

MLP

~~SECRET~~

Sandia Laboratories

Albuquerque, New Mexico 87115

8005140366

October 15, 1979

Dr. M. Picklesimer
NRC/TMI/SIG Rm. 412
6935 Arlington Road
Bethesda, MD 20014

Dear Dr. Picklesimer:

Enclosed are the results of our TMI Bubble Analysis investigation. The stated objectives of the project were:

1. Determine the quantities of non-condensable gases formed and how they were formed.
2. Determine the time distribution of non-condensable gases in the primary system.
3. Determine distribution of gas between reactor primary system and containment.

Our conclusions, as previously reported by telephone, follow. Several aspects are expanded on in the attachments.

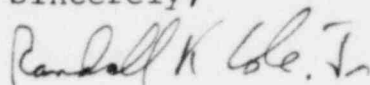
Our best estimate (based on previous work and a new computer calculation) is that approximately 450 kg of hydrogen was produced, essentially all by oxidation of zircaloy fuel cladding. A small amount (less than 10 kg based on rates "Core-Meltdown Experimental Review" SAND74-0382, August 1975) could have been produced by oxidation of stainless steel. We concur in the general conclusion that radiolysis was not important and that there was never any significant amount of free oxygen in the primary system.

Of the total hydrogen, we find that approximately 300 kg was released to containment before 9 hrs., with the remaining approximately 150 kg removed over the next several days. These results, from the computer simulation, are reasonably well supported by a back calculation from the hydrogen burn at 9.9 hrs., and by extrapolation of bubble sizes measured later.

The chemical results are inconclusive. Radionuclide concentrations in the primary system are consistent with the calculated core damage, if it is assumed that a significant fraction of the 270,000 gal of water from the BWST somehow did not pass through the core. It appears that leaching is as likely a method as volatilization for removal of some of the fission products from the fuel, so that one need not infer extended periods of extremely high temperature. We were unable to find any satisfactory explanation of measured boron concentrations.

Some of this work, in particular, the computer simulation and the bubble calculation, will be expanded during the next several weeks. New results will be reported as they become available.

Sincerely,



Randall K. Cole, Jr.
Reactor Safety Studies
Division 4441

Enclosure: 1) Computer Simulation
2) The Hydrogen Bubble
3) Hydrogen Burns

A computer code was developed, primarily by Michael I Baskes of Sandia's Livermore Lab, to provide a simplified model of the TMI2 reactor coolant system. The code uses all available data including pressure, temperatures, pump flows, and valve positions as input. It calculates core response and hydrogen production, and water and hydrogen inventories in various components including the reactor vessel, hot and cold legs, steam generators, and the pressurizer.

Both mass and energy conservation are considered for the reactor vessel. Core physics was initially treated much as described in NUREG/CR-0913*, including finite heat transfer coefficients between fuel and liquid and between fuel and steam, but neglecting the radial power profile. For all other components, mass alone is conserved. Flow through the PORV is evaluated using HEM critical flow and an effective area of 1.09 in^{2**} .

Calculations with the original core model predicted hydrogen generation shortly after $2\frac{1}{2}$ hrs at such a rate that the calculated partial pressure of hydrogen exceeded the measured total pressure. The model was therefore modified to consider the flow of molten zirconium clad down the rods to cooler regions of the core, delaying its oxidation sufficiently to eliminate the contradiction. The relative ease with which

← ZrO₂ reacts with UO₂ to dissolve UO₂ liquid phase

* R.K.Cole, Jr., "Generation of Hydrogen during the First Three Hours of the Three Mile Island Accident", NUREG/CR-0913, SAND79-1357, Sandia Laboratories, Albuquerque, NM, July 1979.

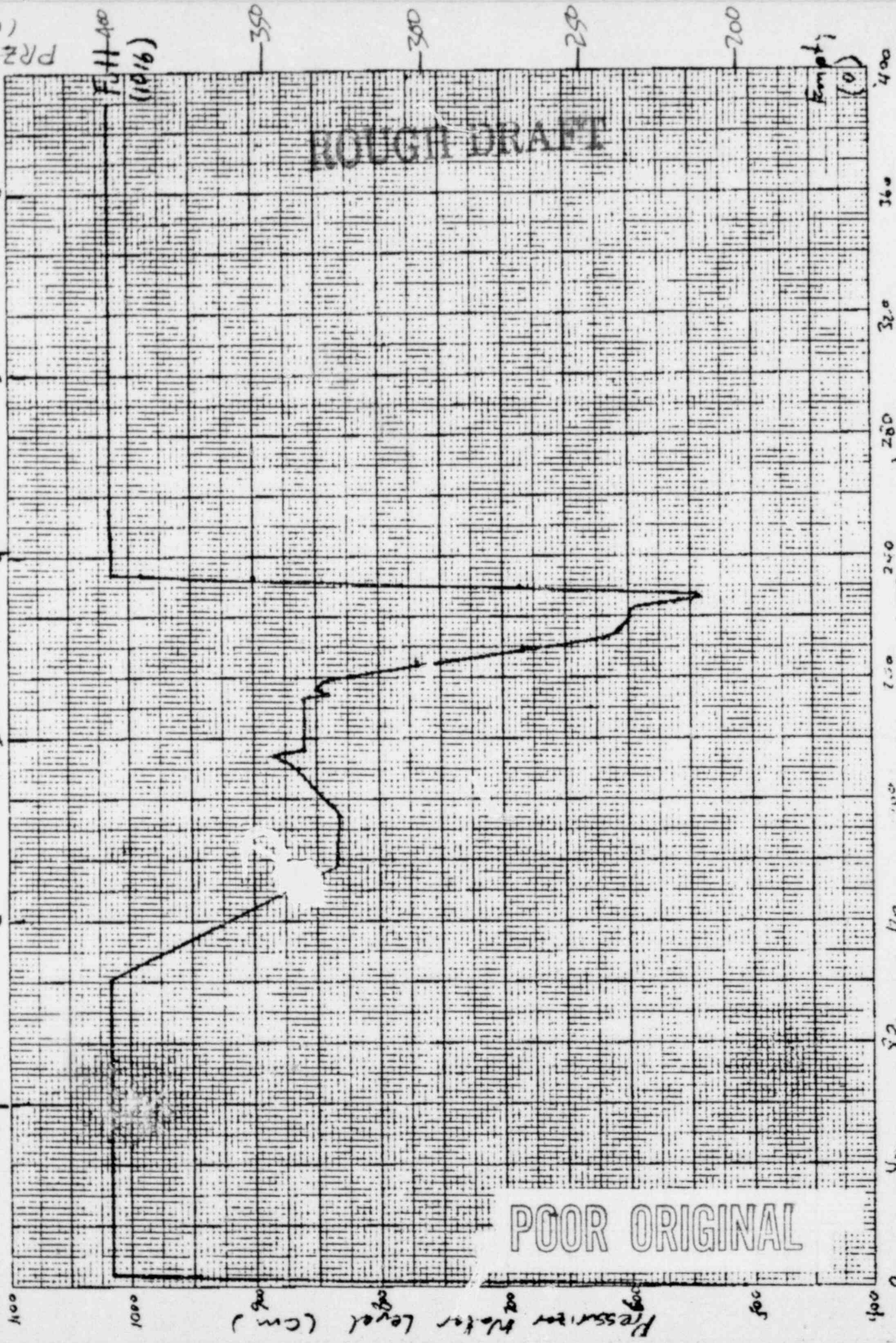
**Reasonable results could not be obtained using the 0.87 in^2 effective area of NUREG/CR-0913.

this modification could be made results from the basic structure of the code. The various relationships to be modelled are represented by a set \dot{x} of ordinary differential equations (the current version employs more than 100) and solved by a general purpose library routine. This routine controls the time step by monitoring its own accuracy, and will terminate (with appropriate messages) if severe difficulties are encountered.

~~P~~reliminary results for the first $6\frac{1}{2}$ hours of the accident are shown in Figs 1-3. We anticipate that during the depressurization between about $7\frac{1}{2}$ and 9 hours approximately one half of the hydrogen remaining in the reactor vessel and steam generator B will be lost, leaving about 150 kg in the primary system and about 300 kg in containment. These figures are consistent with back calculations based on later bubble sizes and on the hydrogen burn just before 10 hours.

FIG 1, PRESSURIZER LEVEL

TIME (HOURS AFTER TRIP)



ROUGH DRAFT

POOR ORIGINAL

Full (1016)

Empty (0)

PRZR level (m)

400 360 320 280 240 200 160 120 80 40 0

1400 1000 200 300 250 200

POOR ORIGINAL

ft above bottom of core

12 11 10 9 8 7 6 5

400

360

320

280

240

200

160

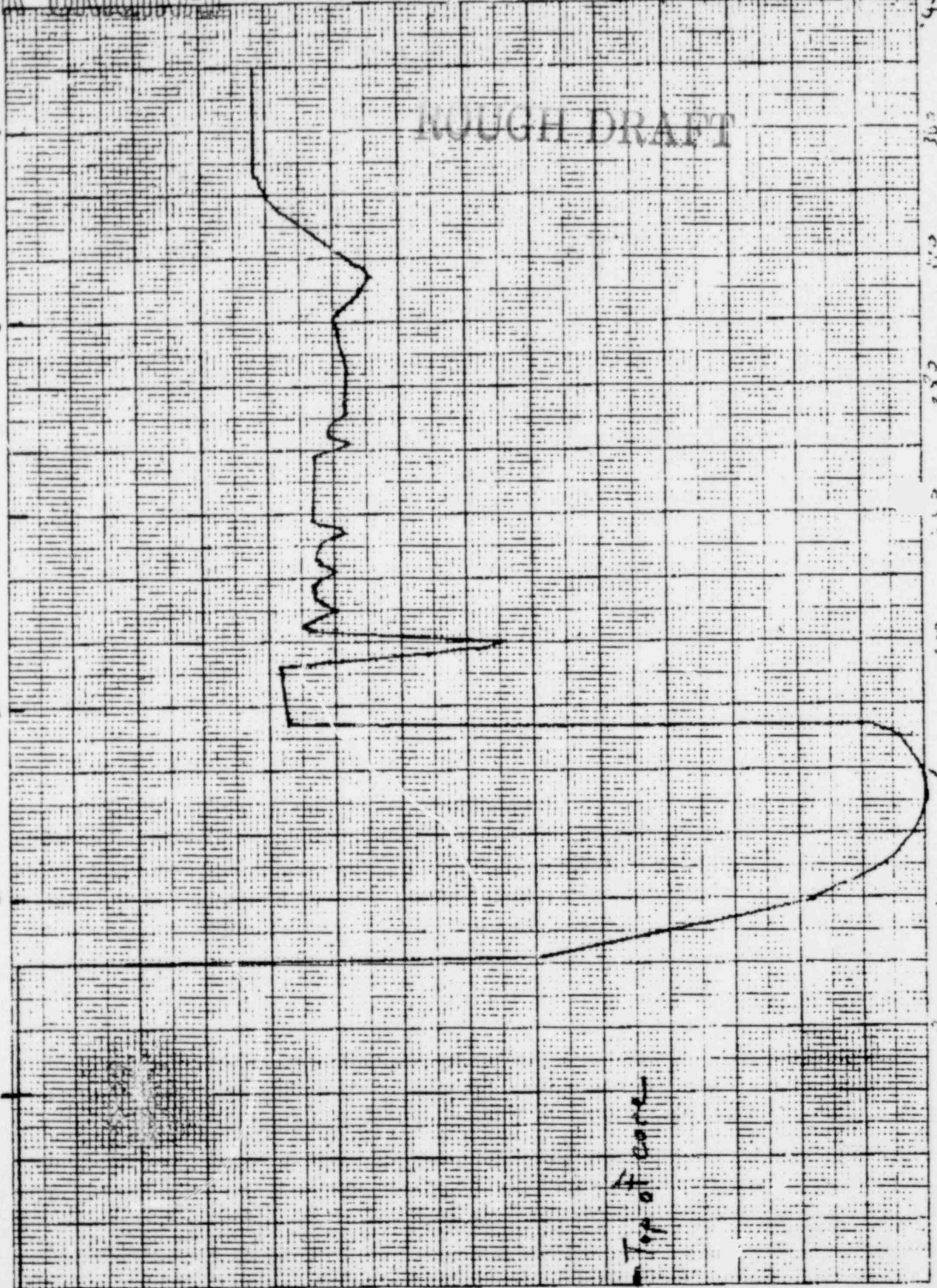
120

80

40

0

ROUGH DRAFT



Top of core

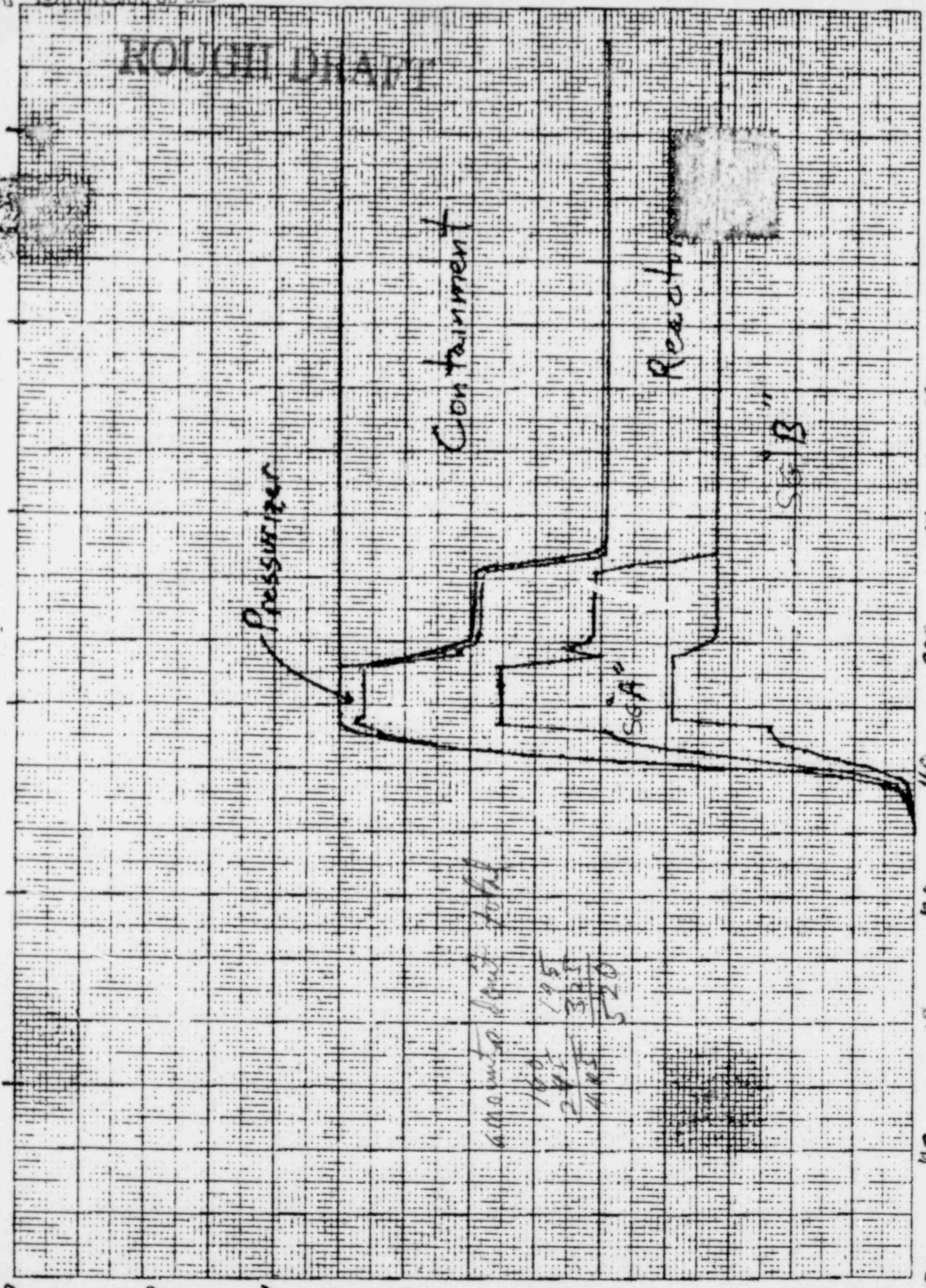
cm above core bottom

FIG 2 MIXTURE LEVEL IN REACTOR TIME (HOURS AFTER START)

ADDITIONAL INFORMATION

ROUGH DRAFT

FIG 3 HYDROGEN INVENTORY TIME (HOURS AFTER TRIP)



450
400
360
330
280
240
200
160
120
80
40
0

1
2
3
4
5

10⁵ gm
H Inventory
M
L
g

During the period from 29 March through at least 8 April measurements were made to determine the size of the (assumed) hydrogen bubble in the TMI2 reactor coolant system (RCS). The procedure was to define a mass balance for the RCS (exclusive of pressurizer) by recording changes in the levels of the ~~pressurizer~~ pressurizer (PZR) and makeup tank (MUT). From this, the change in bubble size (and therefore the apparent compressibility) could be calculated and the bubble size inferred.

We have independently derived a "bubble formula", compared it with the Met Ed and B&W formulae, and used it to reduce the raw data in the "Bubble Book". The total hydrogen content of the RCS, including hydrogen in solution, was then fit ~~by~~ by a ~~xxx~~ straight line (as a function of time) to estimate the average removal rate.

THE BUBBLE FORMULA

We assume that the bubble is a mixture of hydrogen gas and water vapor. Because of the low temperature and water vapor pressure (280°F, 50psia) Dalton's Law should apply. The ~~xx~~ change in water vapor ^{pressure} due to the partial pressure of hydrogen*

$$P_v - P_{sat}(T) \approx (P - P_{sat}(T)) \frac{P_{v sat}}{P_{l sat}} \quad (1)$$

is negligibly small--about 2 psi at a total pressure of 1000psia. The partial density of water vapor in the bubble is essentially the saturation density $\rho_{v sat}(T)$.

The total water mass in the RCS is given by

* P.M. Morse, Thermal Physics, W A Benjamin Inc, 1969, pl24.

$$M_w^{RCS} = (V^{RCS} - V^B) \rho_e (P^{RCS}, T^{RCS}) + V^B \rho_{vsat} (T^{RCS}). \quad (2)$$

If Eq 2 is evaluated for two (P, T) states and the results subtracted, one finds

$$(\rho_e - \rho_v)_2 V_2^B - (\rho_e - \rho_v)_1 V_1^B = (V \rho_e - M_w)_2^{RCS} - (V \rho_e - M_w)_1^{RCS}. \quad (3)$$

With the general notation (f is any quantity)

$$\bar{f} \equiv (f_1 + f_2) / 2 \quad (4)$$

$$\Delta f \equiv f_2 - f_1 \quad (5)$$

Eq 3 becomes

$$(\bar{\rho}_e - \bar{\rho}_v) \Delta V^B + \bar{V}^B \Delta (\rho_e - \rho_v)^{RCS} = \bar{V}^{RCS} \Delta \rho_e^{RCS} + \bar{\rho}_e^{RCS} \Delta V^{RCS} - \Delta M_w^{RCS} \quad (6)$$

This equation (which contains no approximations) may be simplified by elimination of several small terms.* At 1000psia and 280°F, $\rho_e \approx 58 \text{ lb}_m/\text{ft}^3$ and $\rho_{vsat} \approx .12 \text{ lb}_m/\text{ft}^3$. Also, $V^{RCS} \approx 10300 \text{ ft}^3$ and (we will find) V^B is typically several hundred cubic feet while ΔM_w^{RCS} is typically a few thousand pounds for a pressure change of 100 psi. The expansion of a cylindrical vessel is given by

$$\Delta V \approx 2 \frac{R}{E} \frac{\Delta P}{E} V \quad (7)$$

where R/t is the ratio of radius to thickness, about 8 for the RCS, and E is Young's modulus, roughly 3×10^7 psi for steel. Thus $\bar{\rho}_e \Delta V^{RCS}$ is typically 30 lb_m , a 1% effect. Temperature changes were almost never greater than 1°F. Therefore, from Steam Tables, we find $\Delta \rho_e \approx .05 \text{ lb}_m/\text{ft}^3$ and $\Delta \rho_v \approx .002 \text{ lb}_m/\text{ft}^3$. Thus $\bar{V}^B \Delta (\rho_e - \rho_v)^{RCS}$ is a few tens of pounds, again a 1% effect, while $V^{RCS} \Delta \rho_e^{RCS}$ may be several hundred pounds and is ^{more} significant. Finally, neglecting ρ_v compared to ρ_e we find

*The reader may skip to Eq 8, an obvious approximation, and miss only a discussion of the accuracy of that approximation.

V^{RCS} does not contain V^B

$$\Delta V^B \approx (V^{RCS} \Delta \rho_e^{RCS} - \Delta M_w^{RCS}) / \bar{\rho}_e^{RCS}, \quad (8)$$

accurate to a few percent.

The mass increase in the RCS is simply the net mass loss from makeup tank and pressurizer, reduced by net leakage. Because the pressure and temperature in the makeup tank are essentially constant the change in its density may be neglected, but this is not the case for the pressurizer. The resulting expression is

$$\Delta M_w^{RCS} = - (\rho_L A \Delta L)^{MUT} - \Delta (\rho_L V_L + \rho_U V_U)^{PZR} - \Delta M_w^{leak} \quad (9)$$

Here

$$V_L^{PZR} = [A(L+L_0)]^{PZR} \quad (10)$$

$$V_U^{PZR} = V^{PZR} - V_L^{PZR}, \quad (11)$$

where the A's are horizontal cross-sectional areas, L's are levels, L_0 is the effective height of the hemispherical section of the pressurizer below the lower sensing nozzle (2/3 the radius or 28 in) and ΔM_w^{leak} is the unknown leakage term.

The hydrogen content of the bubble is simply

$$m_H^B = P_H V^B / RT \quad (12)$$

moles where R is the gas constant and

$$P_H = P - P_{sat}(T) \quad (13)$$

is the partial pressure of hydrogen. The solubility of hydrogen is proportional to its partial pressure and (neglecting the compressibility of the water) is given by $\mathcal{S}'(T) P_H$ moles per unit volume. The total hydrogen content of the RCS (exclusive of the pressurizer) is given by

$$n_H^{RCS} m_H^{RCS} = m_H^B + \mathcal{S}'(T) P_H (V^{RCS} - V^B) \quad (14)$$

assuming that a bubble is present. If n_H^B is eliminated from Eq 14 by using Eq 12, the resulting equation is

$$V^B = \max \left[\frac{m_H^{RCS} RT}{(1 - \mathcal{S}' RT) P_H} - \frac{\mathcal{S}' RT}{1 - \mathcal{S}' RT} V^{RCS}, 0 \right] \quad (15)$$

where the dimensionless quantity

$$S'_{RT} = S'_{P_H} / (P_H / RT) \quad (16)$$

is simply the ratio of the volumetric concentration of hydrogen in solution to that in the vapor phase, and is approximately 0.03 at 280°F*. Equation 15 explicitly includes the possibility that all hydrogen is in solution with no bubble present.

If measurements are made at two pressures but nearly equal temperatures so that changes in S' and T may be neglected

Eq 15 may be used to show that**

$$n_H^{RCS} = \frac{1 - S'_{RT}}{RT} \max \left[-P_{H_2} P_{H_1} \frac{\Delta V^B}{\Delta P_H}, P_{H \min} \left(|\Delta V^B| + \frac{S'_{RT}}{1 - S'_{RT}} V^{RCS} \right) \right] \quad (17)$$

where the Δ notation of Eq 5 has been used, and $P_{H \min}$ is the lesser hydrogen content of the system has been calculated from Eq 17,

Eq 15 may be used to calculate the size of the bubble at any pressure. If a bubble is present in both states 1 and 2, the first term in brackets in Eq 17 is the greater, and the bubble volume is given by

$$V^B = - \frac{P_{H_2} P_{H_1}}{P_H} \frac{\Delta V^B}{\Delta P_H} - \frac{S'_{RT}}{1 - S'_{RT}} V^{RCS} \quad (\text{Bubble at } P_{H_1}, P_{H_2}, P_H) \quad (18)$$

If a bubble is present only at the lower pressure, the second term in Eq 17 is the greater, and

$$V^B = \frac{P_{H \min} |\Delta V^B|}{P_H} + \frac{P_{H \min} - P_H}{P_H} \frac{S'_{RT}}{1 - S'_{RT}} V^{RCS} \quad (\text{Bubble at } P_{H \min}, P_H) \quad (19)$$

Finally, if no bubble is present in either state, Eq 15 yields an upper bound on hydrogen content given by the amount soluble at the lower pressure, and Eq 15 gives zero volume for any pressure about which we have knowledge. In practice, because of experimental errors, it may be difficult to distinguish the latter

*Calculated from H.A. Pray et al, "Solubility of Hydrogen, Oxygen, Nitrogen, and Helium in Water", Industrial and Engineering Chemistry 44 (5) 1146-1151, 1952.

**We assume that n_H^{RCS} is constant during the measurement. Changing hydrogen content could be included with minor changes in the following equations.

two cases, and it may be better to interpret n_H^{RCS} from Eq 17 as an upper bound whenever the second term dominates.

For the case where a bubble is present at both states 1 and 2 our ~~equation~~ bubble formula/ given by Eqs 8, 9, and 18. The B&W formula is in close agreement, although matching the solubility terms takes a little work. The main differences are B&W's neglect of changes in vapor mass in the pressurizer, and of liquid below the lower sensing nozzle (L_0 in Eq 10), which are offsetting effects of a few percent each. Also, they appear to use total pressure rather than hydrogen partial pressure in/Eq 18, which would lead to a 5% underprediction of bubble size.

The Met Ed formula also uses total pressure, and ~~also~~ ^{Further} neglects all compressibility and thermal expansion terms for the water. The most important of these ^{neglected} terms/is thermal expansion in the pressurizer under increasing saturation temperature, leading to a 10% underprediction of bubble size. The next largest term, Δp_e^{RCS} is dominated by temperature changes and therefore not consistent in sign. Much more important is the ~~complete~~ neglect of the solubility of hydrogen. The effect is shown by the last term of Eq 18 to be a systematic 300 ft³ overprediction of bubble volume ($\rho RT \approx .03$ and $V^{RCS} \approx 10300ft^3$). While the previously mentioned errors tend ~~to produce~~ toward underprediction, they are substantially smaller so that the ^{overprediction through the} neglect of solubility is the overriding ^{effect.} ~~error.~~ Finally, the ~~XXXXXXXXXX~~ data reduction in the Bubble Book contains a number of arithmetic and/or transcription errors.

$V^{RCS} = 11,800 ft^3$
 $V^{RCS} - V^{PZB} = 10,300 ft^3$

RESULTS

The bubble formula derived in the preceding section has been used to reduce the raw data presented in the Bubble Book. The ~~xx~~ RCS temperature and the makeup tank temperature were taken as 280°F and 80°F, respectively, when these data were not recorded. We observed that the pressurizer temperature and system pressure were not consistent in that the pressure was consistently 50 psi lower than $P_{\text{sat}}(T^{\text{PZR}})$. This discrepancy, in the wrong direction to be explained ~~xx~~ by a partial pressure of hydrogen, could be due to a 5°F error in T^{PZR} . However, we also noted that for the earliest data sets pressure was reported from the wide range recorder as well as from the computer, and is typically 50 psi higher. This problem has not been resolved. Therefore, in those cases where T^{PZR} was reported as well as P , the pressurizer mass term ~~xx~~ was evaluated twice, first using P to determine saturation densities and then ^{using} T , and the average used. In no case was the difference significant.

The results of these calculations* are presented in Figures 1-3. The first shows total mass while the second and third give bubble volumes at the average system pressure of 1000psia and at the MetEd-established standard 875psia. Also shown in these figures are ~~xx~~ generalized** least-squares fits to the data from 3/31/79 through 4/3/79 with an approximate one-standard-deviation confidence band on the fit, and a fit presented by B&W.***

*The mass-leak term was taken as zero. We intend to repeat the calculations using the value mentioned later in the text.

**The generalization involves points where the bubble was "almost gone", and the formula yields an upper bound.

***Memo from James H. Taylor to John Bickel, 20 July 1979. We consider only the data "With Solubility Correction".

We feel that the difference between the two fits may be due to B&W's apparent use of only 5 data points. The standard deviation of the points used in our fit from the fit line is ± 12 kg, in reasonable agreement with the error bars shown by B&W, corresponding to a mass error of ± 8 kg.

We find an average removal rate of $1.7(\pm 0.3)$ kg/hr. This corresponds to the complete degassing of $60(\pm 10)$ gpm of letdown flow. This is not meant to imply that all the hydrogen was removed through letdown, but merely to note that typical letdown rates are sufficient to remove most of it. For comparison, the B&W line implies a significantly larger removal rate of 3.0 kg/hr, again perhaps due to the small data set used.

Our fit suggests that the bubble was gone at 1000psia at 1800 on 4/1/79 (± 3 hrs). During this time period the pressure was being cycled between about 950 and 1050psia. The bubble would be eliminated at the higher pressure about an hour earlier.

If the constant removal rate is used to estimate the hydrogen content of the RCS at 16 hours--a very questionable extrapolation of inaccurate data--one finds a total mass of 190 (± 40) kg. At 1400 psia and 360°F there would be roughly 45 kg in solution and an 1100 ft^3 bubble. Two or three hours later when the pressure had fallen to 1000psia, and the temperature was also lower, we would estimate a bubble size of 1300 ft^3 .

We have not included the effects of any leakage of water or loss of hydrogen during the course of a measurement. The former clearly exists: An analysis in the Bubble Book shows

replenishment of the makeup tank at an average rate equivalent to $46 \text{ ft}^3/\text{hr}$ at RCS temperature and pressure. The excess over the bubble shrinkage rate of $16 \text{ ft}^3/\text{hr}$ ~~xxx~~ is presumably ^{un}accounted leakage, a mass loss of perhaps $1700 \text{ lb}_m/\text{hr}$ (3 or 4 gpm). This is confirmed in the data reduction in that, for cases where it is clear that no bubble exists (on 4/2/79 and 4/3/79), the RCS still appears to accept an excess 500-1000 lb_m of water during a typical pressure excursion. When time permits, we intend to repeat the analysis, including this average leakage rate in the mass balance from which the bubble size is inferred. We do not anticipate any large change in results.

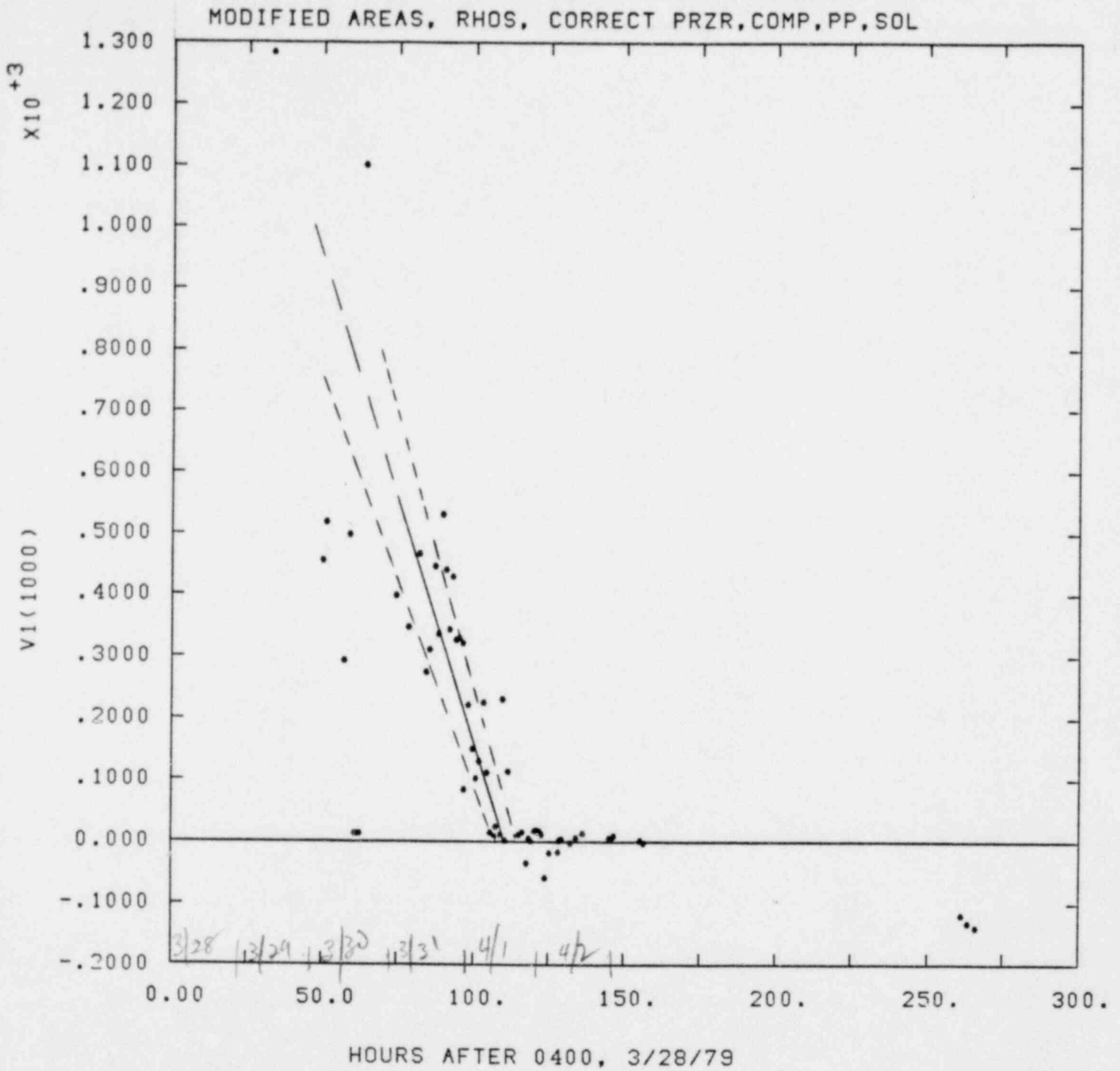


Fig 2 Bubble Volume at 1000 psia

DIKROFZ
 PLOT * 3
 * WORDS 3833
 TOTAL ERRS 0
 S.C. TIME 6
 TOTAL TIME 10

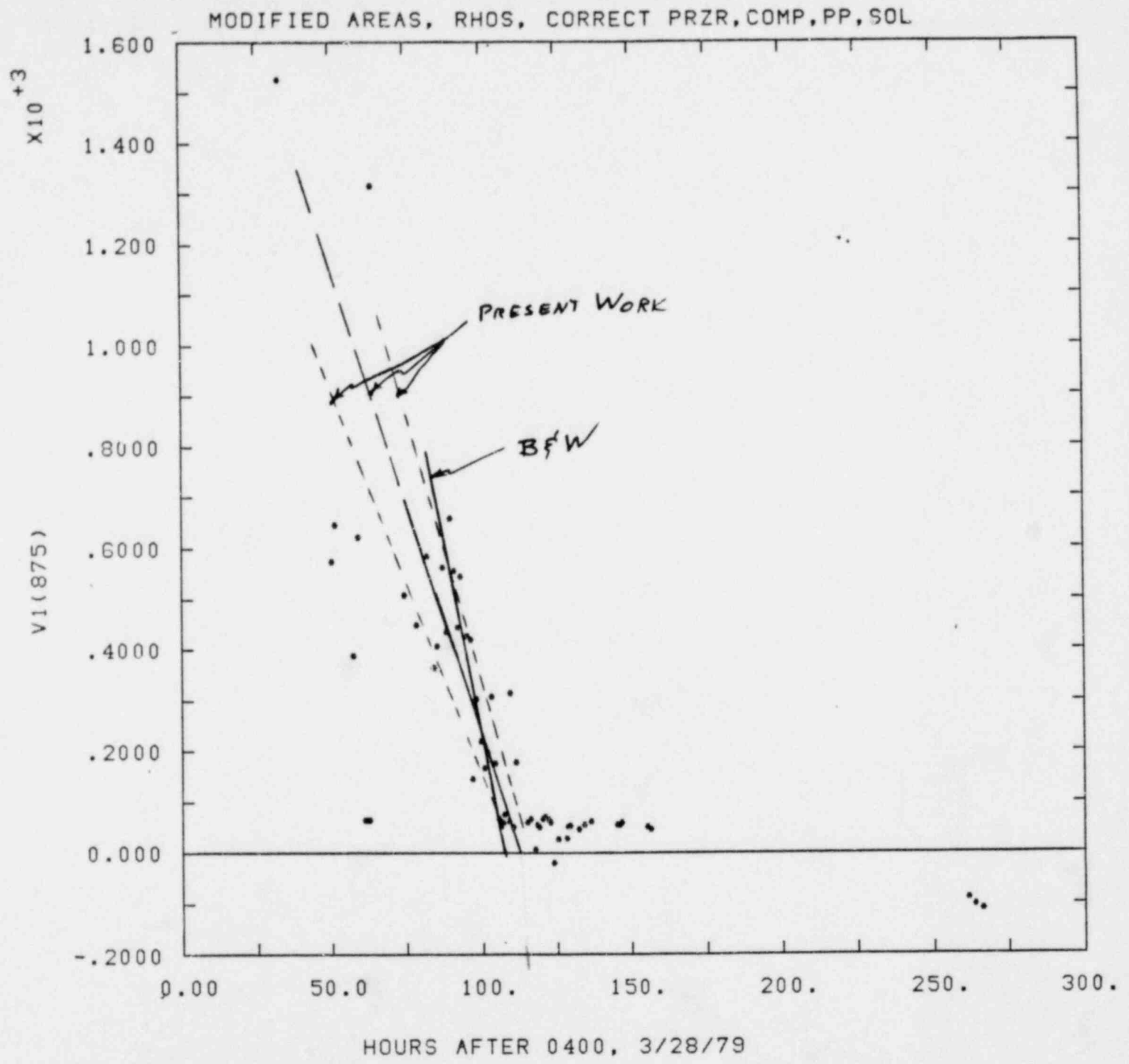


Fig 3 Bubble Volume at 875 psia

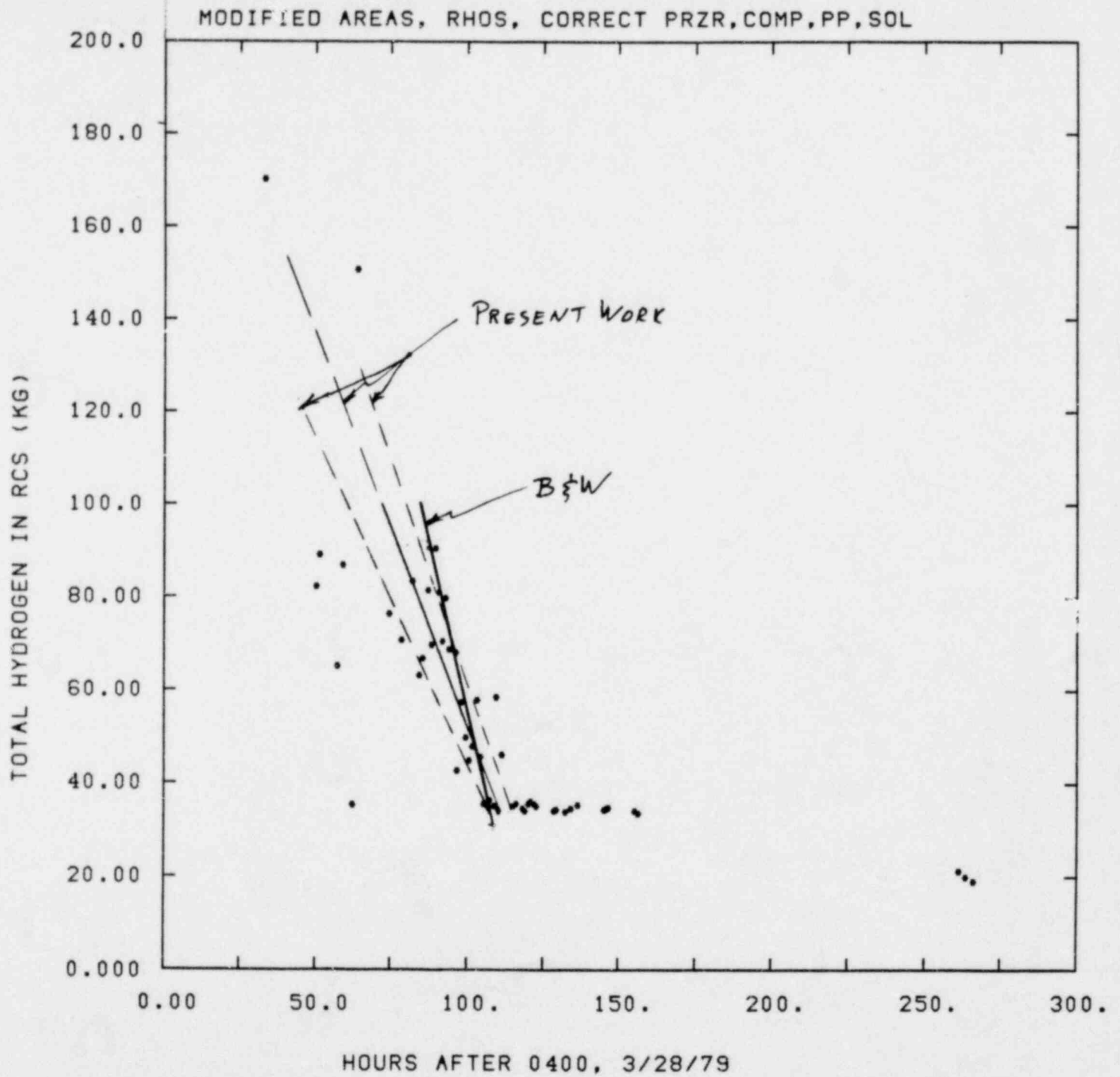


Fig 1 Total Hydrogen in RCS

R.K.Cole, Jr.
Sandia Labs, Albuquerque N.M
15 October 1979
HYDROGEN BURN

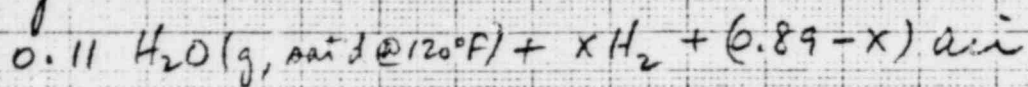
The attached graph was constructed assuming a containment atmosphere initially saturated with water at a temperature of 120°F at a slight overpressure of 1.5 psig relative to atmospheric pressure of 14.3 psia. It shows that an overpressure of 26 to 28 psig requires an initial hydrogen concentration of 7.1 to 7.6%.

If the containment building volume is taken as 2.05×10^6 ft³, we find that this requires 165 to 180 kg moles (330 to 360 kg). The burn will produce ~~x~~ 165 to 180 kg moles of water in addition to the initial 250 kg moles, and the post-burn atmosphere will have a dew point near 141°F. Therefore, we feel (but have not shown conclusively) that the primary mechanism for pressure decay is cooling by evaporation of additional water.

condensation?
no

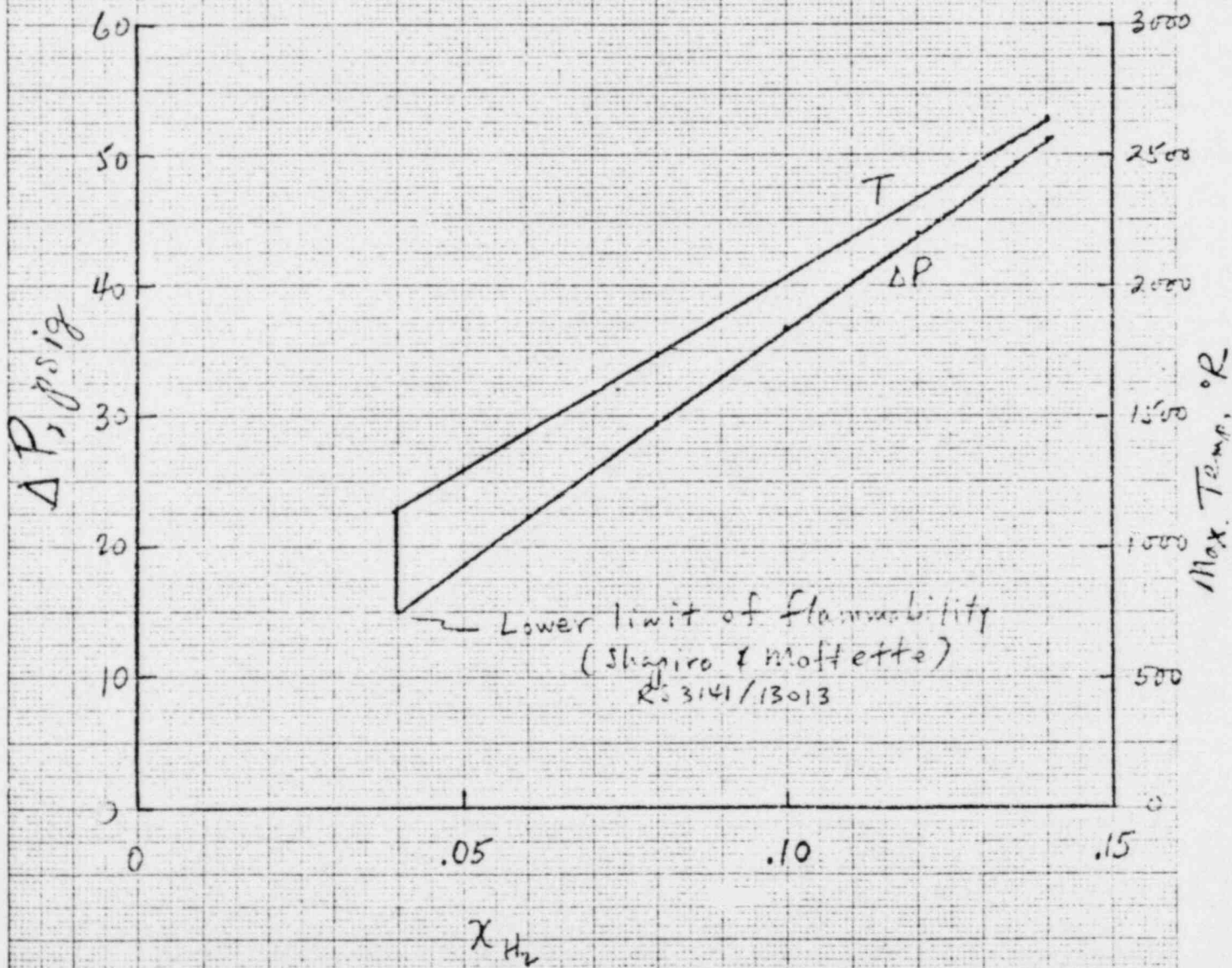
Conditions Following Hydrogen Combustion

Init. Gas Composition:



$$P_0 = 15.8 \text{ psia}, T_0 = 580^\circ \text{R}$$

Note: Lower detonation limit is at $x \approx 0.18$.



POOR ORIGINAL