

**HYDROGEN FLAMMABILITY DATA
AND APPLICATION TO PWR
LOSS-OF-COOLANT ACCIDENT**

CONTRACT AT-11-1-GEN-14

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PITTSBURGH, PENNSYLVANIA**

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TABLE OF CONTENTS

	<u>Page</u>
INTRODUCTION AND SUMMARY.....	1
I. GENERAL DISCUSSION REGARDING THE FLAMMABILITY OF HYDROGEN.....	3
A. Limits of Flammability.....	3
B. Conditions for Propagation of Flames in Mixtures of Gases...3	3
1. Ignition.....	3
2. Size of Vessel.....	5
3. Pressure.....	5
4. Temperature.....	5
5. Turbulence.....	8
6. Stratification.....	8
7. Effect of Chemically Inert Substances.....	8
8. The Limits of Flammability of Hydrogen in Air and Water Vapor.....	9
C. Spontaneous Ignition Temperature.....	9
1. Static Systems.....	9
2. Dynamic Systems.....	9
II. APPLICATION TO FWR LOSS-OF-COOLANT ACCIDENT.....	16
A. Combustion of Steam-Hydrogen Mixture Escaping from Rupture as It Is Evolved.....	16
B. Delayed Combustion of Accumulated Hydrogen.....	19
C. Vacuum Due to Combustion of Hydrogen.....	21
REFERENCES.....	23

LIST OF ILLUSTRATIONS

<u>Fig. No.</u>		<u>Page</u>	<u>Westinghouse Neg. No.</u>
1	Effect of Pressures above Normal on Flammability Limits of Hydrogen in Air	6	18678-2
2	Influence of Temperature on Limits of Flammability of Hydrogen in Air	7	18678-4
3	Flammability Limits of Hydrogen-Air-Steam Mixtures	13	18678-3
4	Variation of Minimum Spontaneous Ignition Temperatures (S.I.T.) with Hydrogen Concentration of Hydrogen-Water Vapor-Air Mixtures	14	18678-1
5	Variation of Minimum S.I.T. Values and Maximum Explosion Pressures with Water Vapor Concentration of Hydrogen-Water Vapor-Air Mixtures	14	18678-1
6	Variation in the Minimum S.I.T. with Exit Velocity of Hydrogen	15	18678-5
7	Variation in the Minimum S.I.T. of Hydrogen-Water Vapor Mixtures Flowing into Air	15	18678-5
8	Pressure in Plant Container after Large Rupture of Main Coolant System	18	20204
9	Percentage of Metal Reacted Vs. Quantity of H ₂ Formed	20	18678-6
12 of	WAPD-SC-544. Temperature of Gas Stream Leaving Reactor Vessel	17	20431-12
14 of	WAPD-SC-544. Composition of Gas Stream Leaving Reactor Vessel	17	20431-14

HYDROGEN FLAMMABILITY DATA
AND APPLICATION TO PWR LOSS-OF-COOLANT ACCIDENT

By Dr. Z. M. Shapiro and T. R. Moffette

Introduction and Summary

The purpose of this report is: (a) to summarize generalized data on the flammability of hydrogen in steam-air mixtures; and (b), of primary importance in PWR Safeguards considerations, to apply the flammability data to the postulated PWR loss-of-coolant accident and to determine the potential pressure effects on the PWR plant container.*


Other documents** of this series related to PWR Safeguards discuss the possibility and results of a major loss-of-coolant accident in PWR (the Shippingport Atomic Power Station). There it is shown that a zirconium-water (or steam) reaction is a possible result of such an accident, evolving significant quantities of heat and hydrogen. It is therefore necessary to assess the possibility of damage to the PWR plant container due to high pressure resulting from detonation or combustion of hydrogen.


A rupture of the reactor coolant system, causing loss of the primary coolant, is considered to be extremely unlikely. Furthermore, in order for this accident to result in an extensive zirconium-water reaction, with hydrogen and heat release: (1) the rupture of the primary coolant system would have to be located below the level of the core and in the part of the system within the reactor chamber of the reactor plant container; and (2), the rupture size must be sufficiently large so the Safety Injection System cannot replenish the water faster than it escapes from the rupture; and/or (3), the Safety Injection System supplies less than its maximum capacity, or does not function at all.

In the specific application of the general information on hydrogen flammability to the PWR loss-of-coolant accident in this report, it is shown that the hydrogen would probably be evolved from the rupture at such an elevated temperature that spontaneous ignition should occur, preventing a large amount of unburned hydrogen from accumulating. The evolution of hydrogen is

* The PWR reactor plant container is a pressure vessel completely surrounding the reactor, primary coolant system, and major auxiliary systems that contain hot, compressed, radioactive coolant. Its function is to enclose any radioactive material as may be accidentally released, preventing its escape to the outside atmosphere. The container was designed to withstand a 52.8 psig internal pressure, the calculated maximum that would occur due to release of hot-compressed coolant due to a rupture of the reactor coolant system.

** WAPD-SC-541, "PWR Hazards Summary Report"; WAPD-SC-543, "Zirconium-Water Reaction Data and Application to PWR Loss-of-Coolant Accident"; WAPD-SC-544, "PWR Loss-of-Coolant Accident - Core Meltdown Calculations."

sufficiently slow so that if the hydrogen is burned as evolved the pressure within the plant container would be raised by only a few psi, at a time when the total pressure would be considerably less than the plant container design pressure. 

In the event the hydrogen does not burn as it is evolved, (for example if it should escape from the reactor vessel through a water trap with resultant cooling of the gas) it is unlikely that a detonation would occur. If a uniform mixture of steam, air, and hydrogen is obtained in the plant container, the maximum hydrogen concentration would be below that required for detonation. Therefore, no detonation should occur unless localized pockets of hydrogen concentration occur. Even should such a pocket detonate, it is believed the peak pressure would not exceed the plant container design pressure. 

I. GENERAL DISCUSSION REGARDING THE FLAMMABILITY OF HYDROGEN

A. Limits of Flammability

A mixture of flammable gases such as hydrogen and air, or hydrogen and oxygen, may be diluted with one of its constituents or with other gases until it is no longer flammable. The marginal composition at which such a mixture will become flammable or non-flammable by a slight change is defined as the "limit of flammability."

A combustible gas mixture generally has two limits of flammability, an upper limit and a lower one. Only when the composition is well between these limits will the gaseous mixture burn or explode. In the case of hydrogen-air mixtures, these limits are generally quoted as 4.1% and 74% hydrogen for the lower and upper limits, respectively. The picture is not as clear cut as these figures would lead one to believe, however, since these limits vary greatly depending on whether the mixture is at room temperature or above, and whether it is at atmospheric pressure. Simple quotation of the limits of flammability is also misleading in that these do not indicate the kind of combustion which occurs, i.e., simple burning, explosion, or detonation. In handling explosive mixtures it is obviously desirable to do this in a manner such that the lower limit of flammability will be raised and the upper limit lowered, and in a manner such that the mixture, if ignited, will not detonate. The factors affecting these limits are discussed more fully in the following section.

B. Conditions for Propagation of Flames in Mixtures of Gases

1. Ignition

If a source of ignition is brought to the center of a combustible mixture, it would be expected that the flame would tend to travel away from the source uniformly in all directions. This would probably be the case if it were not for the convection currents set up by the hot, expanded combustion products which cause an upward movement of the gases. In the case of limit mixtures, the rate of propagation of the flame is less than the rate of rise of the flammable mixture due to convection currents. Under these conditions the flame cannot move downward. A similar condition holds true in the case of horizontal flame propagation, the limits for which lie between those for upward and downward propagation. Accordingly, the range of mixtures between the limits for upward and horizontal propagation will propagate flame upward but not horizontally or downward, whereas, the range of mixtures between the limits for horizontal and downward propagation will propagate flame horizontally and upward, but not downward. Thus, when limits of flammability are given it is important to note the direction of flame propagation to which the limits apply.

The lower flammability limits for hydrogen in air are 4.1% for upward propagation, 6% for horizontal propagation and 9% for downward propagation. Although the upper limits for horizontal and downward propagation are somewhat smaller than the upper limit for upward propagation, these are not well established. Accordingly, all the upper limits are taken as 74% in order to be on the safe side. In all cases, the above limits apply to gases maintained at atmospheric pressure and saturated with water vapor at room temperature.

Ignition of hydrogen-air mixtures, particularly when these mixtures are well within the flammability limits, takes place with an exceedingly small input of energy. A spark having such low energy that it is invisible in a dark room will ignite such a mixture. Common sources of ignition are sparks from electrical equipment, and sparks caused by the discharge of small accumulations of static electrical charges. Even though a mixture is below the limit of flammability, some combustion can be made to occur with a source of sufficient size and intensity. Such flames are not self-propagating, however, and will be extinguished as soon as the source of ignition is removed.

In general, mixtures of hydrogen and air burn with a very pale flame. In the case of the limit mixtures the flames may even be invisible. When mixtures having compositions close to flammability limits are ignited a ring of flame is formed which breaks up into balls of flame that travel to the top of the test vessel. Only a fraction of the hydrogen is burned in these cases. As the percentage of hydrogen in a hydrogen-air mixture is increased, however, an increasing amount of the hydrogen present will be burned. A 5.6% hydrogen mixture, for example, will burn only about 50% of the hydrogen, and complete combustion will not occur until the percentage of hydrogen is increased to 10%. Accordingly, as the percentage of hydrogen in mixtures with air is increased from 4.1% to 10%, the energy released by the combustion increases out of proportion to the increase in the amount of hydrogen.

As indicated by the flammability limits given above, it is not until the hydrogen concentration in air reaches 9% that the flame will be propagated downward. This is not to say that hydrogen and air mixtures having less than 9% hydrogen are not dangerous. This depends largely upon the point of ignition. For example, a mixture of hydrogen and air containing 6% hydrogen, if ignited near the ceiling, would propagate only upward and horizontally until the flame would reach the walls and ceiling where it would be extinguished; whereas the same mixture, if ignited near the floor, would propagate throughout the whole room, reach a mean temperature of 350°C*, and build up a pressure of approximately 1 atm. As the hydrogen concentration increases beyond 9% not only

* It is interesting to note that despite the fact that the spontaneous ignition temperature of a hydrogen air mixture is 585°C and despite the fact that 4.1% hydrogen air mixture has a flame temperature considerably below this, such a mixture will be self propagating. This phenomenon is attributed to the ability of hydrogen to diffuse into the flame more rapidly than other gases and thus maintain the necessary concentration for flame propagation.

does the flame propagate in all directions but the rate of propagation increases rapidly. When the concentration of hydrogen in air reaches 19%, the mixture detonates. This condition is particularly severe until the hydrogen concentration in such a mixture exceeds 57%.

2. Size of Vessel

Since the propagation of flame depends upon the transfer of energy from the burned to the unburned gas, and since in a limit mixture the amount of energy available for transfer is only just enough to propagate the flame, anything that reduces the available energy will affect the limits. Thus, if a vessel has a high surface-to-volume ratio, the effect of cooling by the walls of the vessel becomes appreciable and the flammability limits are narrowed somewhat.

3. Pressure

Normal variations of atmospheric pressure do not affect the limits of flammability appreciably. However, larger variations in pressure cause effects that are different for each mixture. Thus it is impossible to generalize. Fortunately, in the case of hydrogen-air mixtures, the range of flammability for downward propagation is narrowed at both limits by moderate increases of pressure above atmospheric. Above 10 atm and up to 220 atm however, the upper limit for hydrogen increases. The magnitude of this increase is shown in Fig. 1. In addition to the data given in Fig. 1, a series of experiments performed by Eitner ^(a) indicate there is no change in the lower limit over a range from 0.5 to 4.0 atm.

4. Temperature

To propagate flame, the layer of unburned gas next to the burning layer must be brought up to ignition temperature. If the unburned gas is already above room temperature, less heat need be supplied by the burning gas to bring the adjacent layer up to ignition temperature. Thus the range of flammability is widened at both limits by increasing the temperature of the mixture. In the case of hydrogen-air mixtures, as for most mixtures, there is a straightline relation between the limit of flammability and the initial temperature of the mixture. Thus, at 200°C the lower limit for downward propagation of hydrogen-air mixtures decreases to 7.9% while the upper limit increases to 76%; at 400°C the lower limit decreases to 6.3% while the upper limit increases to 81.5%. The change in the limits of flammability of H₂ in air for downward propagation are given in Fig. 2.

(a) Eitner, P. Explosion Limits of Flammable Gases and Vapors, Habilitationen - Schrift, Munchen, 1902.

EFFECT OF PRESSURES ABOVE NORMAL
ON FIAMMABILITY LIMITS OF HYDROGEN IN AIR

EXPERIMENT

- DOWNWARD PROPAGATION; CYLINDER, 37 x 8 CM
- DOWNWARD PROPAGATION, CYLINDER
- SIDE OR CENTRAL IGNITION; SPHERE, 7.6 CM DIAM.

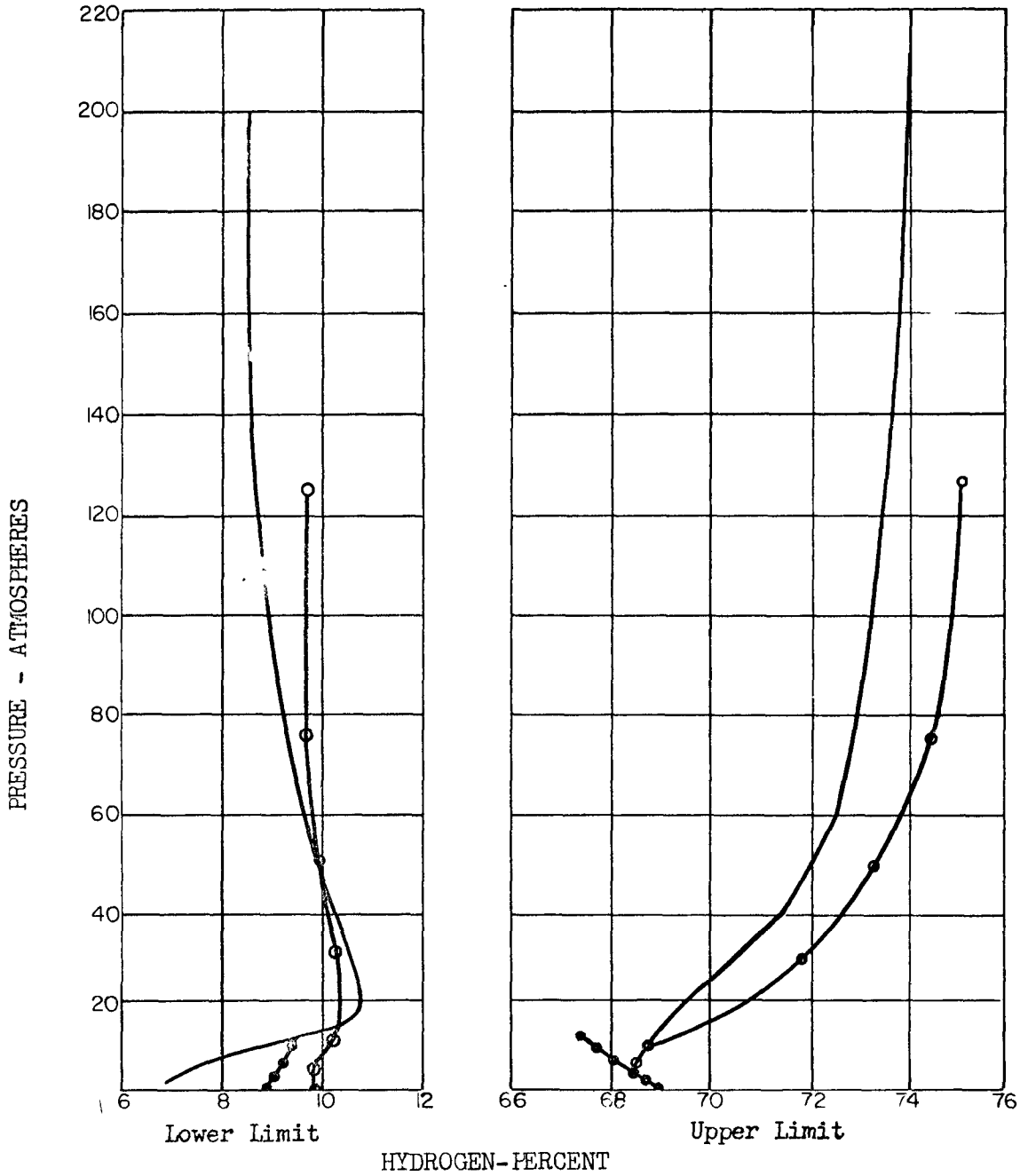


FIG. 1

INFLUENCE OF TEMPERATURE ON LIMITS OF
FLAMMABILITY OF HYDROGEN IN AIR
(DOWNWARD PROPAGATION OF FLAME)

- A. MAXIMUM AMBIENT TEMPERATURE ATTAINED IN CONTAINER
- B. AMBIENT IN THE CONTAINER

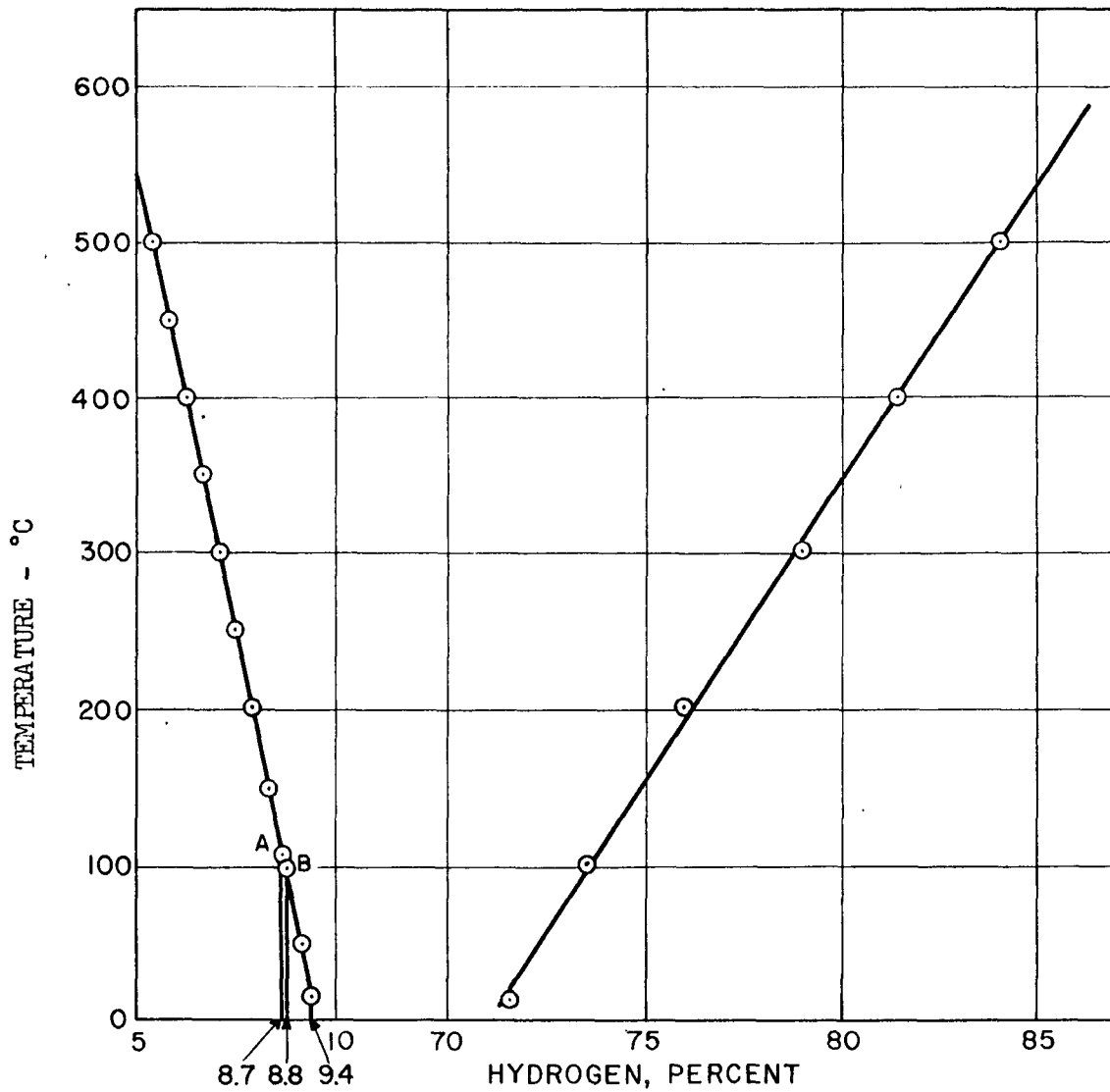


FIG. 2

A recent study* of the combined effects of pressure and temperature on the upper and lower limits of flammability of hydrogen-air-water vapor indicate that at 50 and 100 psig and 300°F the effect of temperature on widening the flammability limits is counterbalanced by the effect of pressure on narrowing the limits. Thus at 50 psig and 300°F the upper and lower limits of flammability for upward propagation are 77 and 5 v/o H₂ respectively, whereas at 100 psig and 300°F the upper and lower limits are 78 and 4 v/o H₂ respectively. As indicated previously, the limits at atmospheric pressure and room temperature are 4.1 and 74 v/o H₂.

5. Turbulence

No observations have been made on the effect of turbulence on the limits of flammability of hydrogen-air mixtures. It has been shown, however, that the lower limits of methane and ethane are reduced somewhat by agitation of the mixture. This effect is expected to be even smaller in the case of hydrogen-air mixtures due to the higher rate of diffusion of H₂ in the atmosphere.

6. Stratification

Because hydrogen has a lower density than that of any other gas, slow leakage of hydrogen near the ceiling of a room will cause the hydrogen to collect near to the ceiling in dangerous concentrations. This effect can be easily overcome by maintaining a slight circulation of the atmosphere in the room, or by venting the ceilings. This stratification does not occur if the leak is near the floor since the hydrogen readily diffuses through the atmosphere. Where leaks occur in high pressure lines even near ceilings, there is less tendency for stratification to occur because of the mixing action of the jet.

7. Effect of Chemically Inert Substances

The addition of increasing amounts of chemically inert substances to mixtures of hydrogen and air causes the upper and lower limits of flammability to approach each other and ultimately to meet. In vessels larger than 2 in. in diam, the effectiveness as a diluent of carbon dioxide, nitrogen, helium and argon decreases in the following order.

CO₂ N₂ He A

To render any mixture of hydrogen and air harmless, 11 volumes of helium, or 10.2 volumes of CO₂ per volume of combustible mixture are required.

* Division of Explosives Technology - Bureau of Mines Report #3543, 9/4/56

8. The Limits of Flammability of Hydrogen in Air and Water Vapor

Water vapor has been shown to be more potent than even CO_2 in its extinctive effect on mixtures of hydrogen and air. Whereas, as indicated above, 10.2 volumes of CO_2 are required to render 1 volume of any hydrogen-air mixture completely non-flammable, only 7.6 volumes of water vapor are required to accomplish the same purpose at atmospheric pressure. Experiments carried out over water in a thermostated spherical vessel with mixtures maintained at atmospheric pressure* indicate that the upper and lower flammability limits of hydrogen-air mixtures converge as the percentage of water vapor increases. As the temperature rises and the water vapor content rises as a consequence, the lower limit rises slowly while the upper limit falls rapidly. When approximately 60% of the hydrogen-air-steam mixture at one atm is composed of steam, the limits coincide at about 10% hydrogen. The available data for hydrogen-air-steam mixtures at atmospheric pressure are given in Tables 1, 2, and 3. Data for nitrogen and CO_2 are included for comparison. The data for hydrogen-steam-air mixtures at 300°F and at 0, as well as 100 psig, are best summarized and compared with the data at standard pressure in the triangular diagram, Fig. 3. It will be noted that there is very little change in the flammability limits in the entire range in which the data were collected.

C. Spontaneous Ignition Temperature

1. Static Systems

Tests conducted at the Bureau of Mines indicate only a small change in minimum spontaneous ignition temperature with pressure in the range between 0 and 100 psig. As shown in Table 4, minimum spontaneous ignition temperatures of hydrogen-air mixtures vary between only 516 and 523°C with the minima occurring at compositions of 30 and 35 v/o hydrogen. While the progressive addition of water vapor to the hydrogen-air mixtures results in a small increase (5%) in the minimum spontaneous ignition temperature, the resulting depression in the maximum pressure generated during the explosion is quite significant (35%). The data obtained at an initial pressure of 100 psig are shown in Figs. 4 and 5.

2. Dynamic Systems

If hydrogen is passed through a heated pipe into the atmosphere, mixtures are formed which are capable of spontaneous ignition after a period of time depending on the hydrogen temperature and the exit velocity. Although, as indicated previously, the minimum spontaneous ignition temperature under static conditions is 520°C**, the minimum hydrogen temperature required to

* J. S. Yeaw and L. Shnidman - A.G.A. Proceedings, Technical Section, pp. 717-745 (1938)

** In the presence of platinum, surface combustion occurs at temperatures as low as 100°C.

effect ignition in the dynamic experiments conducted at the Bureau of Mines was approximately 680°C. Since an incubation period is observed prior to ignition, of length depending on gas temperature and flow rate, it is assumed that the ignition occurs after the mixture formed by the hot hydrogen issuing from the pipe contains at least 4.1 v/o hydrogen, and has achieved a temperature of 520°C. It is important to note, however, that if a solid object is placed in the path of the heated gas, wide diffusion of the hydrogen is apparently prevented and the minimum spontaneous ignition temperature drops from 680°C to that observed in the static case, viz, 520°C. While some variation in the ignition temperature was found with changes in exit velocity, they are relatively small. However, the addition in substantial proportions of water vapor to the hydrogen does raise the minimum spontaneous ignition temperature in a significant manner. These data are summarized in Figs. 6 and 7.

TABLE 1

EXPLOSIVE LIMITS IN STEAM

Summary of Experimental Results for Hydrogen
(one atmosphere total pressure)

<u>Thermostat Temp. °F</u>	<u>% Gas in Gas-Air Mixture</u>	<u>% Steam Absolute</u>	<u>Upper & Lower Explosive Limits % Gas in Gas-Air- Steam Mixture</u>
68°	72.0	2.3	70.3
68°	4.7		4.6
130	68.1	15.3	57.7
130	5.6		4.7
150	62.7	25.7	46.6
150	7.1		5.3
160	58.8	32.7	39.6
160	8.0		5.4
170	51.8	41.1	30.5
170	10.2		6.0
180	40.7	51.5	19.7
180	14.1		6.8
185	30.1	57.6	12.8
185	18.0		7.6
186	28.0	58.9	11.5
186	20.0		8.2
187	no explosion of any mixture	60.1	---

TABLE 2

The Extinction of Combustible Gas Flames by Inert Gases
Minimum Fractions of Inerts in the Inert-Gas-Air
Mixtures to Prevent Flame Propagation

<u>Inert Gas</u>	<u>Volumes of Inert per Volume of Combustible</u>	<u>% Inert in Inert- Gas-Air Mixture</u>	<u>Temperature of Mixture °F</u>
HYDROGEN			
Nitrogen	16.5	71.1	75
Carbon Dioxide	10.2	56.5	75 *
Water Vapor	7.6	58.9	187

* Calculated

TABLE 3

THE EXTINCTION OF GAS FLAMES BY INERT GASES

Minimum Fractions of Inerts in the Inert-Gas Mixtures which are Necessary for the Prevention of Possible Explosive Mixtures Regardless of any Subsequent Dilution with Air*

<u>Inert Gas</u>	<u>Volume of Inert per Volume of Combustible</u>	<u>% Inert in Inert- Gas Mixture</u>	<u>Temperature of Mixture °F</u>
Nitrogen	16.5	94.5	75
Carbon Dioxide	10.2	91.5	75
Water Vapor	7.6	88.3	207

* Assuming no condensation of the steam

TABLE 4

Minimum Spontaneous Ignition Temperatures (S.I.T.) and Maximum Explosion Pressures of Hydrogen-Water Vapor-Air Mixtures in Contact with Stainless Steel (I-33A Apparatus)

<u>Composition, Vol. %</u>			<u>S.I.T. (°C)</u>	<u>Pressure, (p.s.i.a.)</u>		<u>Time Lag (sec.)</u>
<u>H₂</u>	<u>H₂O</u>	<u>Air</u>		<u>Initial</u>	<u>Explosion</u>	
25	--	75	528	14.7	20	13
30	--	70	520	14.7	58	7
35	--	65	546	14.7	30	9
25	--	75	532	50	150	5
30	--	70	523	50	160	6
35	--	65	530	50	140	9
25	--	75	524	115	187	3
30	--	70	516	115	190	2
35	--	65	535	115	185	6
25	10	65	529	115	140	14
30	10	60	525	115	150	2
35	10	55	531	115	145	7
40	10	50	541	115	140	5
30	20	50	557	115	140	2
35	20	45	533	115	140	5
40	20	40	558	115	140	4
45	20	35	562	115	140	4
25	30	45	559	115	120	5
30	30	40	548	115	120	1
35	30	35	548	115	120	7
40	30	30	580	115	120	2

FLAMMABILITY LIMITS OF HYDROGEN-AIR STEAM MIXTURES

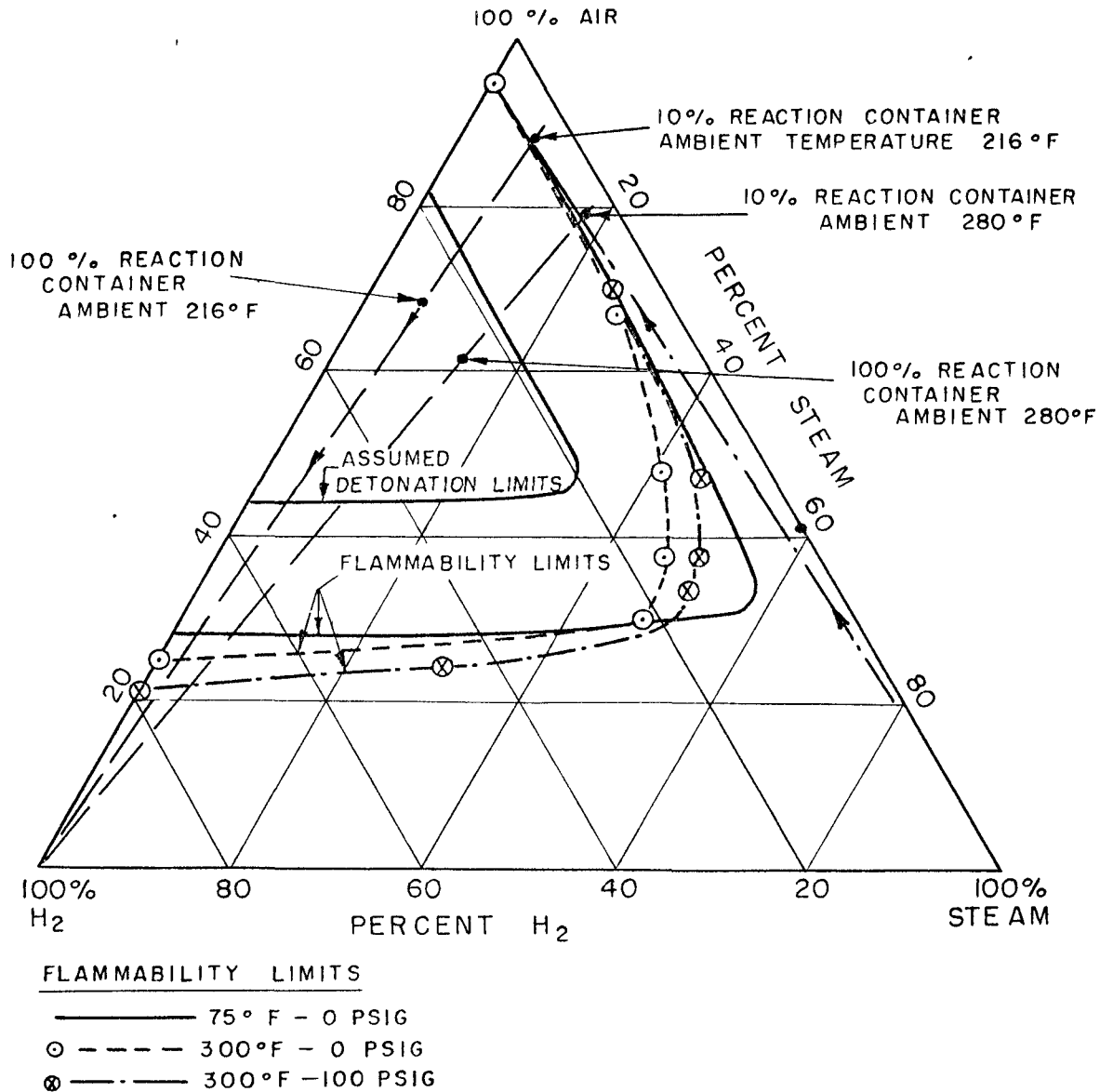


FIG. 3

FIGS. 4 and 5

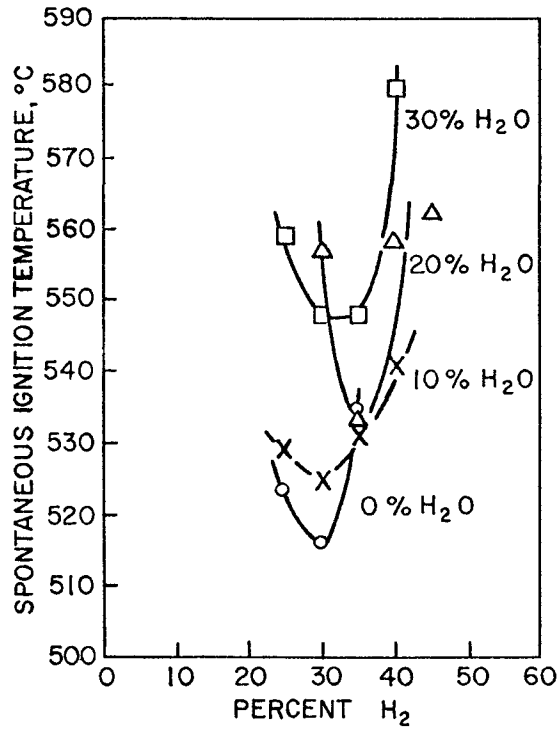


FIGURE 4 - VARIATION IN THE MINIMUM SPONTANEOUS IGNITION TEMPERATURES (S.I.T.) WITH HYDROGEN CONCENTRATION OF HYDROGEN-WATER VAPOR-AIR MIXTURES CONTAINING 0, 10, 20 AND 30 VOLUME PERCENT WATER VAPOR AT 100 P.S.I.G. INITIAL PRESSURE (STAINLESS STEEL APPARATUS.)

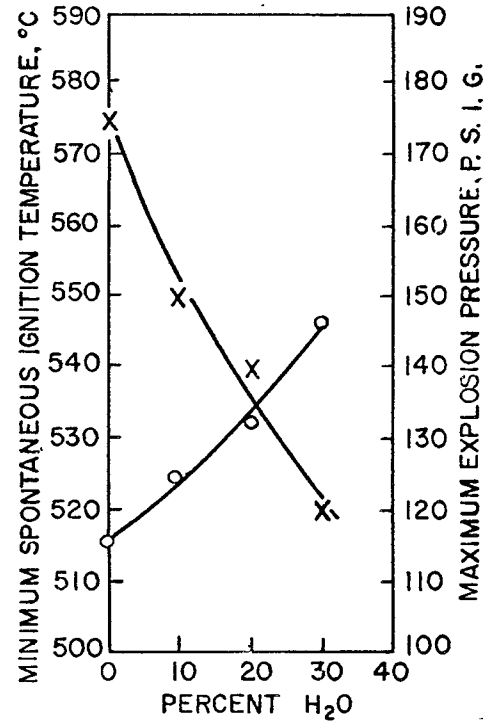


FIGURE 5 - VARIATION IN THE MINIMUM SPONTANEOUS IGNITION TEMPERATURE (S. I. T.) VALUES (O) AND MAXIMUM EXPLOSION PRESSURES (X) WITH WATER VAPOR CONCENTRATION OF HYDROGEN-WATER VAPOR-AIR MIXTURES AT 100 P.S.I.G. INITIAL PRESSURE (STAINLESS STEEL APPARATUS.)

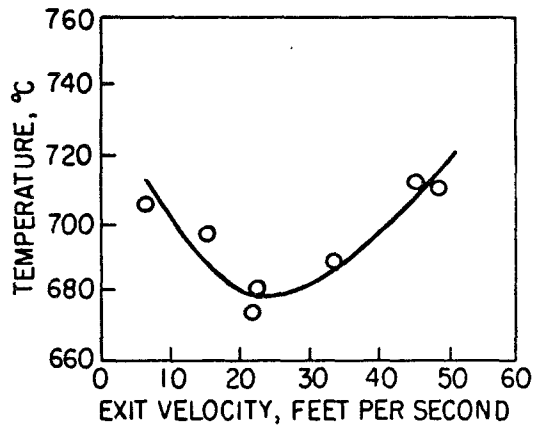


FIGURE 6 - VARIATION IN THE MINIMUM SPONTANEOUS IGNITION TEMPERATURE WITH EXIT VELOCITY OF HYDROGEN FLOWING THROUGH A 0.54-INCH DIAMETER X 59-INCH STAINLESS STEEL PIPE INTO UNHEATED AIR AT ONE ATMOSPHERE PRESSURE.

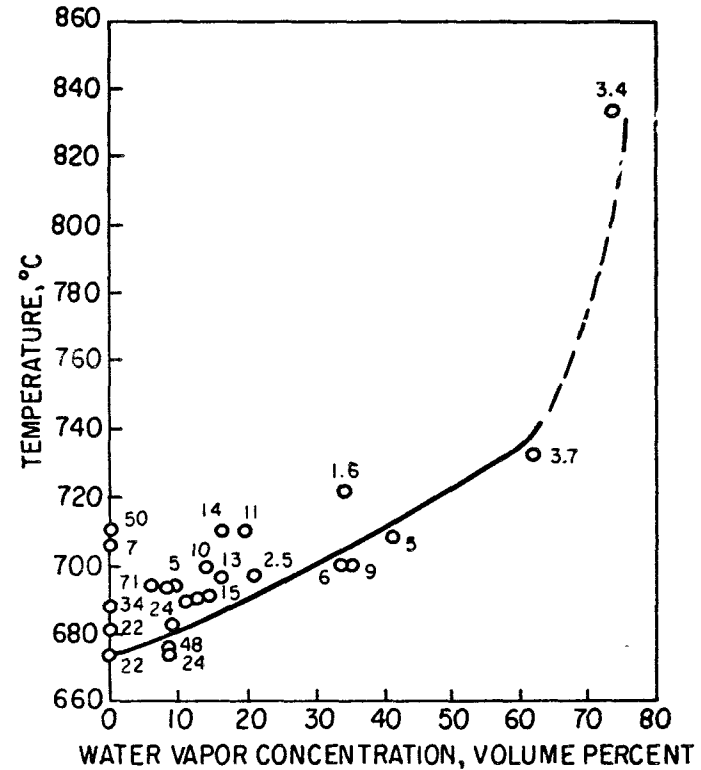


FIGURE 7 - VARIATION IN THE MINIMUM SPONTANEOUS IGNITION TEMPERATURE OF HYDROGEN-WATER VAPOR MIXTURES FLOWING THROUGH A 0.54-INCH DIAMETER X 59-INCH STAINLESS STEEL PIPE INTO UNHEATED AIR AT ONE ATMOSPHERE PRESSURE. (EXIT VELOCITY: 1.6 TO 71 FT. /SEC.)

II. APPLICATION TO PWR LOSS-OF-COOLANT ACCIDENT

A. Combustion of Steam-Hydrogen Mixture Escaping from Rupture as It is Evolved

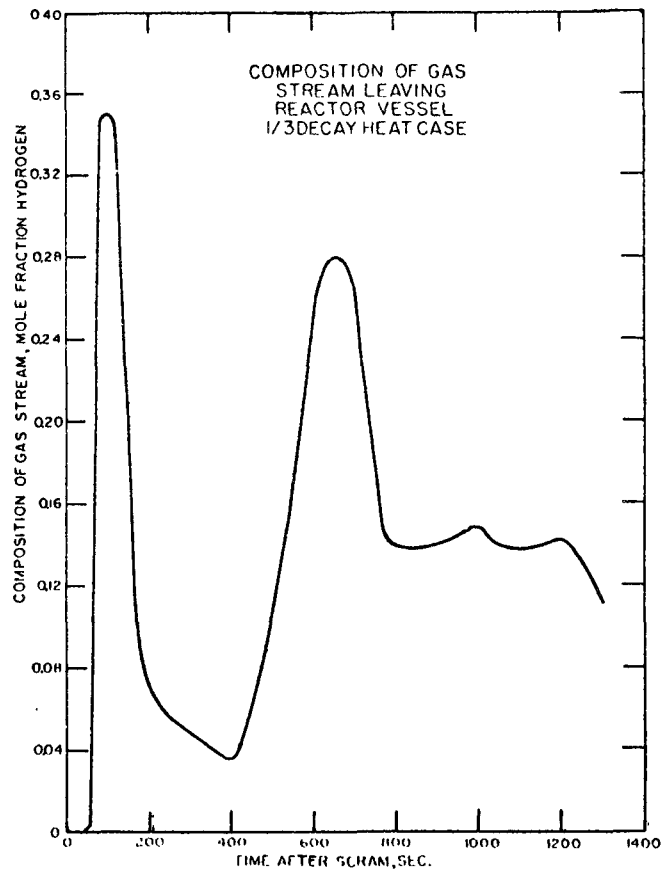
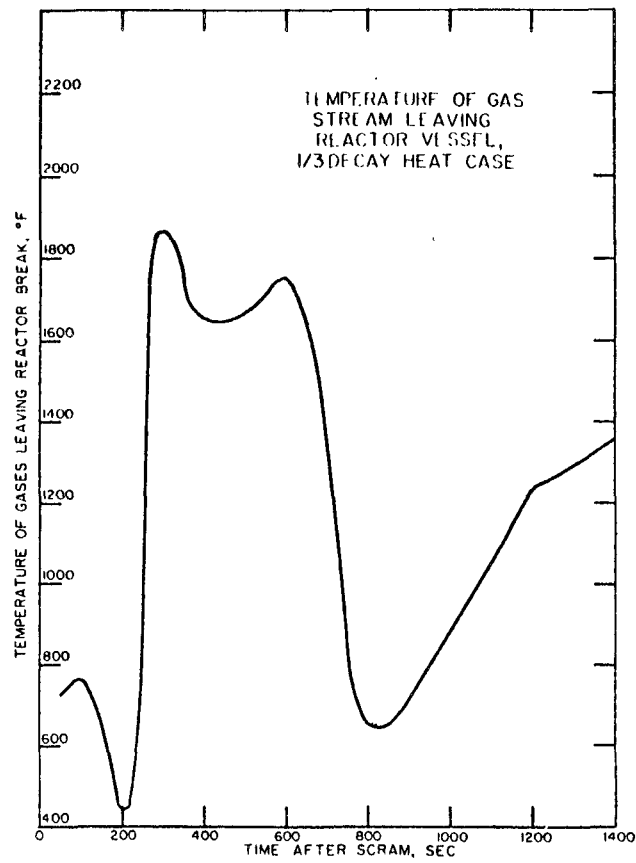
The preceding generalized discussion indicated that for dynamic systems in which a hydrogen-steam mixture issues from a pipe (the rupture), the minimum spontaneous ignition temperature is 680°C (1256°F). However, if the gas issues against a solid object (as would probably occur with a large rupture of the primary piping within the reactor chamber) the minimum spontaneous ignition temperature drops from 680°C to 520°C (968°F). Fig. 20 of WAPD-SC-544 indicates the escaping gas will reach the lower spontaneous ignition temperature in 252 sec after the rupture occurs, and the higher spontaneous ignition temperature will be obtained in 260 sec. After 200 sec the gas temperature quickly increases a second time, eventually reaching a maximum of 1870°F (1020°C) at 300 sec. Unless the gas is cooled before entering the atmosphere of the container (for example, if it should escape from the reactor vessel through a water trap), it appears unlikely that ignition would be delayed after 600 sec, and all hydrogen produced after ignition occurred would probably burn as it evolved. The greater portion of the gas would be produced after 600 sec. Therefore, it appears somewhat unlikely that any large amount of unburned hydrogen would accumulate. (The case of unburned hydrogen accumulating is discussed in subsection "B", below).

The composition of the gas stream escaping from the rupture for the first 1300 sec after the rupture is shown in Fig. 22 of WAPD-SC-544. There it is shown that a 25% hydrogen and 75% steam composition is obtained at 600 sec, (when Fig. 20 of WAPD-SC-544 shows the temperature is 1752°F or 955°C). Inspection of Fig. 7 indicates that for hydrogen-steam mixtures issuing from a pipe into air, with a 75% steam content the minimum spontaneous ignition temperature is about 835°C (1535°F). Therefore ignition of the gas appears assured.

Calculations have been made of the effect on plant container pressure due to burning of the hydrogen gas as it is evolved. Two cases have been calculated and the resulting pressure as shown in Fig. 8 within the plant container has been plotted. The first case is one in which the Safety Injection System is assumed not to function, and the maximum zirconium-water reaction and hydrogen production occurs, as calculated in WAPD-SC-544. The pressure increase within the container is calculated to be less than 5 psi, shown as the upper dashed line of Fig. 8.** The peak total pressure due to the hydrogen combustion is only 24 psig, considerably less than the plant container design pressure of 52.8 psig.

* Repeated in this report for convenience.

** See WAPD-SC-549 "PWR Plant Container Sizing Criteria" for information on the pressure (and temperature) within the plant container after a large rupture, with no zirconium-water reaction, as represented by the solid line of Fig. 8.



FIGURES 20 AND 22 OF WAPD-SC-544 (Repeated here for convenience)
TEMPERATURE AND COMPOSITION OF GAS STREAM LEAVING REACTOR VESSEL

PRESSURE IN PLANT CONTAINER AFTER
LARGE RUPTURE OF MAIN COOLANT SYSTEM

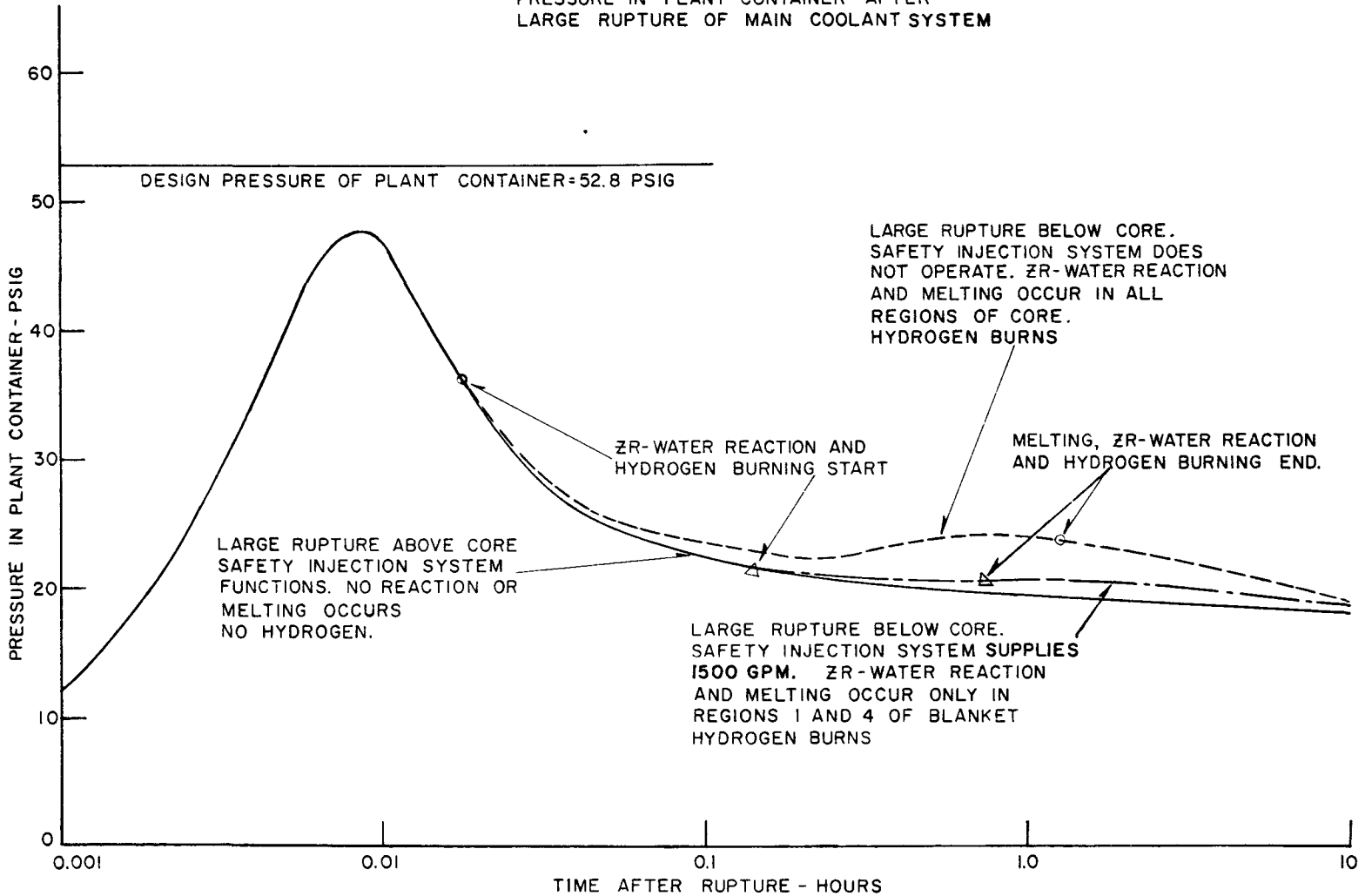


FIG. 8
PRESSURE IN PLANT CONTAINER AFTER
LARGE RUPTURE OF MAIN COOLANT SYSTEM

The second case calculated is one in which the Safety Injection System is assumed to function, but at its minimum output of 1500 gpm. Considerably less zirconium-water reactor and hydrogen production occurs, and as shown in Fig. 8, the pressure rise within the plant container is less severe than the former case.

While there appears to be no danger to the plant container due to over-pressure under the above conditions, the possibility of damage to the container caused by localized high temperatures in the region of combustion is extremely difficult to assess. This is due to the many unknowns to be considered in such a postulated accident, such as the exact location of a rupture.

B. Delayed Combustion of Accumulated Hydrogen

In case the hydrogen does not burn as it is evolved and escapes from the rupture (for example if it should escape from the reactor vessel through a water trap with resulting cooling of the gas), it is necessary to investigate the possibility of detonation of the accumulated hydrogen.

From the worst case accident investigated in WAPD-SC-544, it can be calculated that 3095 lb or 16.5% of the active zirconium in the core might react exothermically with steam or water, and producing 67.75 lb-moles of hydrogen. At standard conditions the hydrogen would occupy 24,330 cu ft. If the hydrogen is assumed to mix uniformly with the steam and air in the plant container, the possibility of burning or detonation may be determined by referring to Figs. 3 and 9. By comparison with the 10% reaction case in Fig. 3, it may be seen that a 16.5% reaction would make a mixture slightly inside or at the limit of the flammable region, depending on the temperature and the resulting fraction of steam in the mixture.

Fig. 9 is a simplification of Fig. 3, using only the flammability and detonation limits at 75°F, and applied specifically to the PWR loss-of-coolant accident. Two cases are presented in Fig. 9, one in which 45,000 cu ft of air have been swept out of the container by steam escaping from the rupture before the plant container ventilation system butterfly valves close, leaving 430,000 cu ft of air (at standard conditions) within the closed container. In the second case it is assumed the container is sealed at the time of rupture and no air escapes, leaving the entire container net free volume of 475,000 cu ft of air. There is no significant difference between the two cases in regards to the specific accident considered here. Here again it is seen that the maximum calculated reaction of 16.5% of the zirconium would produce only enough hydrogen for the air-steam-hydrogen mixture to enter the lower limit of the flammable region. If such a weak mixture should become ignited, only about 50% of the hydrogen would burn, and the energy release would be reduced accordingly, as previously mentioned in Sec. I-B-1. The pressure peak would not be expected to exceed the plant container design pressure, considering the data of Table 4 and the lean mixture.

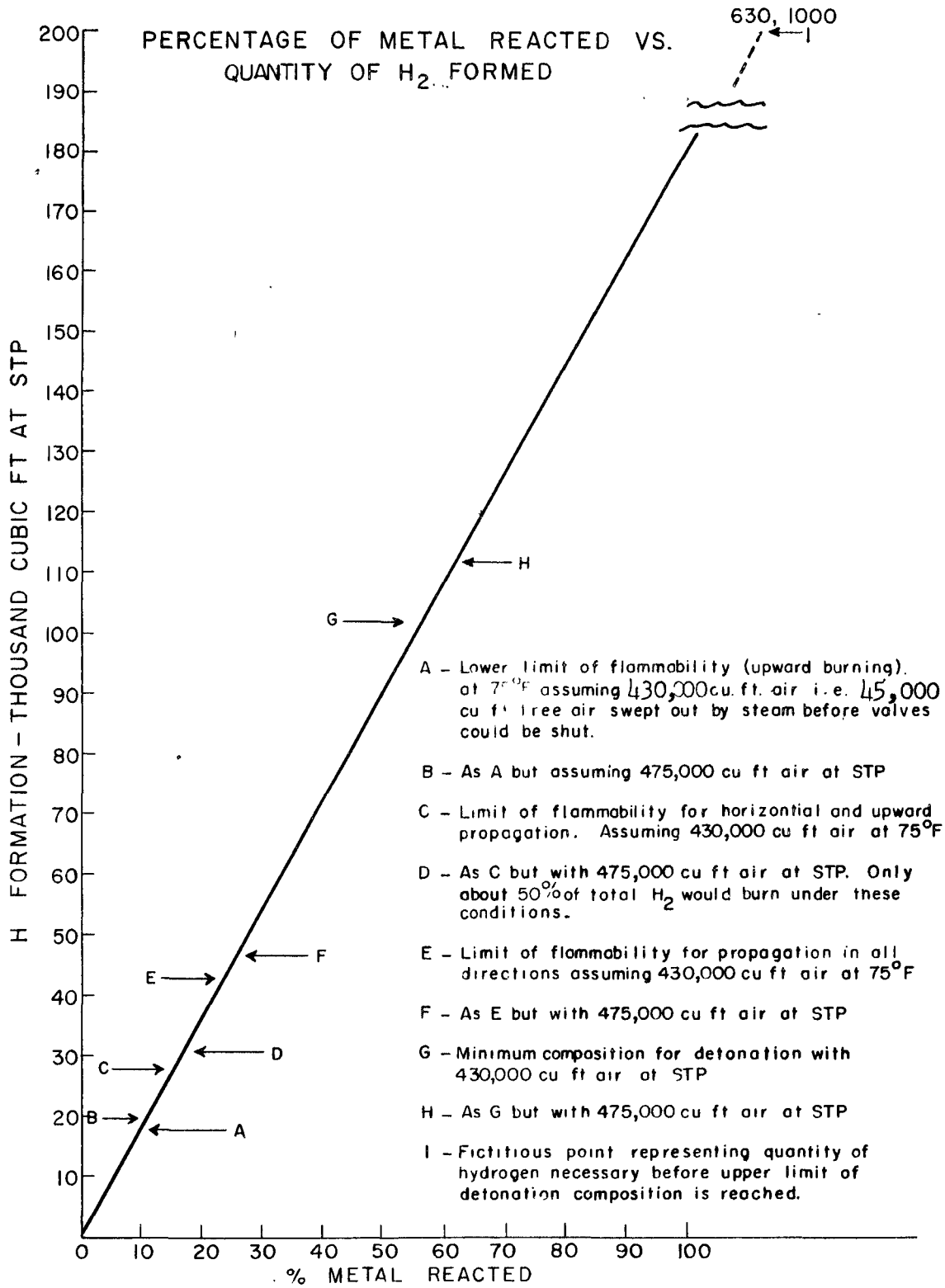


FIG. 9

Approximately four times the 16.5% reaction would be required to produce enough hydrogen to permit detonation, assuming a uniform mixture.

Another case has been calculated involving operation of the Safety Injection System at its minimum expected output of 1500 gpm of cooling water to the core, with melting and zirconium-water reaction limited to regions one and four of the blanket. The maximum zirconium-water reaction involves only 4.5% of the zirconium in the core, considerably less than the case previously considered. By reference to Fig. 9 it may be seen that, if uniformly mixed, the hydrogen product of a 4.5% zirconium reaction would be insufficient to bring the mixture within the flammable limit. Therefore, if the gas is not burned as it is evolved, and if it is uniformly mixed, neither burning nor detonation should occur.

For either of the above cases, however, the multivessel compartmented plant container might hinder uniform mixing of the hydrogen, air, and steam. Local pockets with high concentrations of hydrogen may occur, causing an explosion hazard in that region.

If an explosion occurs, the 475,000 cu ft net volume of the container would cushion the over-all shock considerably. Data on the maximum explosion pressures of hydrogen-air-steam mixtures given in Table 4 indicate the resulting pressure rise should be comparatively slight, due to the presence of steam in the atmosphere. The pressure in the plant container would be less than 24 psig before an explosion, so the peak pressure would not be expected to exceed the plant container design pressure of 52.8 psig.

C. Vacuum Due to Combustion of Hydrogen

After the atmosphere within the container cools and the steam condenses, the internal pressure will be reduced below the initial pressure in proportion to the amount of oxygen or air consumed, creating a partial vacuum. The plant container is designed to withstand an external pressure (internal vacuum) of 3 psi.

The calculated maximum amount of hydrogen that could be produced as a result of the loss-of-coolant accident and a subsequent zirconium-water reaction is 67.75 lb-moles, as mentioned above in Section B. Combustion of all of the hydrogen would consume 33.9 lb-moles of oxygen, or 12,180 cu ft at standard conditions.

Assuming none of the 475,000 cu ft of air in the plant container escapes before the ventilation system butterfly valves close at the start of the accident, the combustion of 12,180 cu ft of oxygen would cause a 0.4 psi vacuum after the steam condensed.

Should as much as 45,000 cu ft of air escape before the butterfly valves close, and with the combustion of 12,180 cu ft of oxygen, the final pressure within the container would be only 1.7 psi below atmospheric pressure.

It is concluded that the postulated accident will not cause the external pressure on (internal vacuum in) the plant container to exceed the design differential of 3 psi.

REFERENCES

The information presented in Section I was taken largely from the following documents:

1. Bureau of Mines Bulletin 503, Limits of Flammability of Gases and Vapors, by H. F. Coward and G. W. Jones.
2. A paper by J. S. Yeaw and L. Shinidman entitled "The Extinction of Gas Flames by Steam" found in the Technical Section of A.G.A. Proceedings for 1938, pp. 717-745.
3. Division of Explosives Technology, Bureau of Mines Report No. 3543, September 4, 1956, by M. G. Zabetakis.