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AN ANALYSIS OF THE HYDROGEN
BUBBLE CONCERNS IN THE THREE-
MILE ISLAND UNIT-2 REACTOR VESSEL

by:

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ABSTRACT

- (1) After identifying and analyzing all sources of non-condensable gases in the "bubble", we determined that a continuing growth of the "bubble" during the period Friday, March 30 through Sunday, April 1 as reported in the press was not possible.
- (2) During the first sixteen hours after reactor shut down, boiling of the primary coolant water took place and therefore in the worst case stoichiometric amounts of hydrogen and oxygen were produced by radiolysis of the primary coolant water. At the end of the sixteen hour period, the maximum amount of oxygen in the non-condensable bubble, including O_2 introduced from addition of air saturated water, was .7% of the hydrogen which is well below the explosion limit.
- (3) After this sixteen hour period when boiling had ceased, no further oxygen was produced by radiolysis of the reactor coolant water. On the contrary oxygen was recombined with hydrogen due to radiation at such a rate that the oxygen dissolved in the water was completely removed in less than five minutes. The subsequent removal rate of oxygen from the bubble by dissolution and radiolysis in the water depended essentially on the rate of dissolution.

Introduction

On Friday, March 30, two days after the accident at the Three-Mile Island Unit-2 Station, reports of a hydrogen bubble inside the reactor vessel began to appear in the news media. The following excerpts taken from a series of articles in the Washington Post give some indication of the nature of the information that was being provided to the general public.

"This bubble had appeared late Thursday or early Friday. At first, the Metropolitan Edison people believed that it was a steam bubble. Then the experts from Babcock and Wilcox which built the plant and the Nuclear Regulatory Commission agreed that, with pressures up to 1000 psi in the reactor, it couldn't possibly be a steam bubble. A steam bubble would have collapsed. That left only one possibility; a gas bubble containing hydrogen, temperamental volatile hydrogen. The bubble, a thousand cubic feet and growing would make Saturday the worst day of the crisis".

"The second alarming development was the gas bubble containing hydrogen, 1000 cubic feet in size at the top of the reactor. The reactor had become so hot that the coolant water

had decomposed to its primary elements, oxygen and hydrogen. The biggest danger was that the bubble would continue to grow forcing all the coolant out of the reactor, allowing the temperature of the fuel rods to build up until they reached 5000°. At that heat, the uranium would begin to melt. Short of the meltdown, there was a possibility of an explosion either in the containment building or in the reactor core. On the first day of the accident, there had been a small hydrogen explosion in the containment - an event Met Ed officials didn't tell state or federal officials about. When NRC experts found out, they launched an immediate effort to analyze the physical chemistry of the bubble. Thornburgh was told that the NRC's analysis showed that the hydrogen could become flammable or explosive in a matter of days. A Princeton University scientist calculated that the energy in the bubble was enough to set off an explosion equal to three tons of TNT. Such a force could rip the top of the reactor dome right off, flooding the containment with radioactive debris. There were also fears that the hydrogen would escape to the containment and explode there. One engineer calculated that a hydrogen explosion three times the force of Wednesday's blast might break

the four foot thick walls of the containment releasing radioactive materials into the air".

"Just before 8:30 p.m. (Saturday, March 31) came the final straw. Associated Press sent out an urgent story warning that the bubble situation had become extremely dangerous. In fact, the story warned, the unnamed experts were warning that the bubble might explode at any minute".

"By 10:00 a.m. on Monday, they were readying bulletin material that would take the word across the country. Minutes later the dispatch was torn from the wire machine at a Harrisburg radio station. An announcer read the news at 10:30: 'the hydrogen bubble was nearly gone and cooling of the reactor was continuing'".

While the details of the hydrogen bubble story vary in the different media accounts, they all contain the basic elements; that is, there was a hydrogen bubble inside the vessel and it was growing and about to explode at any moment. In this paper, we attempt to address both of these points; that is, could the bubble have been still growing during the period Friday through Sunday and was there any possibility of the bubble exploding.

Throughout the period of concern, the water inside the reactor vessel was being maintained at an average temperature of about 280°F and a pressure of 1000 to 1100 psi. Circulation was being maintained by operation of one reactor coolant pump in the A loop and heat was being removed through the A steam generator. The primary system flow as indicated by the flow meter in the hot leg of loop A was about 40% of full flow. The heat generation rate inside the reactor vessel was in the order of 27 MW from fission product decay plus 6.7 MW due to the operation of the one reactor coolant pump for a total of 1.2% of full power. All but one of the outlet thermocouples located directly above the core appear to have been working normally. Most of these thermocouples indicated temperatures within a few degrees of 280°F although, a few were as much as 100 to 200°F higher. However, none were reading in excess of the saturation temperature corresponding to the pressure range of 1000 to 1100 psi (550°F).

To address these two questions, it is necessary to start by identifying all the possible sources of gas which could have contributed to the bubble inside the reactor vessel. The first and most obvious source of gas in the bubble would be water vapor. However, at 280°F the vapor pressure of water is only 49.2 psi absolute. Since the total system pressure was at the order of 1000 to 1100 psi, it can readily be seen that water vapor could only have contributed about 5% of the total volume of the bubble.

The next source of gas to be considered are the stable isotopes of Krypton and Xenon which are formed as a result of radioactive decay of the fission products inside the fuel element. During normal operation, almost all of this gas is retained within the uranium dioxide fuel pellet. The remainder, of the order of a few percent, is released from the ceramic fuel pellets to the void spaces inside the zircaloy cladding of the fuel element. During the overheating of the core which occurred on the first day, the zircaloy cladding on most of the fuel elements was heavily oxidized and cracked. In addition, as the ceramic pellets themselves heated up, some of the gas retained in the UO_2 was released. It is therefore reasonable to assume that some substantial fraction of the stable Krypton and Xenon isotopes in the fuel was released to the gas bubble inside the vessel. If one assumes that all of the Krypton and Xenon stable isotopes that were present in the core were released to the reactor vessel during the overheating, and that none escaped through the stuck open relief valve, the following simple calculation shows that this source of gas could have contributed only a few tenths of one percent of the total volume of the bubble:

POWER HIS DRY: 95 Effective Full Power Days
 95 EFPD x 2772 MW = 2.63×10^5 MWD
 1 MWD = 0.96 gram U^{235} fissioned
 Total Fission yield of stable Kr and Xe

$$\frac{0.25 \text{ Moles of Gas}^{(1)}}{\text{Mole of } U^{235} \text{ fissioned}}$$

$$2.63 \times 10^5 \text{ (MWD)} \times 0.96 \frac{\text{gU}^{235} \text{ Fiss.}}{\text{MWD}} \times \frac{1}{235 \frac{\text{gU}^{235}}{\text{Mole}}} \times 0.25 \frac{\text{Mole Gas}}{\text{Mole } U^{235} \text{ Fiss.}}$$

$$\times 22.4 \frac{\text{liters STP}}{\text{Mole Gas}} \times 0.035 \frac{\text{ft}^3}{\text{liter}} = 211 \text{ ft}^3 \text{ at STP}$$

$$V_2 = V_1 \frac{P_1}{P_2} \frac{T_2}{T_1}$$

$$V_1 = 211 \text{ ft}^3$$

$$T_1 = 492^\circ\text{R} (32^\circ\text{F})$$

$$P_1 = 14.7 \text{ psia}$$

$$T_2 = 740^\circ\text{R} (280^\circ\text{F})$$

$$P_2 = 1000 \text{ psia}$$

$$V_2 = 211 \times \frac{14.7}{1000} \times \frac{740}{492} = 4.7 \text{ ft}^3 \text{ at } 280^\circ\text{F} \text{ and } 1000 \text{ psi}$$

Bubble volume measurements⁽²⁾ made on the 31st indicate about 1500 ft^3 at 970 psig and 280°F . Therefore the contribution of Krypton and Xenon was 0.3%.

(1) D. R. Olander "Fundamental Aspects of Nuclear Reactor Fuel Elements" TID-26711-P1, EROA, 1976, pp. 201-2.

(2) 230th General Meeting - Advisory Committee on Reactor Safeguards.

A second possible source of gas that comes from the fuel pins is the helium that is loaded into the zircaloy tubing along with the uranium dioxide pellets during fabrication. Discussions with the Babcock and Wilcox Company have indicated that the total amount of helium loaded into all of the fuel pins at fabrication amounted to 1,092 standard cubic feet. This helium fill gas also contained small amounts of oxygen and nitrogen impurities. The amount of nitrogen was 32 standard cubic feet and the amount of oxygen was 8.5 standard cubic feet. If it is assumed that all of the pins in the core failed and released all of the fill gas then the volume of this fill gas at the conditions of the bubble, 280°F and a 1000 psi, would have been 25 cubic feet. Again, it is apparent that the fill gas itself is a rather small fraction of the total volume of the bubble.

Another source of gas which has been considered is the nitrogen and oxygen dissolved in the water that was injected into the plant during the accident. The following calculation shows that this source of gas could have contributed at most 1% of the volume of the bubble.

Information received from the Babcock and Wilcox Company indicates that 284,000 gallons of borated water was pumped into the primary system. Assume that this water was saturated with air at about 1 atmosphere pressure and 10°C (50°F). At these conditions about 23 cm³ of air will dissolve in a liter of water. The composition of the dissolved gas will be about 34.5% oxygen.

Amount of Air in Water Added to the System

$$2.34 \times 10^5 \text{ (gal)} \times 3.79 \frac{\text{liter}}{\text{gal}} \times 23 \frac{\text{cm}^3 \text{ Air}}{\text{liter}} = 2.47 \times 10^7 \text{ cm}^3 \text{ Air}$$

$$\frac{2.47 \times 10^7 \text{ (cm}^3 \text{ Air)}}{2.24 \times 10^3 \text{ (cm}^3 \text{/Mole)}} \times \frac{273}{283} = 1.07 \times 10^3 \text{ gram Moles Air}$$

$$\frac{1.07 \times 10^3 \text{ (gram Moles)}}{454 \text{ (grams/pound)}} = 2.35 \text{ pound moles air}$$

Amount of Air Dissolved in the Water in the Primary System

From the TMI safety analysis report, it can be determined that the water volume in the reactor coolant system is about 11,500 ft³. Subtracting 1500 ft³ for the bubble leaves 10,000 ft³ or 74,800 gallons of water in the reactor coolant system. Thus, the amount of water pumped into the reactor coolant system was almost four times the amount present in the system, the remainder escaping through the stuck open relief valve. To provide an estimate of the maximum amount of air that could have been present in the bubble, assume that the water escaping through the relief valve contained no air. With this assumption, all the air from this 284,000 gallons of water pumped in remains in the reactor coolant system either dissolved in the 74,800 gallon of water in the system or in the gas bubble. The distribution of the air between the water and the gas bubble can be calculated from Henry's Law:

$$P_{\text{air}} = K X_{\text{air}} \quad (1)$$

Where: P_{air} = partial pressure of Air in the gas bubble

X_{air} = Mole fraction of Air in the water

K = Henry's Law constant for Air
(1.3×10^6 psia/mole fraction at 280°F)

plus the material balance shown in equation (2).

$$X_{\text{air}} M_1 + Y_{\text{air}} M_g = M_{\text{air}} \quad (2)$$

Where: M_1 = Total moles in liquid phase

M_g = Total moles in gas phase

M_{air} = Total moles of air

Y_{air} = Mole fraction of air in the bubble

and Raoult's Law:

$$P_{\text{air}} = P_t Y_{\text{air}} \quad (3)$$

Where: P_t = Total pressure

From equations (1) and (3)

$$P_{\text{air}} = K X_{\text{air}} = P_t Y_{\text{air}}$$

or
$$Y_{\text{air}} = X_{\text{air}} \frac{K}{P_t}$$

Substituting into equation (2)

$$X_{\text{air}} = \frac{M_{\text{air}}}{M_1 + \frac{K}{P_t} M_g} \quad (4)$$

As calculated previously $M_{\text{air}} = 2.35$ pound moles

Assuming that the total moles in the liquid phase is essentially equal to the moles of water:

$$M_L = \frac{10,000 (\text{ft}^3) \times 57 (\#/\text{ft}^3)}{18 (\#/mole)} = 3.11 \times 10^4 \text{ pound moles of water}$$

$$M_G = \frac{PV}{RT} = \frac{1000 \text{ psia} \times 1500 \text{ ft}^3}{10.73 \times 740} = 189 \text{ pound moles of gas}$$

Therefore:

$$X_{\text{air}} = \frac{2.35}{3.11 \times 10^4 + \frac{1.3 \times 10^6}{3} \times 1.89 \times 10^2}$$

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$$X_{\text{air}} = 8.7 \times 10^{-6}$$

From equation (1)

$$P_{\text{air}} = 1.3 \times 10^6 \times 8.7 \times 10^{-6}$$

$$P_{\text{air}} = 11 \text{ psia}$$

From equation (3)

$$Y_{\text{air}} = \frac{11 \text{ psia}}{1000 \text{ psia}}$$

$$Y_{\text{air}} = 0.011$$

Thus, the maximum amount of air in the bubble is 1.1% or 16.5 ft³ based on a 1500 ft³ bubble at 280°F and 1000 psia. The amount of air dissolved in the water would be ($X_{\text{air}} \times M_L$) = 0.26 pound moles.

Approximately eight hours after the beginning of the accident the reactor coolant system pressure was reduced to approximately 400 psi. Under these conditions, some fraction of the borated water contained within the core flood tanks would have been injected into the reactor coolant system. During normal operations,

the core flood tanks are maintained at a pressure of 600 psi by a nitrogen gas blanket. The water level in the tanks is maintained by a series of check valves which open as the reactor coolant pressure falls below 600 psi. The following analysis shows that the amount of dissolved nitrogen brought into the primary system from the core flood tanks amounts to only a fraction of 1% of the total volume of the gas bubble.

Each of the two core flood tanks normally contain 1040 ft³ of borated water (2270 ppm Boron) and 270 ft³ of nitrogen at 600 ± 25 psig. The minimum pressure indicated during the accident was 440 psig. Under these conditions the nitrogen would have expanded discharging about 1000 gallons of water from each tank.

$$2000 \text{ (gal)} \times 8.6 \frac{\text{pounds}}{\text{gal}} \times 454 \frac{\text{grams}}{\text{pound}} = 7.8 \times 10^6 \text{ grams of water}$$

$$P_N = K X_N$$

$$P_N = 585 \text{ psia}$$

$$K = 1.4 \times 10^3 \text{ psia cc-STP/g of water at } 50^\circ\text{C}$$

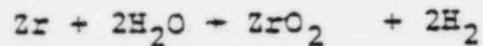
$$X_N = \frac{585}{1.4 \times 10^3} = 0.42 \text{ cc-STP/g of water}$$

$$0.42 \left(\frac{\text{cc-STP}}{\text{g of water}} \right) \times 7.8 \times 10^6 \text{ g of water} = 3.2 \times 10^6 \text{ cc-STP}$$

$$3.2 \times 10^6 \text{ (cc)} \times \frac{1}{1000 \text{ (cc/ft}^3\text{)}} \times 0.035 \frac{\text{ft}^3}{2} = 114 \text{ ft}^3 - \text{STP}$$

At the conditions of the bubble, 1000 psi and 280°F, this nitrogen would have contributed 2.5 ft³.

The total volume of all the gas sources identified thus far is of the order of 125 cubic feet at 280°F and 1000 psi or 7% of the gas bubble. The two sources of gas which have not yet been considered are hydrogen from the corrosion of the zircaloy by water according to the reaction:



and the hydrogen and oxygen formed by the radiolysis of water. The core of the Three-Mile Island reactor contains approximately 21 tons of zircaloy cladding not including fuel pin end fittings, instrument tubes, or other zircaloy components in the reactor. From the above reaction, it can be seen that one pound mole of zirconium reacting with water will produce two pound moles of hydrogen gas. The 21 tons of zircaloy cladding corresponds to 460 pound moles of zirconium which if completely reacted would produce 920 pound moles of hydrogen. It was shown earlier that the 1500 cubic foot bubble at 280°F at a 1000 psi would contain of the order of 200 pound moles of gas. It is therefore apparent that hydrogen from the zirconium water corrosion reaction is more than sufficient to contribute the other 92% of the observed gas bubble volume.

The final potential sources of gas, hydrogen and oxygen from radiolytic decomposition of water, will be considered in detail later. However, it is appropriate at this point to consider whether any of the sources of gas which have been identified thus far could have contributed to the impression that the gas bubble was still growing. To address this question,

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We will make the arbitrary assumption that a 10% per day increase in the gas bubble size, that is, 150 cubic feet per day will be considered as significant. While this is admittedly a purely arbitrary choice, it seems unreasonable that a much smaller growth rate (e.g. 1% per day) would have been reported by the news media as the immediate danger indicated by the excerpts from the Washington Post. It should be remembered that the bubble measurements over the period in question exhibited a downward trend, albeit with considerable scatter in the results, and that the bubble growth "threat" disappeared quickly from the media. However, it is appropriate to ask if the information at hand was sufficient to preclude an immediate threat due to growth of the bubble.

All the sources of non-condensable gas, except for hydrogen from corrosion of the Zircaloy and radiolytic decomposition which has not yet been discussed, amount to at most 50 ft³. Since most of these gases would have been released early in the sequence, and their total volume was small, they could not have contributed to the impression that the bubble was growing. As will be shown later, radiolytic decomposition of water in the presence of a substantial over pressure of hydrogen could not have produced more gas. In fact the reactor was normally operated with an overpressure of hydrogen to prevent radiolytic decomposition of the water. Thus steam formation and additional corrosion of the Zircaloy are the only possible sources of gas which need to be addressed with respect to the question of continued bubble growth.

Given that the reactor coolant was at average temperature of 280°F, a pressure of 1000 to 1100 psi, and under forced circulation corresponding to about 20% of full flow (40% in one loop), continued bubble growth due to steam formation was not possible. While some local boiling or even steam blanketing in damaged regions was possible, the steam leaving such a region would tend to condense in the surrounding subcooled water. If steam did manage to reach the gas liquid interface and enter the bubble it would condense on the cold walls of the primary system which by this time would have been in equilibrium with the reactor coolant at 280°F.

The possibility of additional corrosion generated hydrogen giving rise to significant bubble growth during the period in question is somewhat more complex. However, it is quite reasonable, given a general knowledge of the Zircaloy corrosion rate as a function of temperature and the existing plant conditions, to conclude that it was very unlikely that significant corrosion of the Zircaloy could still have been taking place. Figure 1 shows the corrosion of Zircaloy in terms of oxygen pickup as a function of time and temperature^(3,4). An oxygen pickup of 0.15 grams/cm² corresponds to the complete corrosion of the 0.026" thickness of the fuel pin cladding. The vertical line at 0.15 gram/cm² indicates that cladding regions at temperatures above ~1150°C would have been completely oxidized during the first day and thus could not have been contributing additional hydrogen. For a clad region to contribute hydrogen throughout the period between the first and fifth day its temperature would have to be below ~1000°C.

(3) ORNL/NOREG - 17

(4) Lustman and Kenzie "The Metallurgy of Zirconium"

Using the same 10% per day rate as the definition of significant bubble growth, and recalling that the 1500 ft³ bubble contained 139 pound moles of gas, significant growth would be 19 pound moles of hydrogen generated or about 9 pound moles of Zircaloy oxidized per day. Nine pound moles of Zircaloy corresponds to all of the cladding on about 750 fuel pins.

The oxidation of 9 pound moles of Zircaloy would consume 1.3×10^5 grams of oxygen per day. From Figure 1, the average corrosion rate during the five day period at 1000°C (complete oxidation of the clad thickness) is 3×10^{-2} grams/cm² per day. Therefore, to consume 1.3×10^5 grams of oxygen per day would require that 4.3×10^6 cm² or ~ 10% of the total clad surface area in the core be at 1000°C. Lower clad temperatures would require a larger area with a temperature of 800°C corresponding to the involvement of half of the total cladding area in the core. Such high temperatures in large regions of the core which were essentially intact at this point (i.e. regions which have enough unoxidized Zircaloy left to represent a possible source of corrosion) is not plausible given the plant conditions of 280°F, 1000 psi and forced circulation. These regions would have to be steam blanketed since the saturation temperature at 1000 to 1100 psi is only 550°F. Steam blanketing at decay heat power levels and 20% full flow could only occur within or downstream of a flow blockage (i.e. heavily damaged region). Since the core uncovering would have proceeded generally from the top of the core downward, cladding in the upper regions of the core (i.e. downstream of a flow blockage) would have been heavily oxidized during the accident. Therefore, even if significant flow blockages were present during the time in question leading to temperatures

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of the order of 1000°C (1832°F), the clad in these regions could not have been contributing much hydrogen since it would have already been heavily oxidized.

Hydrogen Explosive Limits

Based on the previous discussion the composition of the bubble at the end of the first day, excluding the effects of radiolytic decomposition, is indicated in Table 1. From the information in Figure 2 of reference 5, the lower limit of flammability for a hydrogen, 5% steam mixture occurs at 5% oxygen and the lower limit for detonation occurs at 12% oxygen. As can be seen from Table 1, the maximum concentration of oxygen in the bubble, excluding any contribution from radiolysis, is 0.4%. Thus the key question becomes, could radiolytic decomposition of the water have contributed enough oxygen to reach levels of the order of 5 to 12% in the bubble?

Table 1

<u>Component</u>	<u>Volume at 1000 psi and 280°F (ft³)</u>	<u>Mole %</u>
Steam	74	5
Kr + Xe	4.7	0.3
N ₂ from Air	10.8	0.7
N ₂ from Core		
Flood Tanks	2.5	0.2
N ₂ from Helium		
Fill Gas	0.7	0.05
Helium	23	1.6
O ₂ from Air	5.7	0.4
O ₂ from Helium		
Fill Gas	0.2	0.01
Hydrogen	<u>1349 (balance)</u>	<u>91.7</u>
TOTAL	1471	100

Analysis of O₂ Evolution by Radiolysis

The absorption of radiation in pure water in a light water reactor results in decomposition of the water through a complex series of reactions resulting initially in the production of reactive species such as: OH radical, hydrogen atoms (H) and hydrated electron (e^-_{aq}) as well as the products H₂ and H₂O₂. These species then react in a variety of ways. These reactions are listed in Table 2 (see for example reference 1).

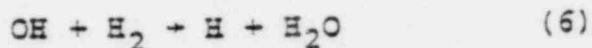
Table 2

<u>Reaction</u>	<u>k(25°C), (L mol⁻¹ s⁻¹)</u>	<u>E_a (kcal mol⁻¹)</u>
(1) H + H → H ₂	1 × 10 ¹⁰	3.0
(2) OH + OH → H ₂ O ₂	5.0 × 10 ⁹	3.0
(3) e ⁻ _{aq} + OH → OH ⁻	3 × 10 ¹⁰	3.5
(4) e ⁻ _{aq} + H → H ₂ + OH ⁻	2.5 × 10 ¹⁰	3.5
(5) e ⁻ _{aq} + e ⁻ _{aq} → H ₂ + 2 OH ⁻	5 × 10 ⁹	5.2
(6) OH + H ₂ → H + H ₂ O	4 × 10 ⁷	3.2
(7) OH + H ₂ O ₂ → HO ₂ + H ₂ O	2.25 × 10 ⁷	1.95
(8) H + H ₂ O ₂ → OH + H ₂ O	9 × 10 ⁷	3.0
(9) H + O ₂ → HO ₂	1.9 × 10 ¹⁰	3.0
(10) H + HO ₂ → H ₂ O ₂	2 × 10 ¹⁰	3.0
(11) OH + HO ₂ → H ₂ O + O ₂	1 × 10 ¹⁰	3.0
(12) HO ₂ + HO ₂ → H ₂ O ₂ + O ₂	1.6 × 10 ⁶	2.1
(13) e ⁻ _{aq} + H ₂ O ₂ → OH + OH ⁻	1.3 × 10 ¹⁰	3.5
(14) e ⁻ _{aq} + O ₂ → O ₂ ⁻	1.9 × 10 ¹⁰	3.5
(15) e ⁻ _{aq} + H ⁺ → H	2.3 × 10 ¹⁰	2.8
(16) e ⁻ _{aq} + H ₂ O → H + OH ⁻	16	6.7
(17) H + OH → H ₂ O	1.5 × 10 ¹⁰	3.0
(18) H + OH → e ⁻ _{aq} + H ₂ O	1.5 × 10 ⁷	4.0

The initial yields of the primary species, H, OH, e^-_{aq} , H_2O_2 and H_2 are each expressed in terms of a number called G which is the number of the species formed per 100 ev of radiation energy absorbed by the water. These initial yields depend principally on the nature of the radiation being absorbed by the water. This is a reflection of the density of ionization produced by the radiation along the track in which it is absorbed. In general particles which result in a high density of radiation along the track (high LET) favor a high initial yield of H_2 and H_2O_2 . Such particles are α particles, protons, deuterons and high energy neutrons (the effect of the neutrons in H_2O is due to the high energy proton recoils produced by elastic collisions with the neutrons). The particles producing a lower density of ions along the track (low LET) such as γ rays and β particles favor the production of H atoms, OH radicals and e^-_{aq} (2,3).

Therefore, if water is subjected to high LET radiation alone, the result would be a high initial yield of H_2 and H_2O_2 and, as the concentration of H_2O_2 builds up, reactions with H, OH and e^-_{aq} will decompose the H_2O_2 to water and oxygen. A stage is eventually reached when a stoichiometric amount of H_2 and O_2 would continuously form as the high LET radiation is absorbed.

In a mixed field of radiation, e.g., γ rays and protons (or proton recoils from fast neutrons), we have a situation where the H_2 and H_2O_2 resulting principally from the high LET radiation undergo reaction with H atoms and OH radicals produced principally by the low LET radiation. These reactions are illustrated by reactions 6 and 8 in Table 2:



It can be seen from these reactions that the OH and H atoms, produced principally by the low LET radiation (e.g. γ rays) recombine the hydrogen and hydrogen peroxide to form water in a chain reaction. Reaction (6) consumes an OH radical destroying a molecule of H_2 while producing an H atom; reaction (8) consumes an H atom destroying a molecule of peroxide and producing an OH radical which then can react by reaction 6 again resulting in an effective recombination of the hydrogen and hydrogen peroxide formed. Thus the formation of H_2 and O_2 is prevented.

Reactions listed in Table 2 which compete with this chain recombination of hydrogen and hydrogen peroxide to form water are for example reaction (7), (10), (11) and (12) which lead to the formation of oxygen. However, reaction (9) is a fast reaction with H atoms leading to the formation of the HO_2 radical which by reaction (13) forms hydrogen peroxide again.

It can be seen that anything which leads to the increased formation of H atoms and OH radicals will promote the recombination of hydrogen peroxide and hydrogen. One of the most effective additives is hydrogen gas which promotes the chain sequence of reactions (6) and (8) by increasing the rate of reaction (6) as well as (9). Another effective method is to increase the ratio of γ radiation (low LET) to fast neutrons (high LET).

Other factors which must be considered are: the impurities such as Cl^- . These tend to recombine the radicals H and OH and prevent their availability in recombining the hydrogen and hydrogen peroxide^(3,4). High temperature has little effect on the reaction rates producing the hydrogen and hydrogen peroxide but do have an appreciable effect on the reaction recombining these products. Hence high temperature tends to decrease the formation of hydrogen and oxygen. If hydrogen and oxygen are allowed to escape the aqueous phase by boiling or by being swept out with an inert gas, decomposition to H_2 and O_2 will be enhanced due to suppressing the back reaction (recombination)⁵.

With these considerations, the data one needs to calculate the rate of O_2 and hydrogen peroxide formation during full power operation, at shut down, and during the cooling period are:

1. γ ray flux
2. Fast neutron fluxes
3. β activity in the moderator
4. Thermal neutron fluxes
5. Initial conditions for any period including the concentration of the following materials in the moderator
 - a) O_2
 - b) H_2
 - c) Cl^- , F^- (major probable impurities on* the moderator of importance)
 - d) Boric acid
 - e) NaOH
- 6) Rate constants for elementary reactions (Table 2)
- 7) Temperature
- 8) Boiling

The following data were used in the computer calculation of O_2 formation.

Q (total neutron flux at full power) =

$$2.790 \times 10^{14} \text{ n cm}^{-2} \text{ sec}^{-1}$$

\bar{E}_n (average initial energy of neutrons = 2 MeV

$\bar{\Sigma}_a$ (flux weighted macroscopic cross section of core) =

$$2.28773 \times 10^{-2} \text{ cm}^{-1}$$

Volume of core = $3.08 \times 10^7 \text{ cm}^3$

Volume of coolant in core = $(3.08 \times 10^4 \times .58 \text{ L} = 17,858 \text{ L})$

Above data were taken from Appendix A.

*Since the rate constant for $OH + \text{halide ion}$ is less than $10^6 \text{ L mol}^{-1}\text{sec}^{-1}$, a simple calculation shows that at the low halide ion concentration present in the reactor coolant (.1 ppm), this reaction need not be considered.

The neutron dose then is:

$$\frac{2.79 \times 10^{14} \times 2.238 \times 10^{-2} \times 3.08 \times 10^7 \times 2 \times 10^6}{17,358} =$$

$$2.2 \times 10^{22} \text{ eV/L sec}$$

The photon dose at full power at shut down

$$= 2.2 \times 10^{21} \text{ eV/L sec}$$

Calculated from data
in Appendix A

Energy absorbed due to total α , Li recoils from B^{16} (n, α)
Li⁷ reaction⁶:

$$\frac{dE}{dt} \text{ (eV/sec mole boron)} = E_r f \sigma N[B]$$

$$E_r = 2.33 \times 10^6 \text{ eV} = \text{total } \alpha, \text{ Li recoil energy}$$

$$f = 4.74 \times 10^{13} \text{ n/cm}^2 \text{ sec (thermal) (Appendix A)}$$

$$[B] = 1.66 \times 10^{-5} \text{ mol cm}^{-3} \text{ (ref. 7)}$$

$$\sigma = 7.5 \times 10^{-22}$$

$$\begin{aligned} \frac{dE}{dt} &= 2.33 \times 10^6 \times 4.74 \times 10^{13} \times 7.55 \times 10^{-22} \\ &\quad \times 6.02 \times 10^{23} \times 1.66 \times 10^{-5} \\ &= 8.33 \times 10^{17} \text{ eV/L sec} \end{aligned}$$

This is small compared to the proton recoil dose rate and is neglected. Total activity due to fission products in the primary coolant is 6×10^{11} disintegrations $L^{-1} \text{ sec}^{-1}$. If we estimate 1 MeV average energy per disintegration and total absorption one would arrive at a dose rate of $6 \times 10^{17} \text{ eV L}^{-1} \text{ sec}^{-1}$, a factor of about 3×10^4 less than the γ dose rate at shut down. Even after 30 days following shut down the γ dose rate from the fuel is about 500 times that contributed by the fission products found in the primary coolant. Therefore, we neglect this contribution to the low LET dose.

The G values were calculated in the following way.

The average logarithmic decrement of energy loss per fast neutron collision with a target of atomic wt A is

$$\ln \frac{\bar{E}_1}{\bar{E}_2} = 1 + \frac{(A-1)^2}{2A} \ln \frac{A-1}{\ln(A+1)}$$

for A = 1 (proton):

$$\bar{E}_1 = 2.71 \bar{E}_2$$

\bar{E}_1 = average energy of neutron before collision

\bar{E}_2 = average energy of neutron after collision

$$\text{Proton recoil energy} = \bar{E}_1 - \bar{E}_2$$

The energies of the proton recoils from the first three neutron collisions are tabulated in Table 3. After 3 collisions, the proton recoils make only a small contribution to the dose compared to the first three collisions and their contribution to the dose is therefore neglected.

The LET values in Table 3 were determined by interpolation and extrapolation from data in reference 9. The $G(e^-_{aq})$ values in Table 3 are taken from reference 10. The values for $G(H)$, $G(H_2O_2)$ and $G(OH)$ were calculated from a diffusion kinetic model¹¹.

-Table 3

<u>Collision #</u>	<u>\bar{E}_n (MeV)</u>	<u>\bar{E}_p (MeV)</u>	<u>LET ($\frac{\text{MeV}}{\text{cm}}$)</u>	<u>G(H)</u>	<u>G(H₂O₂)</u>	<u>G(OH)</u>	<u>G(e⁻_{aq})</u>
	2						
1	.73	1.27	238	.49	.91	.46	.21
2	.272	.458	562	.30	.99	.24	.05
3	.100	.172	2000	.12	1.15	.09	0

Energy weighted G values for above three groups of proton energy are $\bar{G}(\text{H}) = .41$, $\bar{G}(\text{H}_2\text{O}_2) = .95$, $\bar{G}(\text{OH}) = .37$, $\bar{G}(\text{H}_2) = .855$ and $\bar{G}(\text{e}^-_{\text{aq}}) = .15$, all in units of particles per 100 eV absorbed.

We performed two groups of calculations using our computer code WR122 which is a modified version of code WR20¹².

Since there is some ambiguity of the actual pH of the water we made the calculations for 2 pH's namely 6 and 8 (measured at room temperature), assuming in the first case that pH 6 was achieved by addition of boric acid and in the second case by addition of the same amount of boric acid and the required amount of NaOH. The calculations were carried out for a temperature of 280°F under non-boiling conditions and the actual H⁺ and OH⁻ concentrations at that temperature were calculated for the above mixture and used. As confirmed by our later calculations, we found that the results were not very sensitive to the pH in this range.

The first calculation served to verify that during normal full power operation (with mixed neutron and γ radiation), the steady state concentrations of H_2 , H_2O_2 and O_2 are small. We used the dose rates cited in the text. For the γ yields, we used the following G values: $G(OH) = 2.7$, $G(H) = 0.45$, $G(H_2O_2) = 0.7$, $G(e^-_{aq}) = 2.81$, $G(H_2) = 0.45$.

The neutron G value have been described in the text. The ratio of dose rate to neutron dose rate was about ten to one and weighted average G values were accordingly calculated. We assumed an initial O_2 concentration of 0.1 ppm = $3.12 \times 10^{-6} \text{ Ml}^{-1}$ (maximum permissible O_2 concentration, table 5.2-12¹³). The initial hydrogen concentration used was $1 \times 10^{-3} \text{ Ml}^{-1}$, a value in the middle range of values specified in the above cited table. For the calculations to be realistic, we had to take into account the water circulation in the core. We used a value of $137.5 \times 10^6 \text{ lbs. water hr}^{-1}$ for the circulation rate and a total coolant volume of $2.14 \times 10^5 \text{ L}^{13}$. From these data and the water volume in the core we calculate that the total water volume circulates once every 23.6 seconds. As an approximation, we assumed that during this time, the difference in concentration of a species in the core water and the outside loop is reduced by a factor of $1/e$.

The results for the first group of calculations can be summarized as follows:

- A. Oxygen concentration in the core water drops down to essentially zero within 14 milliseconds (Fig. 2).
- B. A steady state concentration of about 9×10^{-6} mol L⁻¹ of H₂O₂ in the core is reached after 50-100 milliseconds (Fig. 3 and 4).
- C. The oxygen concentration in the circulating loop after 100 seconds has diminished to 1.44% of the initial value (Fig. 5).
- D. At the same time, the H₂O₂ concentration in the circulating loop has essentially reached a steady state of 9×10^{-6} mol L⁻¹ (Fig. 6 and 7).

The second group of calculations was carried out for the time 16 hours after shut down when boiling of the water had stopped. For the dose rate we used 3.4×10^{20} ev L⁻¹ sec⁻¹ which was calculated from the data in Appendix A, and zero neutron dose since the asymptotic total neutron dose (Table VII, Appendix A) was negligible compared to the γ dose. Actually this asymptotic neutron dose is higher than quoted in the table by a factor of 10 due to the contribution of the γ -n reaction on the deuterium in the coolant water¹⁵ but this value is still negligible and is neglected.

The calculation for the initial condition (H₂ and O₂ concentrations in the water) was based on a bubble size (H₂, O₂ and steam) of 1471 ft³ at 180°C and 970 psi. We assumed that during the boiling stoichiometric amounts of

O_2 and H_2 were produced with a G value for O_2 of .225 molecules per 100 ev energy absorbed in the water (one-half of $G(H_2)$). We also assumed that no gas production took place in uncovered portions of the core. We further assumed that the fraction of O_2 vented out during the boiling period was equal to the fraction of hydrogen and steam (added together) vented during this period. We arrive at a ratio of oxygen to hydrogen in the bubble at the end of the boiling period (16 hrs. after shut down) of 0.7%.

For the O_2 concentration in the water, we obtained a value of 2.6×10^{-4} mol/l. We calculate the hydrogen concentration to be 5.7×10^{-2} mol/l.

The details of these calculations are given in Appendix B.

The flow of cooling water in the core was taken into account as described for the first group of calculations, with the following modification:

1. The flow rate was reduced by a factor of 5 since only one pump in one cooling leg was in operation¹⁴.
2. The total water volume was reduced by the volume of the bubble

Using these initial conditions and assumptions and the computer code described above, we obtained results that can be summarized as follows:

1. Within about 280 seconds after boiling ceased, the O_2 in the core water is totally removed if during this time no further oxygen is dissolved from the bubble (Fig. 8).
2. The O_2 removal curve for the cooling loop (not shown) is essentially the same except for a slight delay due to the finite circulation rate.
3. The rate of removal of oxygen is approximately linear with time and therefore independent of O_2 concentration within a wide range. From the rate of removal at the dose rate used, we calculated an approximate G value for oxygen removal, $G(-O_2)$, of about 1.4 molecules/100 eV.
4. The behavior of the H_2O_2 and H_2 concentration in the core water is shown in Figs. 9 and 10, respectively. The corresponding curves for the cooling loop (not shown) are very similar.
5. Similar calculations for lower dose rates which prevail at longer times after shut down yielded analogous results and approximately the same value for $G(-O_2)$.

The value of $G(-O_2)$ quoted above was estimated in the following way: the curve in Fig. 8 shows a slope of about $2.2 \times 10^{-4}/280$ ($\text{mol L}^{-1}\text{s}^{-1}$), the irradiated water volume is 17858 L. Therefore, the total energy deposited per second is

$$3.4 \times 10^{20} \times 17858 = 6.07 \times 10^{24} \text{ eVs}^{-1}$$

The total circulating water volume is 177400 L. Therefore, the total amount of O_2 removed in 280 s is

$$2.2 \times 10^{-4} \times 177400 = 39 \text{ mol } O_2$$

Thus

$$\begin{aligned} G(-O_2) &= \frac{39(\text{mol}) \times 6.02 \times 10^{23}(\text{molec. per mol}) \times 100 \text{ (eV)}}{280 \text{ (s)} \times 6.07 \times 10^{24} \text{ (eVs}^{-1}\text{)}} \\ &= 1.4 \text{ molecules } O_2/100 \text{ eV} \end{aligned}$$

These results show that the only time oxygen was produced was under boiling conditions during the first 16 hrs. after shut down. After this period our calculations show that in the worst case only about 0.7% of O_2 could have been contained in the bubble. After this period, no further production of oxygen could have occurred. In fact, the calculations indicate that oxygen would be continually consumed, the rate being essentially determined by the rate of dissolution of oxygen from the bubble. Even under full power with mixed γ and neutron fluxes and a very small hydrogen overpressure of about 1 atmosphere, oxygen would not be produced but actually consumed until a non-measurably low steady state concentration of oxygen is reached.

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APPENDIX A

ENERGY RELEASE RATES INTO THE CORE COOLANT

The energy deposition in the coolant as it flows through the TMI-2 core is primarily due to two sources: gamma (γ) emissions and beta (β) decay. A tertiary source results from the neutron flux interaction with the water and soluble boron, however, after shutdown it is relatively minor in comparison with the γ and β sources. A description of the calculational procedure which was employed to produce the γ and β release rates follows in Section I. Section II describes the approximation by which the energy release rate is transformed into an energy absorption rate. For completeness, an estimate of the neutron flux level following neutronic shutdown is presented in Section III.

I. γ and β Energy Release Rates

A simplified calculational model for TMI-2 was created for the required calculations. Relevant details concerning such quantities as the heavy metal loading of the initial core, power densities and rod design were obtained from the Final Safety Analysis Report. When certain secondary details could not be obtained from the FSAR, typical values were utilized from similar plants. A summary of the calculational parameters which were utilized in the TMI-2 model is presented in Table I.

The γ and β release rates were calculated with the ORIGEN¹ code. ORIGEN is a point depletion code which solves the first order differential equations which describe the production and decay of the various nuclides. In this analysis, the cross section library contained 821 fission product nuclides, 146 light elements and materials of construction, and 99 heavy nuclides plus their daughters. The code plus cross sections were benchmarked against the ANS decay heat standard and shown to produce reasonably accurate PWR spent fuel decay heat information.

The ORIGEN input was created to match the information of Table I: the key quantities were the heavy metal loading, power level and burnup. The output yields directly the total release rate ($\gamma + \beta$) and the γ release rate. The contribution from the β particles is, therefore, inferred from these two values. The accuracy of this inferred value is limited due to the fact that the ORIGEN output yields only 3 significant digits. Table II lists the photon energy release rate (MeV/sec for the entire core) as a function of time after neutronic shutdown and Table III presents the corresponding values for the β energy release rate.

II. γ and β Energy Absorption Rate in the Coolant

The energy release rate values of Section I can be transformed into whole core energy absorption rates by utilizing the following approximations. First, it is assumed that the entire photon population is absorbed within the core boundaries. This, of course, is a conservative assumption. Secondly, the fraction of photons which are absorbed in the coolant is assumed to be equal to that fraction of the homogenized (whole core) macroscopic photon removal cross section which is due to the coolant alone.

Gamma attenuation (or mass attenuation) coefficients are dependent upon the energy of the photon; this dependency will be neglected in this approximate analysis so as to render the problem manageable. Instead, attenuation coefficients are selected at the energy near which the great majority of photons are released: 1 MeV.* To further simplify the analysis, the Zirc-4 and structural steel in the core is assumed to absorb photons at the same rate as the element iron (Fe) does. Likewise, the heavy metal fuel absorption is modeled by lead's (Pb) γ -absorption properties (this assumption, in fact, is utilized by B&W in their analysis of TMI-2.) With these assumptions, the core photon absorption properties are modeled as in Table IV. Thus, to a first approximation, it can be assumed that the coolant absorbs about 12.6% of the total photons released by the core.

Utilizing a slightly different approach, the coolant absorption fraction is arrived at via mass attenuation coefficients as shown in Table V. By using an average water density of 1.0 (a conservative assumption) the fraction of photons absorbed within the coolant is coincidentally equal to 12.6% again. As the two models agree so closely, this value seems to be an excellent approximation within the context of this analysis.

The TMI-2 FSAR (Appendix 6A) contains a short description of a calculation concerning gamma absorption in the coolant following a LOCA and a MHA (maximum hypothetical accident). [The latter includes the release of 1% of the solids and 50% of the halogen inventory and therefore has greater activity than would be expected for TMI-2's current situation.] The LOCA analysis will serve as a better reference point.

The B&W (FSAR) analysis uses an unspecified proprietary code: salient differences are as follows. The ORIGEN code has over 800 explicit fission product nuclides and hundreds of heavy metal and structural isotopes. The B&W code has only 200 fission products modeled along with ^{239}Pu and ^{239}U . The neglect of the remaining isotopes could lead to non-conservative results, however, details are too sparse to allow such a prediction. The B&W code also allows β penetration through the cladding and energy deposition into the water outside the core boundary. The cumulative effects are to increase the total absorption rate by 5-7%. Finally, the FSAR analysis was for a core at 620 FPD as opposed to our calculations at 60 FPD. The impact of the burnup differential will be discussed below.

Figure 1 reproduces the LOCA energy deposition curve** versus time. Also depicted is the energy deposition into the coolant as predicted with the ORIGEN code in conjunction with the assumptions describes above, i.e. 12.6% of the total will be absorbed in the coolant. (The cumulative energy deposition was found by applying piecewise continuous trapezoidal and Simpson integration to the values of Table II.

*The neglect of the energy dependency of the attenuation coefficients should be a second-order effect.

**TMI-2 FSAR, Vol. 5, Chap. 6, Fig. 6A-3.

It can be seen that the ORIGEN values agree with the FSAR values to within a factor of 1.4. ORIGEN, not surprisingly, produces the more conservative values between the two.

The effect of burnup upon this curve was established by comparing instantaneous γ -release rates (as opposed to the cumulative energy deposition curve of Fig. 1) from the point of shutdown to 30 days after shutdown. The ratio of γ -release rates for BU = 20,000 MWd/t (592 FPD) to the rate at 2,000 MWd/t (60 FPD) ranged monotonically from 0.97 at shutdown to 1.94 at 30 days. The effect of increased burnup would shift the slope of the ORIGEN curve in a direction which is more consistent qualitatively with the shape of the B&W curve; quantitatively the ORIGEN values would then be within a factor of 2 of the LOCA values. It is concluded that the ORIGEN values, in combination with a coolant absorption fraction of 0.126, will yield acceptably accurate values for the γ -energy deposition.

The β -energy deposition values must be created from the values of Table III by accounting for β penetration of the cladding and the possible release of (primarily) fission products directly into the coolant. Penetration of the cladding should contribute only 2+3 percent to the total energy deposition. An estimate of the amount of heavy metal and fission products which were released into the coolant must be made in order to account for the second β source.

III. Neutron Flux Levels

The neutron flux level after reactor scram is composed of several parts. First, the full power flux level decays away, normally within the period of about 100 milliseconds. Secondly, the equilibrium precursor concentration from full power decays, which produces a significant neutron source over a time period of ~30 minutes. Lastly, the flux will reach an asymptotic level which represents the multiplication of the residual sources (spontaneous fission, α -n and the startup sources) which remain after the first two flux components have decayed to negligible values.

1. Flux Level Computational Procedure

The neutron flux level from the first two contributors is calculated via standard point kinetics with 6 delayed neutron groups;

$$\dot{n} = n \frac{\rho - \beta}{\Lambda_{eff}} + \sum_{i=1}^6 \lambda_i C_i, \quad (1)$$

and

$$\dot{C}_i = -\lambda_i C_i + \frac{\beta_i}{\Lambda_{eff}} n. \quad (2)$$

The computer code REKINS² was utilized to solve Eqs. 2 and directly yield the flux level from this source. The transition to the asymptotic flux level was approximated by simply superimposing the above solution upon the flux level which results from the multiplication of the spontaneous fission, α -n source and the startup sources as described below.

The asymptotic steady-state source multiplication term of the point kinetics equation reduces to

$$S_{\text{mult}} \equiv \frac{s}{|\rho|}, \quad (3)$$

with s representing the neutron release rate (n/sec) of the individual source.

Equation 3 yields the steady-state neutron production (n/sec). When this value is multiplied by the effective neutron lifetime, l_{eff} , the total average neutron population (# of neutrons) at any point in time is realized. The neutron density is then formed:

$$n = S_{\text{mult}} l_{\text{eff}} / \text{Volume}_{\text{core}} \quad (\text{n/cm}^3). \quad (4)$$

The classic neutron flux can now be defined as

$$\phi \equiv n \bar{v} \quad (\text{n/cm}^2\text{-sec}), \quad (5a)$$

$$= \frac{s}{|\rho|} \frac{l_{\text{eff}}}{\text{Volume}_{\text{core}}} \bar{v} = \frac{s}{|\rho|} \frac{l_{\text{eff}} \bar{v}}{\bar{\Sigma}_a \text{Volume}}, \quad (5b)$$

with \bar{v} representing the average velocity of the neutron (approximated as $\bar{v} = \frac{l_{\text{eff}}}{\bar{\Sigma}_a}$)

and $\bar{\Sigma}_a$ being a flux and core averaged macroscopic absorption cross section. The prompt neutron lifetime (BOL) for TMI-2 is reported to be 2.71×10^{-5} seconds. [Two different approximations for the effective neutron lifetime utilizing the calculational model of this study yielded values of 3.1×10^{-5} and 1.2×10^{-5} seconds.] For purposes of this approximate calculation, the FSAR value of 2.71×10^{-5} seconds will be utilized as approximately equal to l_{eff} .

2. Source Rate Definitions

The neutron release rates (ν) in the core regions are estimated from 3 separate components: spontaneous fission of the actinides, α -n reactions (i.e. the production of α particles via α -decay in transuranics and their subsequent interaction with ^{17}O and ^{18}O) and the startup sources. ORIGEN provides the spontaneous fission information via the following approximation for ν :

$$\nu = 2.84 + 0.1225 (A - 244), \quad (6)$$

with A representing the atomic weight of each spontaneously fissioning nuclide. The energy dependence of these neutrons is not explicitly given; however, it is adequately approximated with the ^{235}U fission spectrum. Therefore, the fission neutrons will have a mean energy in the range of 1-2 MeV. The validity of this approximation has been checked against selected nuclides and has been shown to be reasonable (see, e.g., DP-984 and DP-939 for ^{242}Cm , ^{244}Cm , ^{252}Cf and ^{238}Pu).

During startup, external sources (Am-Cm-Be), are utilized to initiate the chain reaction. Our information, as of this date, is that 2 fixed sources were in place at the time of the accident. Each source has a strength of 1.4×10^9 n/sec. Since the reported strength has only 2 significant digits, variations in the strength over time are neglected as second-order effects. The exact location of the startup sources is not specified and must be assumed. A conservative assumption is to place the sources far enough into the core so that all of the released neutrons are effectively multiplied and exhibit the characteristic flux shape/spectrum in the core: this is equivalent to assuming a homogeneous distribution of the external sources throughout the core. (In addition to being conservative, this assumption is consistent with the methodology utilized previously.) The flux level is then calculated via standard source multiplication.

The α -n source is predicted by ORIGEN via the following:

$$\text{neutrons/alpha disintegration} = 10^{-10} E_{\alpha}^{2.65}, \quad (7)$$

where E_{α} represents the alpha particle energy in MeV. Equation 7 is based upon experimental results from $^{238}\text{PuO}_2$, yet is applied to all alpha emitters. The validity of this approximation is not known; however, the impact will be insignificant as the startup source dominates the total source term. The mean energy of these neutrons, for several particular isotopes, was in the range of ~2.5-3.0 MeV. Thus, the neutron spectra appear to be similar to the spontaneous fission spectra, but with a slightly higher mean energy.

Table VI lists the fixed (startup), α -n and spontaneous fission neutron sources as a function of time after shutdown. The values are relatively constant due to the extremely long half-lives associated with most of the determining reactions and decay chains.

3. Neutron Flux Level Results

Tables VIIa and VIIb list the neutron flux levels which result from Eqns. 1, 2 and 3. The shutdown k_{eff} value is not accurately known; therefore, two representative values were chosen (0.99 and 0.98) to illustrate the effect upon the neutron flux levels which can result from varying this quantity. During the solution of the point kinetics equations (Eqns. 1 and 2), the shutdown mechanism was a linear reactivity ramp imposed over 1.5 seconds (this corresponds approximately to the time required to SCRAM). Varying the ramp rate had little perceptible effect upon the values, thus the approximation was deemed adequate. It should be stressed that flux levels of Table VII are based upon the assumption that there is no flux shape change during the transient. That is, the spectral shift which results from the shutdown mechanism is neglected and does not enter either the analysis or the spectrum discussion of the following subsection. Additionally, the source multiplication term is based upon an approximate value of $\bar{\nu}_a$ (Eqn. 5). As such, the flux values are approximate.

4. Neutron Flux Energy Spectrum

The energy spectrum of the combined neutron sources will be primarily dependent upon the lattice parameters and slowing down properties of the core. The components of the neutron flux are all created within the fuel, and will thus form their representative energy spectrum in the same fashion as the fission neutrons did during full power operation. The only difference is in the neutrons' initial energy. The fixed startup and spontaneous fission source neutrons have initial energy spectrums which are very similar to the ^{235}U fission source, therefore, the resultant cell averaged spectrum will be virtually identical to the spectrum present during normal operation. The α -n source has initial energy spectrums which are similar in shape to the fission spectrum, but with mean energies which are higher by ~ 1 to 1.5 MeV. These neutrons would, of course, lead to a harder resultant spectrum than the fission neutrons would. However, the difference between it and the spontaneous fission neutron contribution should be relatively small. That is, in a light water moderator environment, the difference between a spectrum which results from fission neutrons at $1\text{--}2$ MeV and the spectrum from α -n reactions with energies nominally near $2\text{--}3.5$ MeV would be virtually eliminated by the dominant slowing down properties of water. High energy (i.e. ~ 1 MeV) neutron reactions would increase significantly, however, this should be a second-order effect (globally). Thus, the neutron spectrum which results from the α -n, the spontaneous fission neutrons and the fixed startup source should be approximately equal to the spectrum which was present during full power operation.

The delayed neutrons, which were created during the full power operation phase, typically have energies in the range of $250\text{--}560$ keV. These energies are smaller than those of the previously described sources, but not considerably so. More importantly, these energies are above the resonance range, and therefore, the delayed neutrons will be affected by nearly all of the important slowing down processes: the only relevant spectral effect which is missing is, of course, the high energy effect. The effect is mitigated to a considerable extent due to the multiplication of the delayed neutrons by the

core. For example, in the case where $k_{\text{shutdown}} = 0.98$, the system multiplication is $|\rho|^{-1} = 49$. Thus, only 1 of every 50 neutrons will be a delayed neutron and the spectral impact is negligible.

The energy spectrum of the cell averaged neutron flux is, therefore, approximated with the full power spectrum. A typical 5 group spectrum is listed in Table VIII for full power and 15 minutes after shutdown. Additionally, the inferred neutron density values are also listed.

5. Approximate Neutron Energy Deposition
During Thermalization Process

An estimate of the energy deposition in the moderator which results from the neutron flux was made with the following assumptions. First, the energy deposition mechanism in the coolant is assumed to be scattering: energy release due to absorptions is neglected. Secondly, the thermalization process is typified as occurring exclusively in the moderator region (a conservative assumption) and that each neutron losses 2 MeV during the process. This last criteria results from the fact that the average fission neutron energy is ~2 MeV and that ~99.8% of all neutrons slow down to at least group 3 of the 5 group structure (E_{max} of group 3 = 553 eV). Therefore, the energy loss per neutron is approximately 2 MeV.

The neutron source rate which produces the flux levels in Table VII can be inferred by rearranging Eqn. 5:

$$\phi = S_{\text{mult}} \frac{1}{\bar{\Sigma}_a \cdot \text{Volume}_{\text{core}}} \quad (\text{n/cm}^2\text{-sec}), \quad (8a)$$

or

$$S_{\text{mult}} = \phi \bar{\Sigma}_a \cdot \text{Volume}_{\text{core}} \quad (\text{n/sec}). \quad (8b)$$

By inserting a flux value into Eqn. 8b (along with the core average macroscopic cross section and core volume from Table I), the neutron source rate is found. Since 2 MeV are deposited per neutron, the energy deposition rate is

$$D_n = 2 \phi \bar{\Sigma}_a \cdot \text{Volume}_{\text{core}} \quad (\text{MeV/sec-core}), \quad (9a)$$

or

$$D_n = 1.40878 \times 10^6 \phi \frac{\text{MeV}}{\text{sec-core}}. \quad (9b)$$

is to the conservative assumption that all of the energy is deposited into the coolant, the units of Eqn. 9b are also MeV/sec-coolant. Of course, the coolant refers only to that volume of water which is within the core boundaries: energy-deposition densities should be based upon this volume, not the core volume.

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IV-2. NORTH & SOUTH

Hours after Shutdown

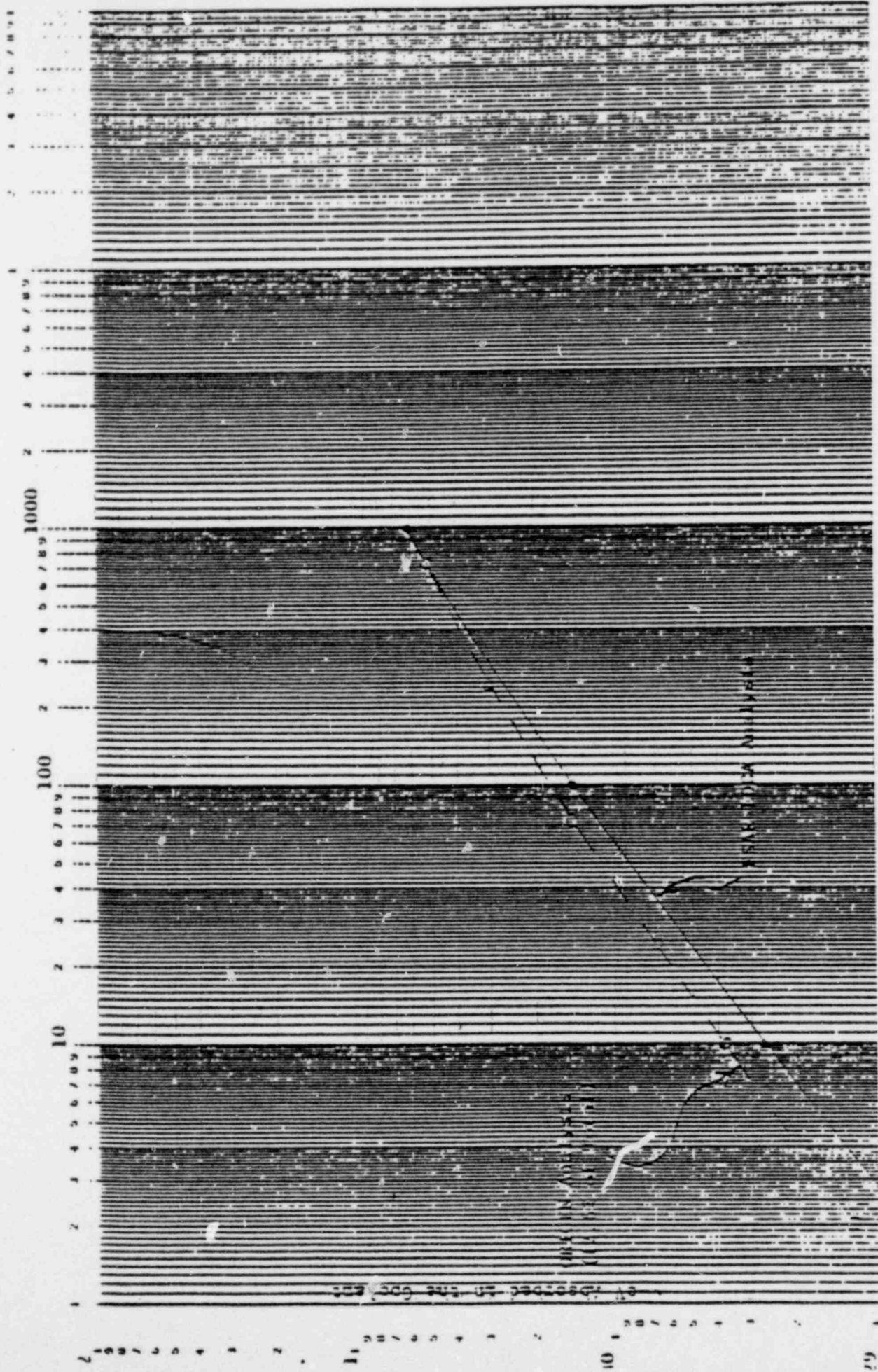


Fig. 1. Cumulative Photon Energy Deposition in the Coolant

TABLE I. Three Mile Island Unit 2 Calculational Parameters for Initial Loading

Power, MWt	2772
Power Density, kW/l core	90.0
Core Volume, l	30,790
Fuel Assembly Volume Fractions	
Fuel	0.303
Moderator	0.580
Zircaloy	0.102
Stainless Steel	0.003
Void	0.012
Initial Loading	
kg U	82064
kg ^{235}U	2111
Average Enrichment wt%	2.57
Power Density MW/MTHM	33.78
Burnup MWd/MTHM	2000
Effective Full Power Days	59.2
β_{eff}	0.00691
λ_p , sec ($\sim = \lambda_{\text{eff}}$)	2.71×10^{-5}
λ_d (longest lived precursor), sec^{-1}	0.0124
$\bar{\lambda}_a$ (ϕ weighted)	2.28773×10^{-2}

TABLE II. Proton Release Rates after Neutronic
Shutdown, MeV/sec, Enriched Core

Time	Light Element, Clad and Structure	Fission Products	Actinides and Daughters	Percent of Total FROB Fission Products	Total
shutdown	9.68 +16	3.04 +20	1.34 +19	95.8	3.175 +20
1 min.	7.90 +16	1.69 +20	1.20 +19	93.3	1.811 +20
1 hour	7.37 +16	1.10 +20	1.03 +19	91.4	1.204 +20
1 hours	7.33 +16	8.47 +19	9.64 +18	89.7	9.442 +19
1 hours	7.72 +16	4.94 +19	8.64 +18	85.0	5.812 +19
1 hours	7.69 +16	3.73 +19	7.28 +18	83.5	4.466 +19
1 days	7.64 +16	2.51 +19	4.03 +18	85.9	2.923 +19
1 day	7.58 +16	2.06 +19	2.25 +18	89.9	2.293 +19
2 days	7.44 +16	1.46 +19	5.28 +17	96.0	1.520 +19
3 days	7.13 +16	9.16 +18	3.35 +16	98.9	9.265 +18
7 days	6.77 +16	6.48 +18	3.87 +15	98.9	6.552 +18

TABLE III. Beta Release Rate^a after Neutronic Shutdown, MeV/sec, Entire Core

Time	Light Element, Clad and Structure	Fission Products	Actinides and Daughters	Percent of Total from Fission Products	Total
Shutdown	9.55 +16	5.41 +20	3.48 +19	93.9	5.759 +20
5 min.	8.99 +16	1.34 +20	2.61 +19	83.6	1.602 +20
1 hour	8.99 +16	8.49 +19	1.47 +19	85.2	9.969 +19
2 hours	8.92 +16	6.62 +19	1.11 +19	85.5	7.739 +19
3 hours	8.68 +16	3.56 +19	9.46 +18	78.8	4.515 +19
14 hours	8.49 +16	2.35 +19	7.96 +18	74.5	3.154 +19
3 days	7.93 +16	1.47 +19	4.42 +18	76.6	1.920 +19
5 days	7.49 +16	1.22 +19	2.46 +18	82.8	1.473 +19
10 days	6.62 +16	9.17 +18	5.79 +17	93.4	9.815 +18
20 days	5.43 +16	6.30 +18	3.84 +16	98.5	6.393 +18
30 days	4.56 +16	4.87 +18	5.90 +15	99.0	4.922 +18

^aEqual to the difference between the total release rate ($\gamma+\beta$) minus the γ release rate: since only 3 digits are yielded from ORIGEN for these quantities, the accuracy of the above numbers is less than the number of digits presented.

TABLE IV. Photon Removal - Model 1

Region (Material)	Σ_r Removal Cross Section for 1 MeV Photons (Reference 2)	V.F. (Volume Fraction)	$\Sigma_r^*V.F.$	Percent of Core Homogenized Removal
Fuel (Pb)	0.776	0.393	0.04914	12.1
Clad and Structure (Fe)	0.468	0.105	0.23513	15.4
Moderator (H ₂ O)	0.0706	0.580	0.04095	12.6
			Total 0.32522	
			(Core Homogenized value)	

TABLE V. Photon Attenuation - Model 2

Region (Material)	Mass Attenuation Coefficient ^a (Ref. 3)	Assumed Density (g/cc)	Product	Product Averaged	Percent of Homogenized Total
Fuel (Pb)	0.0684	10.4	0.7114	0.2155	73.6
Clad and Structure (Fe)	0.0595	6.5	0.3868	0.0406	11.9
Moderator (H ₂ O)	0.0706	0.7+1.0	0.0635 (using $\bar{\rho} = 1.0$)	0.0369 <u>0.2930</u>	12.6
				Homogenized Total	

^a1 MeV γ 's (cm²/g)

TABLE VI. Neutron Sources After Neutronic Shutdown, Neutrons/sec

Time	Fixed Neutron Source	α -n ^a	Spontaneous Fission	Total
Shutdown	2.8 +9	1.22 +7	5.40 +6	2.818 +9
15 min.	2.8 +9	1.22 +7	5.40 +6	2.818 +9
1 hour	2.8 +9	1.22 +7	5.40 +6	2.818 +9
2 hours	2.8 +9	1.22 +7	5.40 +6	2.818 +9
10 hours	2.8 +9	1.23 +7	5.41 +6	2.818 +9
24 hours	2.8 +9	1.24 +7	5.42 +6	2.818 +9
3 days	2.8 +9	1.26 +7	5.43 +6	2.818 +9
5 days	2.8 +9	1.27 +7	5.43 +6	2.818 +9
10 days	2.8 +9	1.29 +7	5.42 +6	2.818 +9
20 days	2.8 +9	1.29 +7	5.40 +6	2.818 +9
30 days	2.8 +9	1.29 +7	5.38 +6	2.818 +9

^a e.g. $^{240}\text{Pu} + \alpha \rightarrow ^{236}\text{U}$, and subsequent $^{236}\text{U} \rightarrow ^{8}\text{O} + \alpha + ^{21}\text{Ne} + \text{n}$.

TABLE VIIa. Total Neutron Flux Levels ($n/cm^2\text{-sec}$) $k_{\text{shutdown}} = 0.99$

Time	Startup, Spontaneous Fission Plus $\alpha\text{-n}$ Sources	Residual Full Power ρ Plus Delayed Neutrons	Total ^a
Shutdown	3.960 +5	2.790 +14	2.790 +14
0.5 sec	3.960 +5	1.826 +14	1.826 +14
1 sec	3.960 +5	1.292 +14	1.292 +14
5 sec	3.960 +5	6.836 +13	6.836 +13
30 sec	3.960 +5	2.430 +13	2.430 +13
1 min	3.960 +5	1.175 +13	1.175 +13
2 min	3.960 +5	3.467 +12	3.467 +12
5 min	3.960 +5	2.253 +11	2.253 +11
10 min	3.960 +5	5.630 +9	5.630 +9
15 min	3.960 +5	1.495 +8	1.499 +8
20 min	3.960 +5	3.975 +6	4.371 +6
25 min	3.960 +5	1.057 +5	5.017 +5
30 min	3.960 +5	2.809 +3	3.988 +5
35 min	3.960 +5	74.683	3.961 +5
40 min	3.960 +5	1.985	3.960 +5
45 min	3.960 +5	0.053	3.960 +5
1 hour	3.960 +5	-	3.960 +5
2 hours	3.960 +5	-	3.960 +5
10 hours	3.960 +5	-	3.960 +5
1 day	3.960 +5	-	3.960 +5
3 days	3.961 +5	-	3.961 +5
5 days	3.961 +5	-	3.961 +5
10 days	3.961 +5	-	3.961 +5
20 days	3.961 +5	-	3.961 +5
30 days	3.961 +5	-	3.961 +5

^aApproximate thermal ($E < 0.625$ eV) flux can be obtained by taking 17% of this total value: see Table V and section III.

TABLE VIII. Total Neutron Flux Levels (n/cm^2 -sec) $K_{shutdown} = 0.98$

Time	Startup, Spontaneous Fission Plus ^{235}U Source	Residual Full Power : Plus Delayed Neutrons	Total ^a
Shutdown	7.600 \pm	2.790 ± 14	2.790 ± 14
0.5 sec	7.600 \pm	1.348 ± 14	1.348 ± 14
1 sec	7.600 \pm	8.260 ± 13	8.260 ± 13
5 sec	7.600 \pm	3.745 ± 13	3.745 ± 13
30 sec	7.600 \pm	1.173 ± 13	1.173 ± 13
1 min	7.600 \pm	5.365 ± 12	5.365 ± 12
2 min	7.600 \pm	1.469 ± 12	1.469 ± 12
5 min	7.600 \pm	9.225 ± 10	9.225 ± 10
10 min	7.600 \pm	2.233 ± 9	2.233 ± 9
15 min	7.600 \pm	5.648 ± 7	5.656 ± 7
20 min	7.600 \pm	1.429 ± 7	1.437 ± 7
25 min	7.600 \pm	3.617 \pm	1.122 ± 5
30 min	7.600 \pm	915.5	7.692 \pm
35 min	7.600 \pm	23.167	7.602 \pm
40 min	7.600 \pm	0.586	7.600 \pm
45 min	7.600 \pm	0.015	7.600 \pm
1 hour	7.600 \pm	-	7.600 \pm
2 hours	7.600 \pm	-	7.600 \pm
10 hours	7.600 \pm	-	7.600 \pm
1 day	7.600 \pm	-	7.600 \pm
3 days	7.601 \pm	-	7.601 \pm
5 days	7.601 \pm	-	7.601 \pm
10 days	7.602 \pm	-	7.602 \pm
20 days	7.602 \pm	-	7.602 \pm
30 days	7.602 \pm	-	7.602 \pm

^aApproximate thermal ($E < 0.625$ eV) flux can be obtained by taking 17% of this total value: see Table V and section III.

TABLE VIII. Approximate Neutron Energy Spectrum

Neutron Flux Spectra (Neutrons/cm ² -sec)				
Energy Group	Energy Range	Normalized to 100	Full Power Values	15 Minutes After Shutdown (k _s = 0.98)
1	0.821-10 MeV	23.6	6.58 +13	1.33 +7
2	5.53-821 keV	32.7	9.14 +13	1.85 +7
3	1.855-553 eV	23.9	6.68 +13	1.35 +7
4	0.625-1.855 eV	2.8	7.70 +12	1.58 +6
5	10 ⁻⁵ -0.625 eV	17.0	4.74 +13	9.60 +6
Totals		100	2.79 +14	5.65 +7

Neutron Density Spectra

Energy Group	Lethargy Range [u = ln(10 MeV/E)]	\bar{u}	E (\bar{u})	\bar{v} [E(\bar{u})] (cm/sec)	Full Power Density		\bar{n} Normalized to 100
					$\bar{n} = \frac{\rho}{\bar{v}}$ (Neutron/cm ³)		
1	2.5-0	1.25	2.865 MeV	2.370 +9	2.776 +4		0.012
2	9.803-2.5	6.152	21.303 keV	2.043 +8	4.474 +5		0.195
3	15.5-9.803	12.652	32.017 eV	7.923 +6	8.431 +6		3.671
4	16.588-15.5	16.044	1.077 eV	1.453 +6	5.299 +6		2.308
5	27.631-16.588	22.120 (19.81) ^a	0.025 eV ^a	2.20 +5 ^a	2.154 +8 ^a		93.814 ^a

$$\bar{v} \text{ (n weighted)} = 1.2153 \times 10^6 \text{ cm/sec}$$

^aAssuming a Maxwellian distribution in the thermal group.

APPENDIX B

ESTIMATE OF THE OXYGEN CONTENTS OF THE BUBBLE

1) Total oxygen production

a) Radiolysis under partial boiling conditions

Table 1

Time Interval After Shut Down (Hours)	Avg. Dose Rate (10^{20} eV L ⁻¹ s ⁻¹)	Total Dose (10^{24} eV L ⁻¹)	Efficiency Factor ^a	Effective Dose (10^{20} eV L ⁻¹)
0-1	12.48	4.49	0.8	3.59
1-2	7.42	2.67	0.7	1.87
2-3	6.24	2.25	0.35	0.79
3-4	5.62	2.02	0.3	0.61
4-8	4.71	6.78	1	6.78
8-14	3.81	8.23	1	8.23
14-16	3.46	2.49	0.8	1.99
			TOTAL	23.86

^aReflects partial covering of core and/or incomplete boiling. Values are considered conservative.

Maximum O₂ produced by radiolysis:

$$\frac{2.386 \times 10^{25} \times 0.225}{100 \times 6.02 \times 10^{23}} = 0.089 \text{ mol L}^{-1}$$

$$\text{or } 0.089 \times 17900 = 1593 \text{ mol O}_2 \text{ (total)}$$

b) O₂ added with feed water

Air saturation assumed (2.33×10^{-4} mol L⁻¹ O₂).

234,000 gal = 1.075×10^6 L H₂O contain 250.5 mol O₂

Total O₂ produced or added (worst case): 1343 mol O₂

(2) Total hydrogen production:

burned in Containment: 226 lb. mol = 10.25×10^4 mol

Still in Containment: 80 lb. mol = 3.63×10^4 mol

in Bubble and Solution* 8.87×10^4 mol

Total H₂ Produced 22.75×10^4 mol

c) Steam produced

284,000 gal. H₂O produce 1.075×10^6 kg

= 5.97×10^4 mol steam

(3) Steam and H₂ in bubble

Volume of Bubble: $1471 \text{ ft}^3 = 41,630 \text{ L}$

Temperature: $250^\circ\text{F} = 138^\circ\text{C} = 411 \text{ K}$

Pressure: 790 psig = 67 atm

Partial pressure of steam
and bubble: = 3.3 atm

Partial Pressure of H₂ = 63.7 atm
(O₂ neglected)

H₂ in Bubble: $\frac{63.7 \times 41630 \times 273}{22.4 \times 411} = 7.86 \times 10^4$ mol

Steam in Bubble 4135 mol

*See calculation below

(4) Contents of solution

Volume of H₂O: 219,000 L - 41,630 L = 177,400 L

Distribution of gas between bubble and solution:

$$\frac{H_2 \text{ (bubble)}}{H_2 \text{ (sol)}} = \frac{41,630}{177,400 \times 0.02} \cdot \frac{273}{411} = 7.79$$

$$\frac{O_2 \text{ (bubble)}}{O_2 \text{ (sol)}} = \frac{41,630}{177,400 \times 0.013} \cdot \frac{273}{411} = 12.0$$

Therefore H₂ in solution:

$$7.86 \times 10^4 / 7.79 = 1.01 \times 10^4 \text{ mol H}_2$$

(5) Fraction of H₂ and steam not vented

Table 2

	<u>Amount Produced (mol)</u>	<u>Amount in Bubble (and solution) (mol)</u>
H ₂	2.27 x 10 ⁵	8.87 x 10 ⁴
Steam	0.60 x 10 ⁵	0.41 x 10 ⁴
	2.87 x 10 ⁵	9.28 x 10 ⁴

Fraction of H₂ + steam retained in RCS: 0.323.

Assuming that the same fraction of O₂ was retained,

this amounts to 0.323 x 1843 = 595 mol O₂

Therefore

O₂ in bubble: 549 mol

O₂ in solution: 46 mol

O₂/H₂ in bubble: 0.73

Concentrations in solution

$$H_2: 1.01 \times 10^4 / 177,400 = 5.7 \times 10^{-2} \text{ mol L}^{-1}$$

$$O_2: 46 / 177,400 = 2.6 \times 10^{-4} \text{ mol L}^{-1}$$

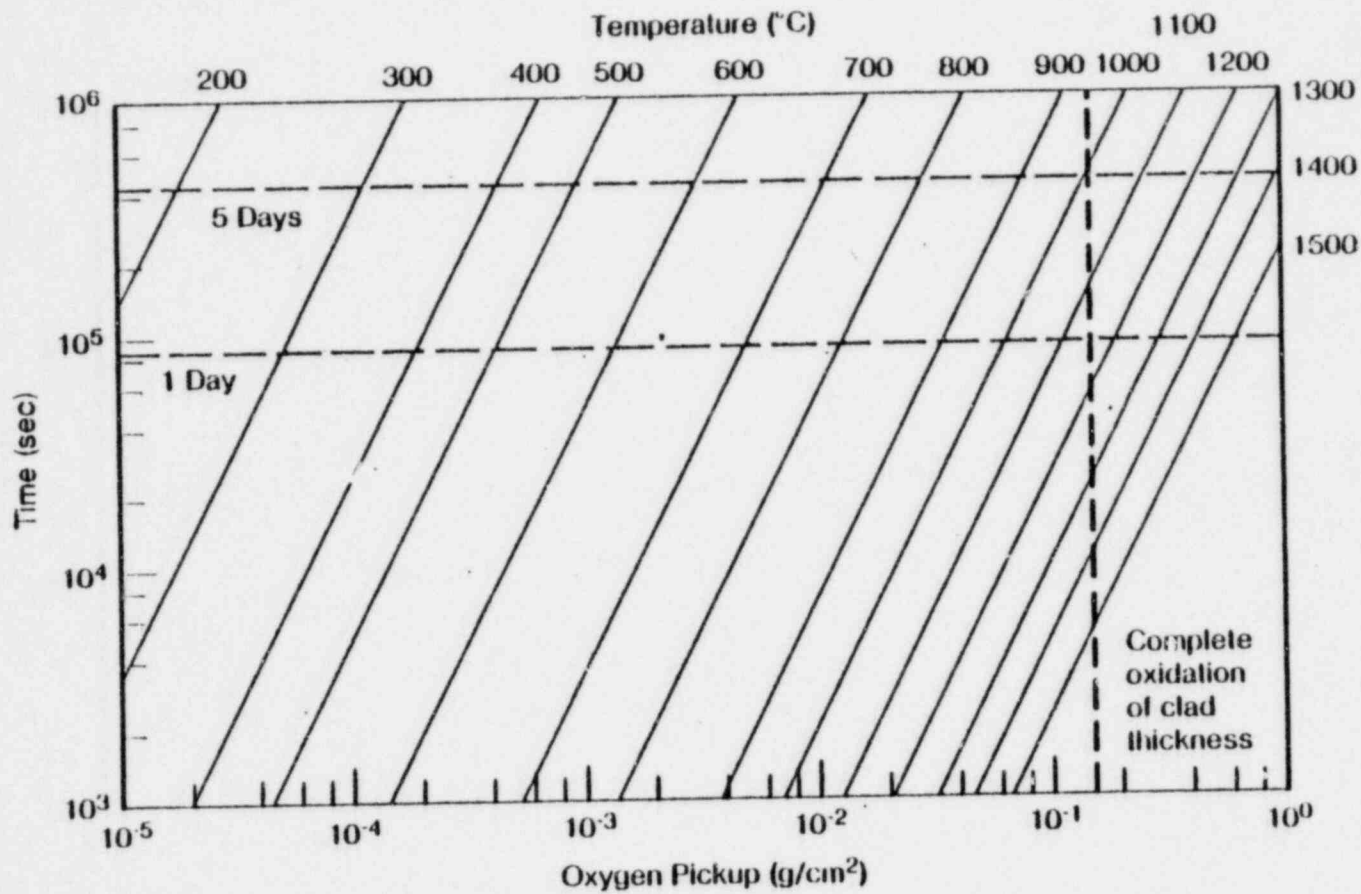


FIGURE 1

FIGURE 2

(O₂) in Core Water Vs. Time
Full Power Conditions
pH = 6

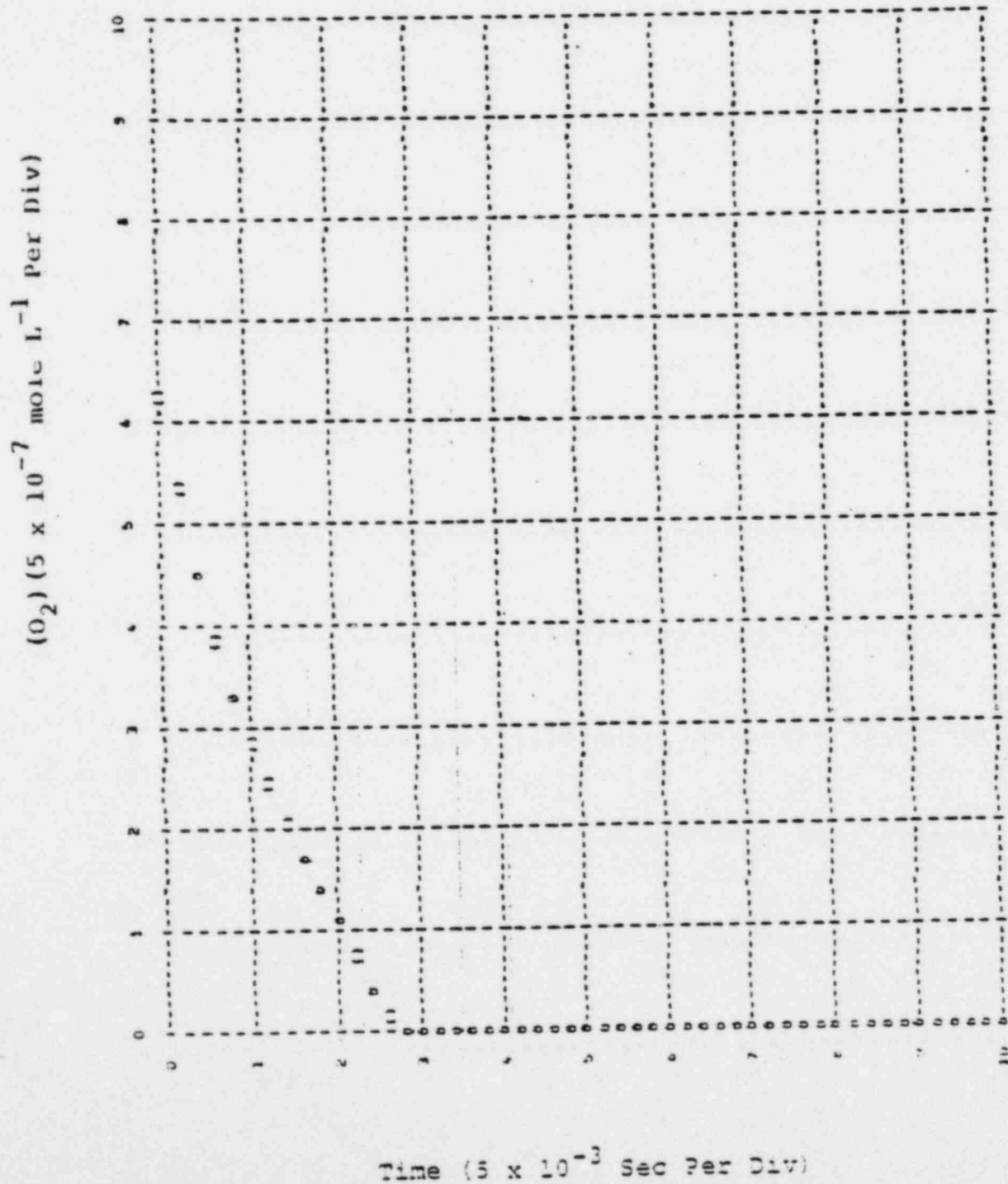


FIGURE 3

(H₂O₂) in Core Water Vs. Time
Full Power Conditions
pH = 6

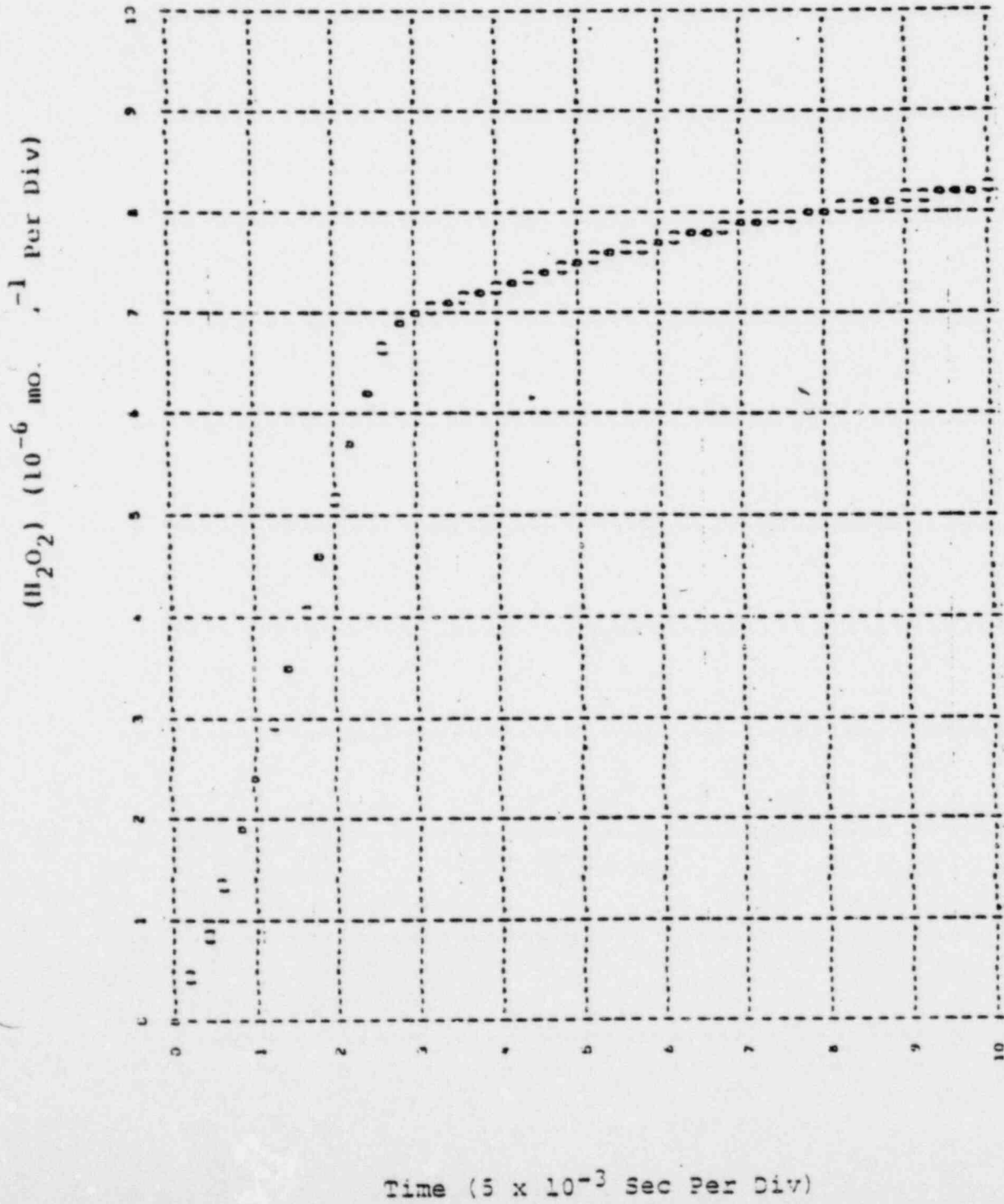
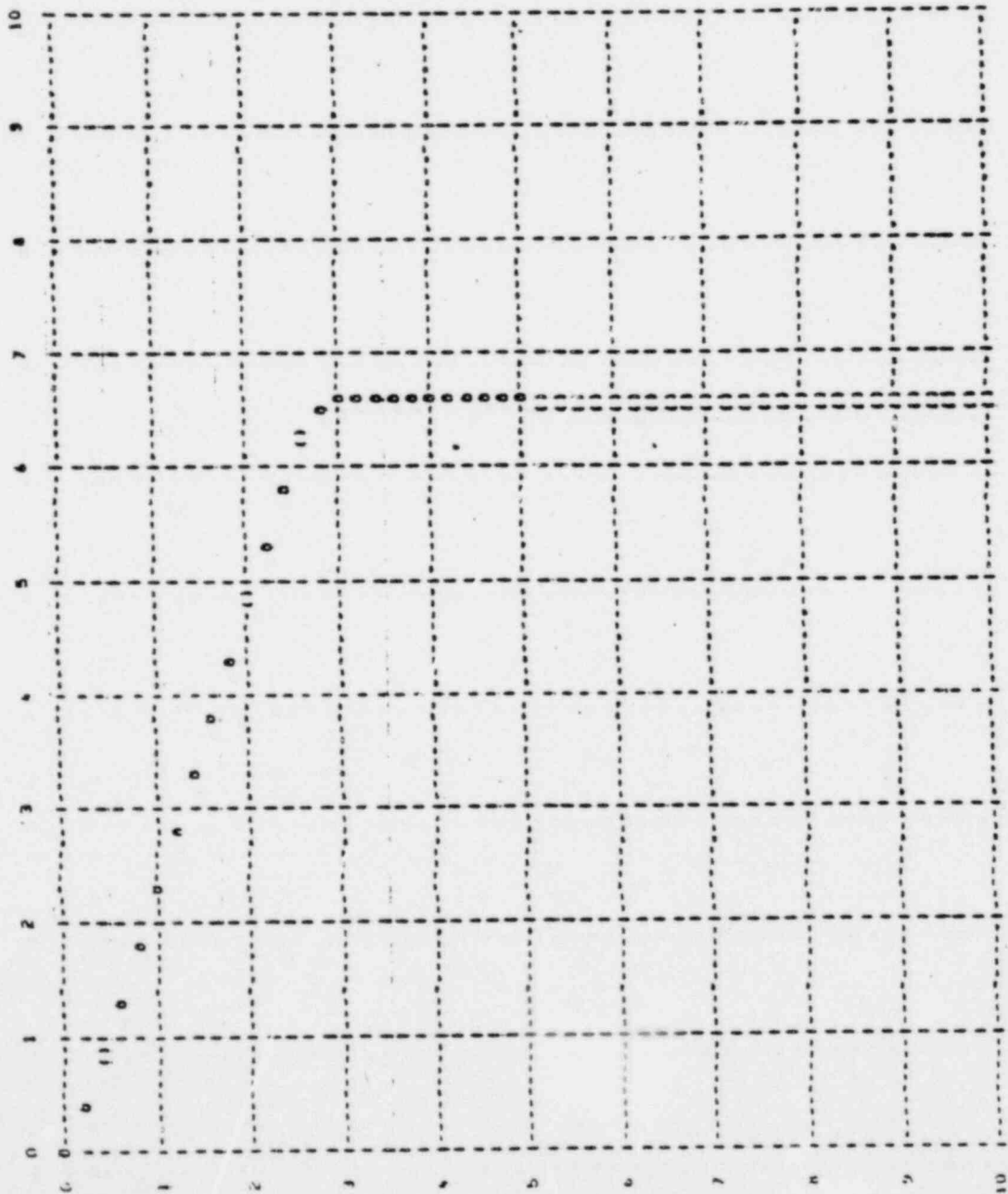


FIGURE 4

(H₂O₂) in Core Water Vs. Time
Full Power Conditions
pH = 8

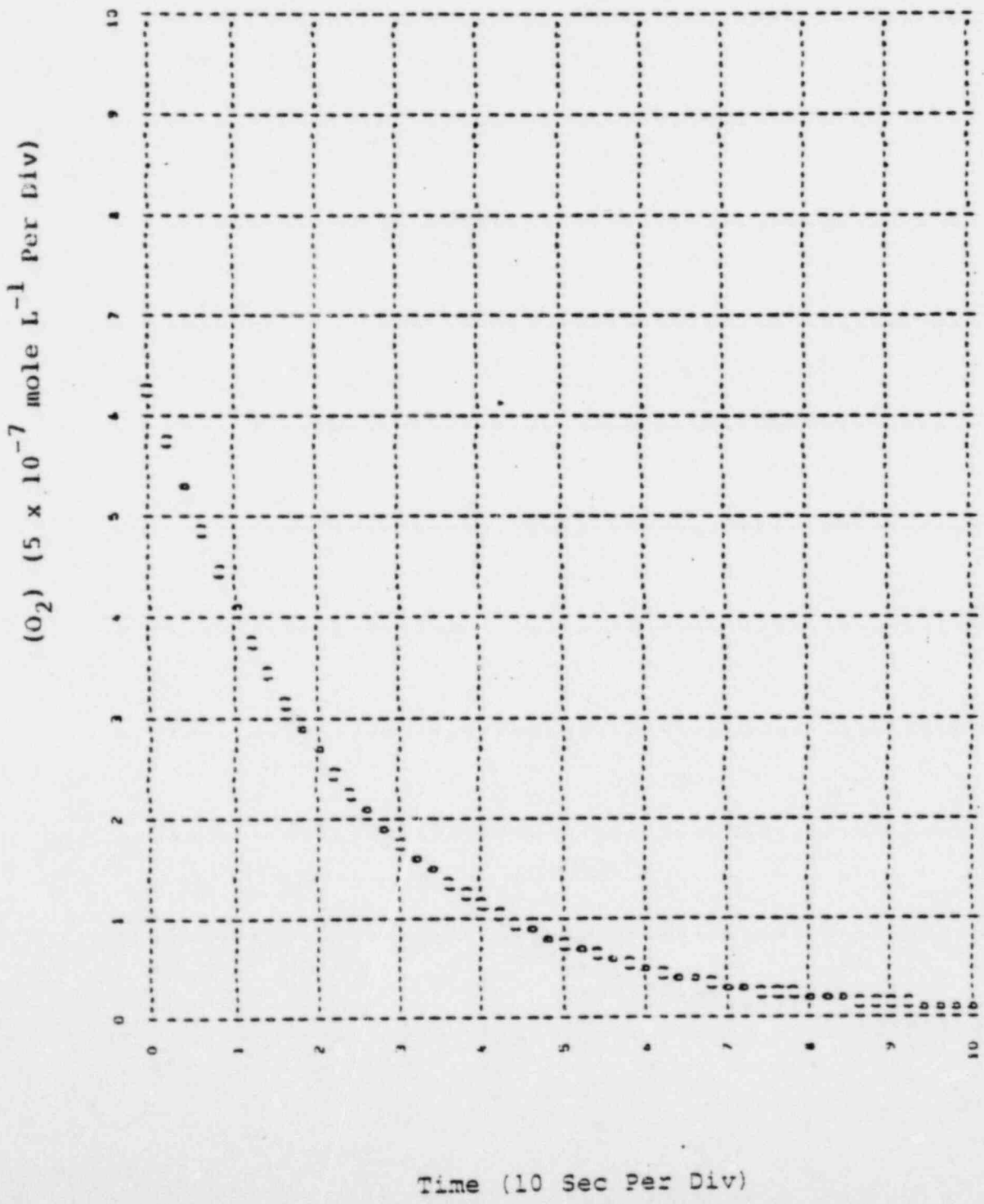
(H₂O₂) (10⁻⁶ mol/L 1 Per Div)



Time (5 x 10⁻³ Sec per Div)

FIGURE 5

(O₂) in Cooling Loops Vs. Time
Full Power Conditions
pH = 6



(H₂O₂) in Cooling Loops Vs. Time
at Power Conditions
PH = 6

(H₂O₂) (10⁻⁶ mole L⁻¹ Per Div)

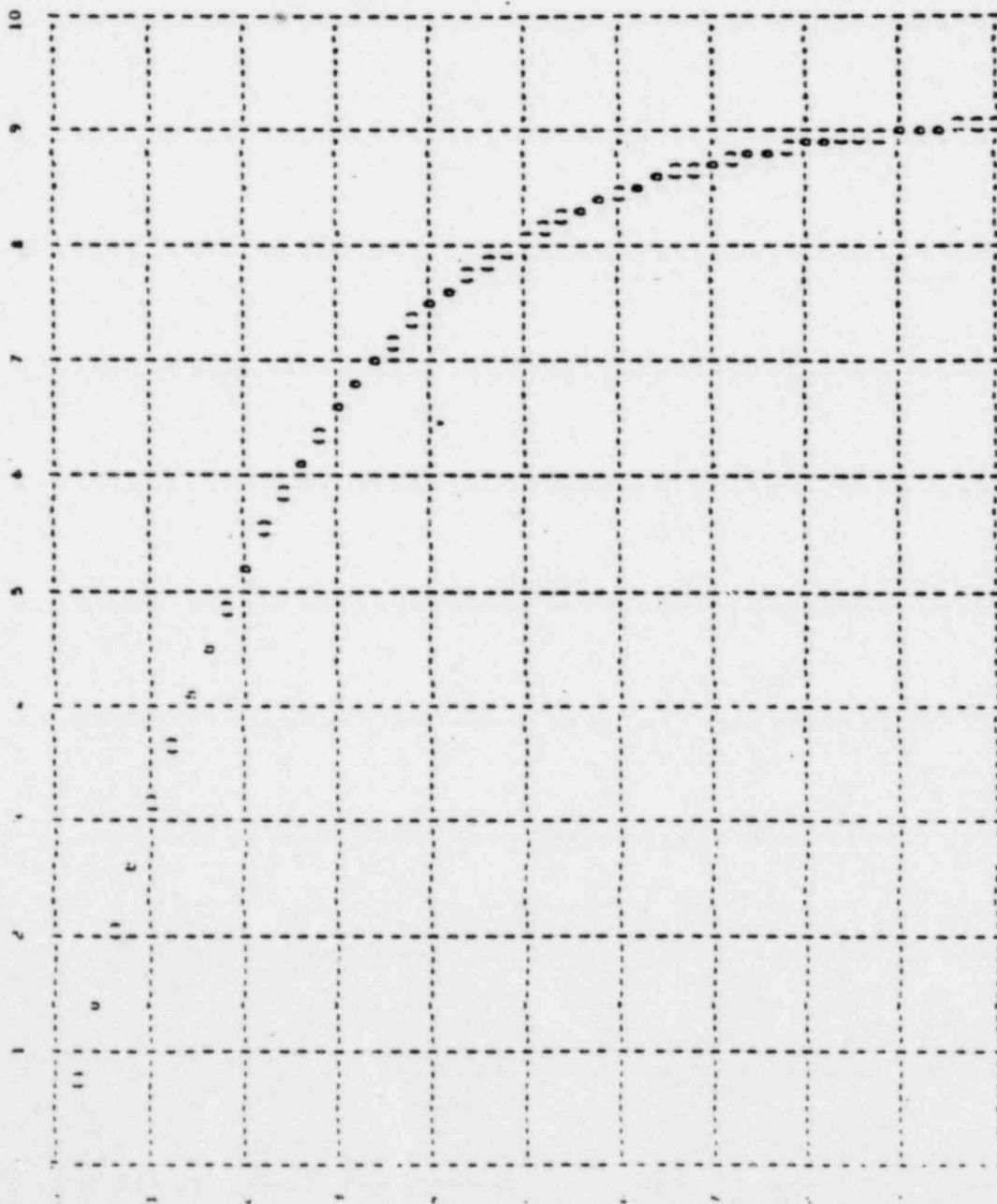


FIGURE 7

(H_2O_2) in Core Water Vs. Time
Full Power Conditions
pH = 3

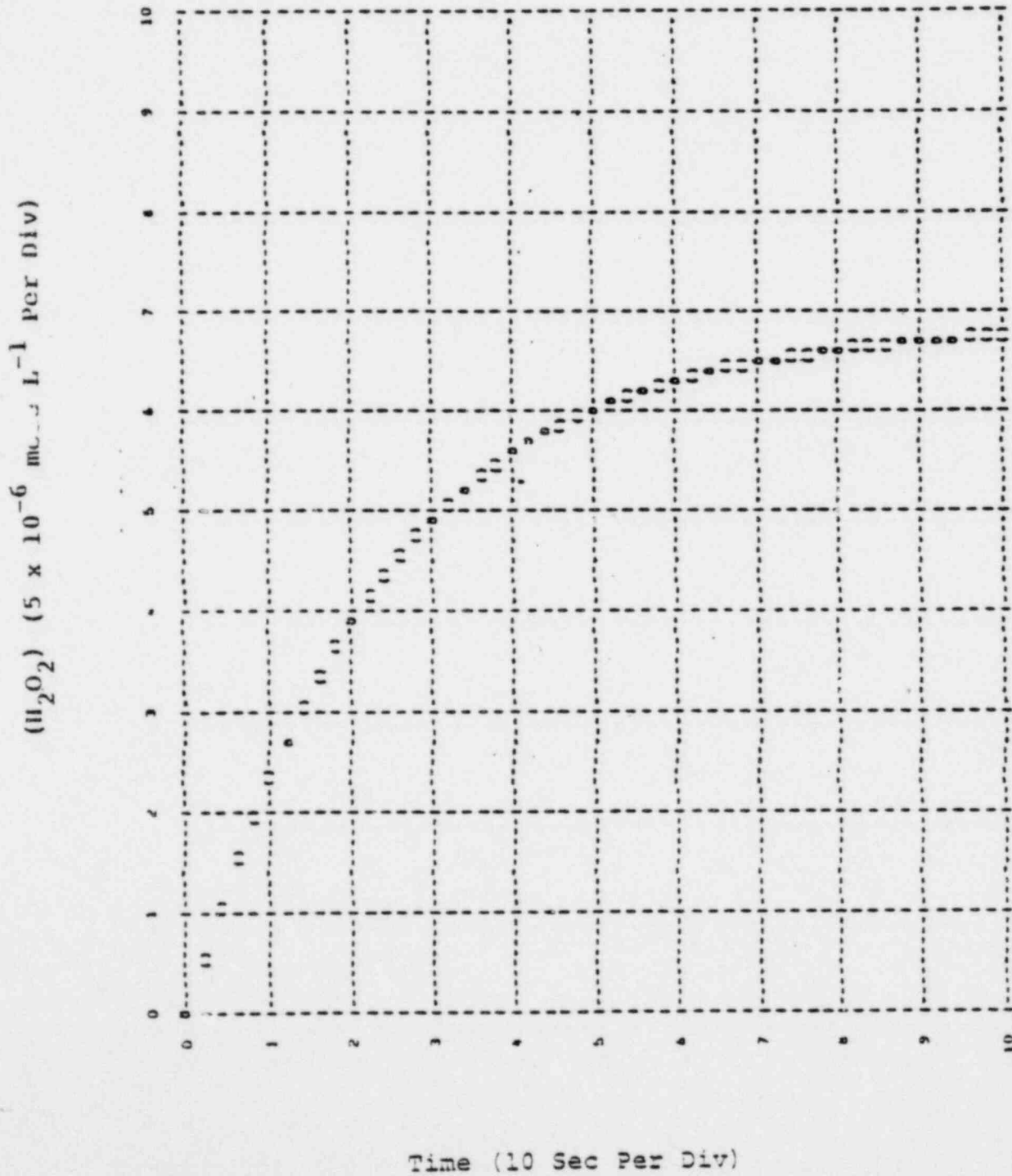


FIGURE 3

(O₂) in Core Water Vs. Time
10 Hours After Shut Down
of # 6

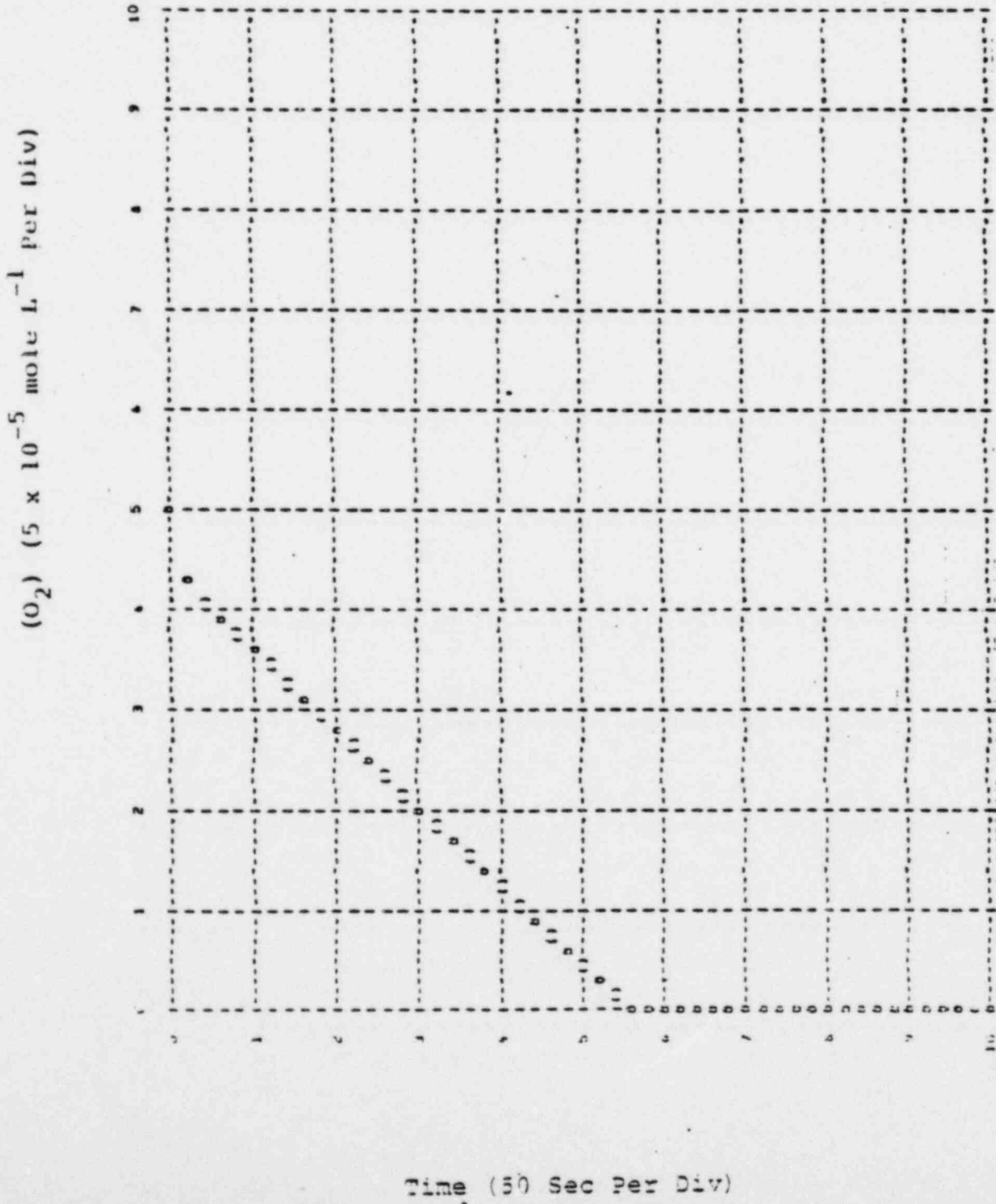


FIGURE 9

(H₂O₂) in Core Water Vs. Time
15 Hours After Shut Down
pH = 6

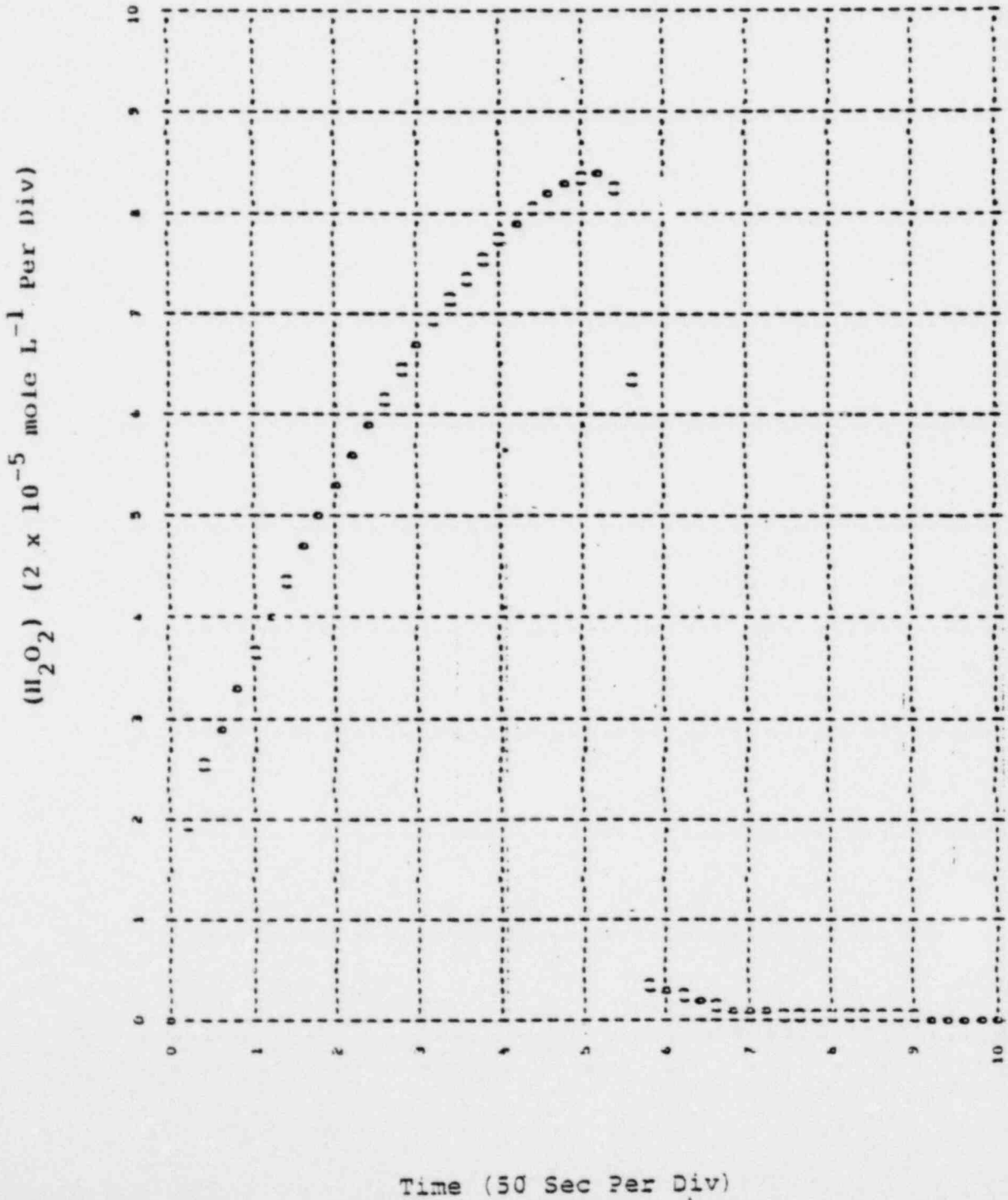


FIGURE 10

(H₂) in Core Water Vs. Time
10 Hours After Shut Down
pH = 6

