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LEACHING OF ^{14}C AND ^{36}Cl
FROM IRRADIATED FRENCH GRAPHITE

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SUMMARY

The leach rates of ^{14}C and ^{36}Cl were measured on solid cylindrical samples prepared from irradiated graphite blocks supplied by the French Commissariat a l'Energie Atomique (CEA). Static leach tests were conducted in deionized water at 20°C for 13 weeks. The graphite samples were completely submerged in the water, and the entire volume of water was changed and analyzed at weekly intervals for the first three weeks and biweekly thereafter.

Large differences in the leach rates of both ^{14}C and ^{36}Cl were observed between samples machined from the different blocks. In general, the leach rates were much higher than those measured in an earlier study with graphite obtained from a block removed from one of the Hanford reactors.

The data from this study are compared with those from the previous study using the Hanford-reactor graphite. Implications of the data from both studies regarding possible rate-limiting mechanisms are discussed.

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INTRODUCTION

The United States Department of Energy (DOE) and the French Commissariat a l'Energie Atomique (CEA) both have active programs aimed at achieving the safe, effective, and economic disposal of surplus graphite-moderated nuclear reactors. To attain this goal, both programs require information on the leaching of radioisotopes from irradiated graphite.

Slight differences in procedures used in the leaching studies at the two sites have hampered direct comparison of data. To determine if the different leaching procedures yield equivalent data, differences in the graphite, or possible differences in the chemical form or distribution of radioactive isotopes within the graphite, must be eliminated. Toward this end, the DOE and the CEA entered into a technical exchange agreement with the immediate objectives of exchanging test procedures, a small number of graphite samples, and data obtained on those samples. Subsequently, Pacific Northwest Laboratory (PNL)^(a) was commissioned by the DOE to conduct the leach tests and other technical aspects of the exchange.

The purpose of this report is to document the results of the leach tests conducted on the samples of graphite obtained from the French. The information exchange will help to illuminate areas where 1) data obtained under one program (DOE or CEA) can complement information derived from the other program, or 2) basic understanding of processes influencing leaching behavior needs further clarification.

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EXPERIMENTAL

LEACH SPECIMENS

Three cylindrical blocks (about 80 mm dia by 75 mm long) of irradiated graphite from the French G-2 Reactor were received in August 1988. The blocks were not marked for identification, but were in individually marked plastic bags. The two cylinders having the highest gross-gamma activities had a one-quarter segment of a fuel channel cut from near the end of the cylinder; these cylinders appear to be segments of cores, extending from the edge of a moderator block to the center of a fuel channel. Our specimens (about 30 mm dia by 30 mm long) were machined from near these fuel channel holes. One end of the third cylinder appeared to be an original surface of a moderator block; our specimen from this cylinder was cut from near the interior (highest activity) end. Figure 1 depicts the location of the leach specimens machined from the original cylindrical blocks.

The three specimens were separately characterized^(a) for density, concentrations of ^{14}C , ^{36}Cl , and gamma-ray emitters, and surface area using the method of Brunauer, Emmett, and Teller (BET). Table 1 summarizes the characterization results for the three specimens.

The ^{14}C and ^{36}Cl concentrations were measured using the aluminum absorption method^(b); corrections for self-absorption and sample geometry were obtained from special standards fabricated to the same size and density as the graphite specimens. For these standards, ^{204}Tl was substituted for ^{36}Cl , because it has nearly the same beta-ray energy (0.763 versus 0.714 MeV) and is easier to handle during the fabrication process.

TEST PROCEDURES

The leach test procedure was based on ANSI/ANS-16.1-1986 "Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term

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- (a) Radiometric measurements were provided by C. W. Thomas, and BET measurements were made by M. C. Burt, both of PNL.
 - (b) ASTM Method E186-76, "Analysis of Radioisotopes," Annual Book of ASTM Standards, Part 45 (1981).

FIGURE 1. Schematic Diagram Showing Location of Leach Specimens Machined from Cylindrical Blocks.

TABLE 1. Characterization Results

	Markings on Plastic Bags		
	NORD SUB	SUP NORD	B A
Gross- γ Activity, (a) μGy	300	100	80
Density, Mg/m^3	1.633	1.658	1.655
BET Area, m^2/g	0.29	0.25	0.29
Mass, g	36.20	37.01	37.27
<u>Activities, MBq/kg</u>			
^{14}C	53.9	65.3	16.3
^{36}Cl	4.46	4.48	0.94
^{60}Co	11.1	11.4	2.12
^{133}Ba	0.093	0.179	0.100
^{134}Cs	0.034	0.025	0.015
^{137}Cs	0.097	0.075	0.011
^{154}Eu	0.525	0.758	0.503
^{155}Eu	0.277	0.374	0.211

(a) Measured on 80-mm-dia x 75-mm-long cylinders, as received. Other measurements were made on 30-mm-dia x 30-mm-long samples cut from the cylinders.

Test Procedure," modified as noted below. The major modification was restructuring of the leachant-renewal schedule to eliminate unnecessary and expensive radionuclide analyses. The same leach-test procedures and equipment were used in tests conducted earlier on graphite specimens removed from one of the Hanford reactors (Gray and Morgan 1988).

Leachant

The leachant was deionized water that was sparged with clean air for 10 to 15 minutes prior to use to ensure air saturation.

Leach Containers

The leach containers consisted of pint-size canning jars with specially designed lids to provide a better seal and thereby prevent the escape of CO_2 or other gases during leaching. Specimen holders made of fused silica were used to hold the graphite specimens near the center of the leach containers.

Ratio of Leachant Volume to Specimen Surface Area (V/S)

Leachant volumes were 406 ± 3 mL in all the tests, and the geometric surface area of each graphite specimen was 43.8 cm^2 ; thus $V/S = 9.27 \text{ cm}$. Although this is slightly different from the ratio of 10.0 cm recommended by ANSI/ANS-16.1-1986, it is considered to be an insignificant difference because V/S ratios are important in leach tests only if the leached species approach their solubility limits in the leachate, which is not the case in these tests for either ^{14}C or ^{36}Cl .

Temperature

A water bath was used to maintain the temperature at $20 \pm 0.5^\circ\text{C}$ during these tests.

Leach Periods

The leachates were changed at intervals of 1, 2, 3, 5, 7, 9, 11, and 13 weeks. One of the important features of the ANSI/ANS-16.1-1986 procedure is that the entire quantity of leachate is removed from the leach container at the end of each leach period and replaced with fresh leachant. Part of the reason for this is to avoid saturating the solution with any of the leached constituents. As extra insurance against saturation, the ANSI/ANS-16.1-1986 procedure calls for frequent renewal of leachant during the early part of the leaching schedule. Since the possibility of saturating the leachate with ^{14}C and ^{36}Cl from the graphite was expected to be remote and short leach periods might have reduced ^{14}C and ^{36}Cl concentrations below detection limits in the current testing program, the leachant renewal frequency during the early part of the schedule was reduced.

At the end of each leach period, the leach containers were removed from the water bath, dried, and weighed to assure that no water had leaked into or out of them. The leachates were removed from the containers and quickly replaced with fresh leachant to prevent the graphite specimens from drying out.

Following removal of the leachates from the leach containers, 3 to 4 mL of 10 M NaOH were added to each to assure that the ^{14}C was maintained in the carbonate form. The NaOH-treated leachates were passed through $0.45\text{-}\mu\text{m}$ filters to remove any graphite particles and then placed in tightly capped sample bottles for transfer to the radioanalytical laboratory.

The radioanalytical laboratory analyzed duplicate 200-mL aliquots of each leachate to determine the concentrations of ^{14}C and ^{38}Cl . The aliquots were placed individually in a distillation flask where known amounts of carbonate and activated chloride (^{38}Cl) carrier were added. Nitric acid was added, and the flask was simultaneously heated and purged with water-scrubbed nitrogen. Gas leaving the distillation flask was passed through a condensation flask and then through a series of $\text{Ba}(\text{OH})_2$ traps where the CO_2 was converted to BaCO_3 . The BaCO_3 precipitate was removed from the $\text{Ba}(\text{OH})_2$ traps by centrifuging and transferred to a counting dish, dried, weighed, and counted for ^{14}C . Weighing the carbonate before and after distillation provided a measure of the chemical yield. A correction curve for self-absorption of the ^{14}C β -radiation was generated as a function of the amount of BaCO_3 precipitate in the counting dish.

Following the carbonate distillation, the solution remaining in the distillation flask was transferred to a beaker, diluted to 400 mL, and AgNO_3 was added. The resulting AgCl precipitate was filtered, and the amounts of ^{38}Cl and ^{36}Cl determined by gamma- and beta-counting, respectively. The radiochemical yield was determined by comparing the amount of ^{38}Cl following the chemical separation with the amount added to the distillation flask. As in the case of ^{14}C , a correction curve for self-absorption of the ^{38}Cl β -radiation was generated as a function of the amount of AgCl on the filter paper.

RESULTS

The concentrations of ^{14}C and ^{36}Cl found in the leachates, as well as other experimental parameters, are given in Appendix A. Data presented in Tables 2 and 3 were derived from the raw data listed in Appendix A according to the following equations:

$$F = \frac{CV}{IM} \quad (1)$$

$$R = \frac{F}{t} \quad (2)$$

$$L = \frac{R}{A} \quad (3)$$

$$W = \frac{CV}{aMA t} \quad (4)$$

where:

F = total fraction of inventory of radionuclide released in a given leaching interval

C = concentration of radionuclide in the leachate, disintegrations per minute per milliliter, d/m•mL

V = volume of leachate, mL

I = inventory of radionuclide in the graphite specimen, disintegrations per minute per gram of specimen, d/m•g

M = mass of specimen, g

R = average fractional leach rate for the period, day⁻¹

t = time, days

L = average leach rate of graphite per unit surface area of specimen, g/m²•day. This is the leach rate that would occur if the entire graphite specimen leached congruently. (See the following discussion for further caveats.)

A = BET surface area of specimen, m²/g

W = average mass loss rate of radionuclide per unit surface area of specimen, g/m²•day. This value is the mass loss rate of the given radionuclide alone.

a = specific activity of radionuclide; 1.65×10^{11} Bq/g for pure ^{14}C and 1.22×10^9 Bq/g for pure ^{36}Cl .

Figures 2 through 8 display the data listed in Tables 2 and 3. Similar data obtained in earlier tests with graphite specimens from one of the Hanford reactors (Gray and Morgan 1988) are also shown in some of the figures for comparison.

The average leach rate, L , defined by Equation (3) requires further clarification. As stated, it is the leach rate that would occur if the specimen leached congruently, which means that the concentration of all elements and isotopes in the leachate are in the same proportion as in the solid. It is not generally true that solids leach congruently, and it may or may not be true of the graphite in this study. In fact, arguments are presented in the following section to suggest that irradiated graphite does not leach congruently, at least during the early stages of leaching. The value, L , is simply a hypothetical concept that is often used to express leaching data.

FIGURE 2. Carbon-14 Fractional Leach Rate in Deionized Water at 20°C

FIGURE 3. Chlorine-36 Fractional Leach Rate in Deionized Water at 20°C

FIGURE 4. Carbon-14 Mass Loss Rate in Deionized Water at 20°C

FIGURE 5. Chlorine-36 Mass Loss Rate in Deionized Water at 20°C

FIGURE 6. Cumulative Fraction of Carbon-14 Inventory Leached in Deionized Water at 20°C

FIGURE 7. Cumulative Fraction of Chlorine-36 Inventory Leached in Deionized Water at 20°C

FIGURE 8. Cumulative Fraction of Carbon-14 Inventory Leached in Deionized Water at 20°C

TABLE 2. Leach Rates of the Three Graphite Specimens

Time, Days	¹⁴ C Leach Rates								
	NORD SUB			SUP NORD			B A		
	R ^(b)	L ^(b)	W ^(b)	R	L	W	R	L	W
3.5	4.37E-5	1.51E-4	4.91E-11	8.44E-5	3.37E-4	1.34E-12	1.75E-4	6.03E-4	5.96E-11
10.5	5.85E-5	2.02E-4	6.58E-11	6.76E-5	2.70E-4	1.07E-12	1.81E-4	6.25E-4	6.17E-11
17.5	6.35E-5	2.19E-4	7.14E-11	6.04E-5	2.41E-4	9.55E-11	1.43E-4	4.93E-4	4.87E-11
28	3.79E-5	1.31E-4	4.26E-11	3.72E-5	1.49E-4	5.88E-11	9.86E-5	3.40E-4	3.36E-11
42	3.20E-5	1.10E-4	3.60E-11	3.60E-5	1.44E-4	5.69E-11	8.42E-5	2.91E-4	2.87E-11
56	1.35E-5	4.65E-5	1.51E-11	2.86E-5	1.14E-4	4.52E-11	5.83E-5	2.01E-4	1.99E-11
70	9.50E-6	3.28E-5	1.07E-11	1.39E-5	5.54E-5	2.19E-11	5.03E-5	1.73E-4	1.71E-11
84	1.17E-5	4.04E-5	1.32E-11	1.19E-5	4.76E-5	1.88E-11	6.73E-5	2.32E-4	2.29E-11
³⁸ Cl Leach Rates									
3.5	4.67E-2	1.61E-1	5.71E-7	1.01E-3	4.03E-3	1.48E-8	4.10E-3	1.41E-2	1.08E-8
10.5	7.40E-3	2.55E-2	9.03E-8	2.03E-4	8.11E-4	2.97E-9	7.97E-4	2.73E-3	2.11E-9
17.5	9.43E-4	3.25E-3	1.15E-8	5.71E-5	2.28E-4	8.36E-10	1.95E-4	6.72E-4	5.15E-10
28	1.85E-4	6.38E-4	2.26E-9	1.68E-5	6.74E-5	2.47E-10	3.85E-5	1.33E-4	1.02E-10
42	9.80E-5	3.38E-4	1.20E-9	1.18E-5	4.73E-5	1.73E-10	1.55E-5	5.34E-5	4.09E-11
56	6.84E-5	2.36E-4	8.36E-10	8.45E-6	3.38E-5	1.24E-10	1.27E-5	4.39E-5	3.36E-11
70	5.73E-5	1.98E-4	7.00E-10	7.50E-6	3.00E-5	1.10E-10	1.21E-5	4.16E-5	3.18E-11
84	3.85E-5	1.33E-4	4.70E-10	5.97E-6	2.39E-5	8.76E-11	9.29E-6	3.20E-5	2.45E-11

(a) The times listed are the midpoints of the different leaching intervals.

(b) See page 9 for definition of these values.

TABLE 3. Cumulative Fraction of Radionuclides Leached
from Three Graphite Specimens

Time, Days	¹⁴ C			³⁶ Cl		
	<u>NORD SUB</u>	<u>SUP NORD</u>	<u>B A</u>	<u>NORD SUB</u>	<u>SUP NORD</u>	<u>B A</u>
7	3.06E-4	5.90E-4	1.22E-3	3.27E-1	7.06E-3	2.87E-2
14	7.16E-4	1.06E-3	2.49E-3	3.79E-1	8.48E-3	3.43E-2
21	1.16E-3	1.49E-3	3.49E-3	3.85E-1	8.88E-3	3.56E-2
35	1.69E-3	2.01E-3	4.87E-3	3.88E-1	9.12E-3	3.62E-2
49	2.14E-3	2.51E-3	6.05E-3	3.89E-1	9.29E-3	3.64E-2
63	2.33E-3	2.91E-3	6.87E-3	3.90E-1	9.40E-3	3.66E-2
77	2.46E-3	3.11E-3	7.57E-3	3.91E-1	9.51E-3	3.68E-2
91	2.63E-3	3.27E-3	8.51E-3	3.91E-1	9.59E-3	3.69E-2

DISCUSSION

One of the striking features of the data shown in Figures 2 through 8 is the large differences between the three French graphite samples, particularly in the ^{36}Cl results. The apparent differences are reduced when the ^{14}C data are plotted on the basis of mass loss per unit surface area (Figure 4), which is independent of the specific ^{14}C inventory, but no similar reduction in differences occurs with the ^{36}Cl data when they are plotted on that basis (Figure 5). It is also interesting that the greatest leach rates for ^{14}C and ^{36}Cl do not occur in the same sample. We are unaware of any differences between the samples that might be expected to lead to the observed differences in leaching behavior.

Another feature of the data shown is the much greater leach rates and cumulative leach fraction of the French graphite compared to the Hanford graphite. The difference is even more striking when the data, particularly those for ^{36}Cl , are plotted on the basis of specific surface areas (Figures 4 and 5). This appears to contradict our belief that graphite leach rates should be proportional to specific surfaces areas given no other differences between samples.

Note (Figure 7) that one of the French samples lost $38 \pm 8\%$ of its ^{36}Cl inventory. To corroborate this, the inventory of this particular sample was remeasured after leaching, and a value $29 \pm 12\%$ less than the original inventory was obtained. The agreement between the two methods of measuring ^{36}Cl loss seems reasonable since the less precise inventory measurements involved subtracting one somewhat uncertain value from another.

In our earlier report (Gray and Morgan, 1988), we discussed some of the features of the leaching data and speculated extensively regarding the possible underlying mechanisms. It is interesting to see how the data obtained with the French graphite support or challenge those arguments.

The ^{14}C data obtained with the Hanford graphite at 20°C was rather scattered whereas the 50°C and 90°C data decreased monotonically with time. We speculated that the scatter may have been due to suspected temperature variations because the leach containers were allowed to stand on the laboratory bench where the temperature was neither controller nor monitored.

Interestingly, the ^{14}C data obtained with the French graphite were also somewhat scattered despite the fact that the temperature was carefully controlled at $20 \pm 0.5^\circ\text{C}$ throughout the entire 13 weeks. Perhaps the scatter in the two sets of data is related to the same, as yet unidentified, cause.

We suggested two possible mechanisms for the observed decrease in leach rates with time: 1) depletion of the ^{14}C and ^{36}Cl sources, and 2) diffusion limited rate of transport of either a reactant such as O_2 , or a reaction product, through the static leachant contained within the tortuous pores of the graphite. The sources were not being depleted, per se, from the Hanford graphite, nor was the ^{14}C depleted from the French graphite. However, significant fractions of ^{36}Cl were depleted from two of the French graphite samples, particularly sample "NORD SUB." Nevertheless, for reasons we described earlier (Gray and Morgan 1988) it seems more likely that just a portion of both sources may have become depleted because they are more reactive or more readily accessible to the leachant. Once depletion of these reactive and/or accessible sources of have occurred, the leach rates of both should approach the leach rate of ^{12}C . As was the case with the Hanford graphite, the leach rates of ^{14}C and ^{36}Cl from the French graphite approach similar values, which lends support to this premise. However, since the leach rates for the French graphite appear to be approaching steady-state values about an order of magnitude higher than for the Hanford graphite, this implies that the French graphite, in general, is more reactive.

The possibility that the rate-limiting mechanism is associated with diffusion of a reactant or product does not appear to be supported by the data for either the French or the Hanford graphite because the cumulative releases are generally not proportional to the square root of time. This is particularly true of the ^{36}Cl data in both studies. Figure 8 shows that the ^{14}C data for French sample "B A" are roughly linear on a square-root-of-time plot, but the other samples do not exhibit linear behavior.

We argued that limited oxygen availability (enough to oxidize about 6 mg of carbon) in the leach containers could not be a limiting factor in the leach tests with the Hanford graphite.^(a) In contrast, more than 6 mg of

(a) Gray (1982) has shown that the mechanism for "leaching" of carbon from graphite is the water-catalyzed oxidation by dissolved oxygen to form CO_2 .

carbon would have been oxidized (leached) from each of the three French samples during most of the leach periods if it is assumed that ^{12}C oxidizes (and therefore leaches) at the same rate as ^{14}C . The extreme case occurred with French sample "B A", from which 0.138% of the inventory of ^{14}C was leached during the 4th period. Again, assuming that ^{12}C oxidizes at the same rate as ^{14}C , a total of 51 mg of carbon would have leached from the 37-g sample. Since only enough oxygen was available to oxidize about 6 mg of carbon during each leach period, the necessary conclusion is that ^{12}C must have oxidized (leached) at a lower rate than the ^{14}C in these tests. This conclusion is also supported by the data of Arnold and Libby (1946), who reported that the initial oxidation rate of ^{14}C is many times that of ^{12}C for irradiated graphite.

Finally, it is of interest to compare the ^{14}C leaching data from both of our studies with those obtained by White et al. (1984). The steady-state leach rates obtained by White et al. at 20°C to 25°C were in good agreement with those we obtained on Hanford graphite at 20°C , but those rates were far less than we found with the French graphite. White et al. reported total fractions of inventories leached after 100 days to be in the range 8.8×10^{-4} to 1.4×10^{-3} . This agrees more favorably with our results for the French graphite (2.6×10^{-3} to 8.5×10^{-3} of inventory) than for the Hanford graphite (3.6×10^{-5} to 9.2×10^{-5} of inventory). White et al. found leach rates that decreased by factors of 50 to 100 after 100 days. This is a far greater decrease than we observed for either the Hanford or the French graphite. Reasons for these similarities and differences can only be speculated upon as we did in our earlier report (Gray and Morgan 1988).

CONCLUSIONS

Data obtained on the French graphite support the idea that the leach rate of ^{14}C from irradiated graphite initially exceeds the leach rate of ^{12}C , in agreement with results published by White et al. (1984). However, since the leach rates of ^{14}C and ^{36}Cl tended toward common values in our tests with both French and Hanford graphite, this supports the premise that both are controlled by some common factor such as the leach rate of ^{12}C following depletion of active or readily accessible portions of the ^{14}C and ^{36}Cl .

Understanding the rather large differences observed between the different French samples requires more information than we currently have regarding their source and their treatment following manufacture. Such information might also help to explain the large differences in leaching behavior between the French and Hanford graphite samples.

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APPENDIX A

SUMMARY OF RAW DATA

TABLE A.1. Leach Data for Graphite Sample NORD SUB

Start		End		Elapsed Time, Days	Leachant Vol. (mL)	¹⁴ C Conc. (d/m•mL)	³⁶ Cl Conc. (d/m•L)
Date	Time	Date	Time				
10/16/88	1340	11/2/88	1045	7	409	88.0 ± 13	7580 ± 810
11/2/88	1050	11/9/88	1500	7	407	118 ± 18	1200 ± 200
11/9/88	1515	11/16/88	1045	7	406	128 ± 19	153 ± 7
11/16/88	1100	11/30/88	1330	14	406	153 ± 23	60.0 ± 4.7
11/30/88	1400	12/14/88	0930	14	408	129 ± 22	31.8 ± 1.2
12/14/88	0945	12/28/88	0920	14	405	54.4 ± 2.6	22.2 ± 1.4
12/28/88	0935	1/11/89	0915	14	406	38.3 ± 7.8	18.6 ± 0.1
1/11/89	0930	1/25/89	1430	14	407	47.2 ± 7.1	12.5 ± 0.6

TABLE A.2. Leach Data for Graphite Sample SUP NORD

Start		End		Elapsed Time, Days	Leachant Vol. (mL)	¹⁴ C Conc. (d/m•mL)	³⁶ Cl Conc. (d/m•mL)
Date	Time	Date	Time				
10/26/88	1340	11/2/88	1045	7	407	211 ± 31	173 ± 17
11/2/88	1050	11/9/88	1500	7	405	169 ± 25	34.8 ± 4.1
11/9/88	1545	11/16/88	1045	7	405	151 ± 23	9.79 ± 0.59
11/16/88	1100	11/30/88	1330	14	409	186 ± 28	5.78 ± 0.06
11/30/88	1400	12/14/88	0930	14	405	180 ± 45	4.06 ± 0.38
12/14/88	0945	12/28/88	0920	14	405	143 ± 18	2.90 ± 0.16
12/28/88	0935	1/11/89	0915	14	405	69.3 ± 5.9	2.58 ± 0.18
1/11/89	0930	1/25/89	1430	14	406	59.5 ± 7.1	2.05 ± 0.92

TABLE A.3. Leach Data for Graphite Sample B A

Start		End		Elapsed Time, Days	Leachant Vol. (mL)	¹⁴ C Conc. (d/m•mL)	³⁶ Cl Conc. (d/m•mL)
Date	Time	Date	Time				
10/26/88	1340	11/2/88	1045	7	405	110 ± 17	148 ± 20
11/2/88	1050	11/9/88	1500	7	408	114 ± 18	28.8 ± 3.2
11/9/88	1515	11/16/88	1045	7	404	89.9 ± 14	7.04 ± 0.03
11/16/88	1100	11/30/88	1330	14	406	124 ± 19	2.78 ± 0.53
11/30/88	1400	12/14/88	0930	14	408	106 ± 8	1.12 ± 0.04
12/14/88	0945	12/28/88	0920	14	407	73.4 ± 21	0.92 ± 0.07
12/28/88	0935	1/11/89	0915	14	408	63.3 ± 10.9	0.867 ± 0.138
1/11/89	0930	1/25/89	1430	14	407	84.7 ± 6.5	0.667 ± 0.032

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