, ^{90}Sr , ^{137}Cs , Sm^{151} , Eu^{152} , Eu^{154} , and $\text{Ho}^{166\text{m}}$. Tracers have been obtained and used to test the procedure for those elements. Group separations are also being tested for ^{93}Zr , $^{178\text{m}}\text{Hf}$ and Mn^{54} , Mo^{93} , Nb^{94} . Fractions will also be reserved for ^3T , ^{14}C , and ^{239}Pu . Due to the relatively high radioactivity levels expected with these samples initial sampling and dissolution will have to be carried out outside our building. Space has been reserved in a nearby building for this work. A specially shielded glove box will be used for sampling, dissolution and removal of ^{60}Co . The samples can then be handled in the 329 Building for the remaining chemistry. The first full scale radiochemical separation will be carried out on a Type 304 stainless steel sample from the Point Beach reactor which was available locally. The sample was cut from a portion of a fuel bundle. It has been out of the reactor for about eight years making it an ideal sample for our purposes. Although this sample is actually part of the fuel cycle it should be representative of other related internals such as the

shroud which should be very important during the early phase of dismantling. If the radiochemical procedure now being tested prove satisfactory, we expect to begin work on this sample some time in October.

Future Sample Opportunities:

Opportunities to obtain samples in the future look excellent, however, protocol problems tend to slow things a bit. At least two samples of irradiated vessel steel will be available when needed from Battelle Columbus Laboratory. A large variety of samples should be available through the surveillance capsule program at Westinghouse Pittsburg. Westinghouse has in hand a large backlog of irradiated vessel steel with matching un-irradiated samples. These span a good range of irradiation conditions. They are quite willing to let us have them if the utilities agree. They also have some irradiated 304 stainless from a coupon hanger which may be useful. Westinghouse has also agreed to help us obtain concrete and rebar samples from reactor of theirs under construction. This should provide good geographical diversity if it works out. The appropriate protocol will of course be necessary individually in each case.

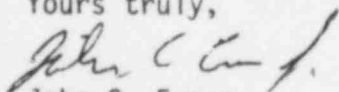
Conclusions:

On the whole I am reasonably satisfied with the progress of the program. A good start has been made on all of the required areas. The analytical data is almost complete on the first batch of samples providing at the same time a good opportunity to evaluate potential analytical problems. They appear to be minimal. At this point we are now ready to begin a major effort to obtain more samples. Samples for radiochemical analysis do appear to be readily obtainable with the exception of activated bioshield concrete. This remains a major problem. I am planning some site visitations in connection with Dave Robertson's program in an attempt to solve this problem.

As this is the first letter report on the program I have tried to be as detailed as possible. On any new program there must always be a lot of loose ends to tie down at first. If you have any further questions please let me know. I will try to be more concise and more prompt with future reports. I will also forward any remaining data as soon as I have it organized better.

957313

Yours truly,



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Senior Research Scientist
Earth and Planetary Chemistry Section
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C. W. Wilkerson

JCE/cms

July 20, 1979

CONCRETE SAMPLES
(ppm unless otherwise noted)

| | H | Li | B | N | % Na | % Mg | % Al | % Si | % S | Cl | % K | % Ca | Sc | % Ti | V | Cr | Mn | % Fe | Co | Ni |
|---------------|---|----|---|---|------|------|------|------|------|------|-----|------|------|------|-----|-----|------|------|------|-----|
| Rancho Seco A | | | | | ---- | ---- | 5.9 | 23.9 | .44 | <800 | .9 | 12.3 | ---- | .36 | 173 | 240 | 699 | 3.8 | ---- | 70 |
| B | | | | | 1.28 | | 5.7 | ---- | ---- | ---- | .74 | 11.3 | 16.2 | .41 | 150 | 209 | 710 | 3.7 | 15.2 | 100 |
| C | | | | | | | | | | | | | | | | | | | | |
| --WPPSS-- | | | | | | | | | | | | | | | | | | | | |
| SP4166E A | | | | | ---- | ---- | 5.3 | 21.0 | .51 | <800 | 1.1 | 13.7 | ---- | .74 | 190 | 42 | 1038 | 5.4 | ---- | 19 |
| B | | | | | 1.64 | | 5.4 | ---- | ---- | ---- | 1.1 | 12.5 | 17.9 | .99 | 190 | 35 | 990 | 5.2 | 25.8 | 40 |
| C | | | | | | | | | | | | | | | | | | | | |
| SP4158 A | | | | | ---- | ---- | 5.7 | 21.1 | .49 | <700 | .9 | 14.7 | ---- | .77 | 170 | 54 | 900 | 5.3 | ---- | 26 |
| B | | | | | 1.53 | | 5.7 | ---- | ---- | ---- | .91 | 12.6 | 17.5 | .69 | 170 | 42 | 850 | 5.0 | 22.3 | <30 |
| C | | | | | | | | | | | | | | | | | | | | |
| SP4715 A | | | | | ---- | ---- | 6.9 | 20.1 | .54 | <700 | .90 | 16.3 | ---- | .73 | 230 | 43 | 1195 | 5.5 | ---- | 23 |
| B | | | | | ---- | 1.40 | 5.2 | ---- | ---- | ---- | .93 | 14.5 | 16.6 | .78 | 180 | 39 | 1100 | 4.84 | 20.7 | 40 |
| C | | | | | | | | | | | | | | | | | | | | |
| SP X A | | | | | ---- | ---- | 5.4 | 23.9 | .28 | <700 | 1.2 | 11.1 | ---- | .72 | 240 | 32 | 1021 | 5.2 | ---- | 22 |
| B | | | | | 1.68 | | 5.8 | | | | .94 | 10.3 | 17.5 | .71 | 180 | 32 | 1010 | 4.9 | 22.1 | 40 |
| C | | | | | | | | | | | | | | | | | | | | |

*A - XRF
B - INAA
C - Other

957314

POOR ORIGINAL

July 20, 1979

CONCRETE SAMPLES
(ppm unless otherwise noted)

| | | Cu | Zn | Ga | As | Se | Br | Rb | Sr | Y | Zr | Nb | Mo | Pd | Ag | Cd | Sn | Sb | Cs | Ba | La | Ce | Nd | Sm | Eu |
|----------------------|---|----|-----|----|-----|-----|-----|----|-----|----|-----|-----|-----|----|----|----|-----|------|-----|-----|------|----|----|-----|------|
| Rancho Seco | A | 46 | 68 | 12 | 5.8 | <1 | <1 | 29 | 422 | 16 | 92 | 5.4 | 2.5 | <3 | <4 | <7 | <3 | <3 | --- | 608 | 15 | 24 | 22 | --- | ---- |
| | B | -- | 48 | -- | 4.1 | <.9 | <.7 | 28 | 390 | -- | 100 | --- | --- | -- | <1 | -- | --- | 1.0 | 1.1 | 660 | 15 | 28 | 14 | 2.3 | .95 |
| | C | | | | | | | | | | | | | | | | | | | | | | | | |
| --WPPSS-- SP4166E | A | 37 | 394 | 15 | 4.7 | <1 | <1 | 36 | 382 | 22 | 140 | 10 | 3.9 | <3 | <4 | <7 | <4 | <3 | --- | 466 | 16 | 40 | 30 | --- | ---- |
| | B | -- | 420 | -- | 3.5 | 1 | <.7 | 34 | 390 | -- | 180 | --- | --- | -- | <2 | -- | --- | .2 | 1.4 | 540 | 20 | 41 | 20 | 4.2 | 1.31 |
| | C | | | | | | | | | | | | | | | | | | | | | | | | |
| SP4158 | A | 72 | 260 | 13 | 104 | <1 | <1 | 30 | 364 | 22 | 142 | 8 | 19 | <3 | <4 | <7 | 10 | ---- | --- | 415 | 13 | 30 | 24 | --- | ---- |
| | B | -- | 270 | -- | 104 | 2 | <.7 | 34 | 370 | -- | 130 | --- | 23 | -- | <1 | -- | --- | 48.7 | 0.8 | 500 | 17.5 | 36 | 18 | 3.8 | 1.19 |
| | C | | | | | | | | | | | | | | | | | | | | | | | | |
| SP4715 | A | 37 | 421 | 12 | 7 | 1.0 | <.8 | 36 | 355 | 23 | 140 | 9 | 5.5 | <3 | <4 | <7 | 10 | <3 | --- | 415 | 15 | 34 | 29 | --- | ---- |
| | B | -- | 430 | -- | 5.8 | 2 | 1.2 | 36 | 360 | -- | 160 | --- | --- | -- | <1 | -- | --- | 3.3 | 0.9 | 480 | 26.5 | 50 | 22 | 4.4 | 1.19 |
| | C | | | | | | | | | | | | | | | | | | | | | | | | |
| SP X | A | 31 | 289 | 16 | 4 | <1 | <.8 | 38 | 366 | 23 | 153 | 10 | 3.7 | <3 | <4 | <7 | 3.1 | <3 | --- | 492 | 20 | 36 | 31 | --- | ---- |
| | B | -- | 290 | | 1.7 | 2 | <.8 | 33 | 400 | | 130 | --- | --- | -- | <1 | -- | --- | .7 | 1.1 | 540 | 19.6 | 40 | 18 | 4.2 | 1.9 |
| | C | | | | | | | | | | | | | | | | | | | | | | | | |

*A - XRF
B - INAA
C - Other

957315

POOR ORIGINAL

July 20, 1979

CONCRETE SAMPLES
(ppm unless otherwise noted)

| | | Tb | Dy | ¹⁴⁰ Sm | Yb | Lu | Hf | Ta | W | Pb | Bi | Th | U |
|-------------|---|-----|-----|-------------------|-----|-----|-----|-----|-----|----|-----|-----|-----|
| Rancho Seco | A | --- | --- | --- | --- | --- | --- | --- | --- | 8 | --- | --- | --- |
| | B | .48 | 2.9 | <.4 | 1.6 | .31 | 2.9 | .46 | <1 | -- | --- | 3.5 | 1.8 |
| | C | | | | | | | | | | | | |
| --WPPSS-- | | | | | | | | | | | | | |
| SP4166E | A | --- | --- | --- | --- | --- | --- | --- | --- | 8 | --- | --- | --- |
| | B | .71 | 4.7 | <.9 | 2.5 | .40 | 3.8 | .78 | 1 | -- | --- | 4.7 | 1.4 |
| | C | | | | | | | | | | | | |
| SP4158 | A | --- | --- | --- | --- | --- | --- | --- | --- | 72 | --- | --- | --- |
| | B | .65 | 4.0 | 1.2 | 2.2 | .34 | 3.6 | .64 | <1 | -- | --- | 3.6 | 2.1 |
| | C | | | | | | | | | | | | |
| SP4715 | A | --- | --- | --- | --- | --- | --- | --- | --- | 9 | --- | --- | --- |
| | B | .72 | 4.2 | .9 | 2.4 | .34 | 3.9 | .72 | <1 | -- | --- | 4.9 | 1.7 |
| | C | | | | | | | | | | | | |
| SP X | A | --- | --- | --- | --- | --- | --- | --- | --- | 5 | --- | --- | --- |
| | B | .70 | 4.1 | .9 | 2.4 | .37 | 3.9 | .79 | 2 | -- | --- | 4.5 | 1.7 |
| | C | | | | | | | | | | | | |

*A - XRF
B - INAA
C - Other

957016

POOR ORIGINAL