CHAPTER 8

HYDROGEN COMBUSTION

8.1 Introduction and Background

The role of hydrogen in severe accident phenomenology and related containment integrity concerns is well known. This role was actually played out in the TMI-2 accident where a large quantity of hydrogen was generated and a spontaneous "burn" event in the containment was experienced. Vivid demonstrations of spontaneous burns have been found in the SURTSEY facility, where relatively large scale corium melt simulants are expelled by high pressure steam into a containment atmosphere. Among all different containment loading mechanisms, the "hydrogen combustion" one had been singled out by Theofanous and Saito (1981) as a principal concern.

It is interesting to note that among the various severe accident phenomena "hydrogen generation" is the only one that already has found a place in existing regulations, and severe accident management, in the US. This happened following the TMI-2 accident. In particular, the 1978 limits (10 CFR 50.44 and Reg. Guide 1.7) on metal-water reaction in the ECCS evaluation criteria were reconsidered, and as a result of hydrogen combustion consequences the Mark I and II containments have been inerted and the ice condenser and Mark III containments have been provided with thermal igniters. [[The effectiveness of thermal igniters in controlling hydrogen has been demonstrated for a wide range of premixed and continuous injection conditions (Haugh et al., 1991). Nevertheless,]] it can be mentioned at the outset that both of these "fixes" have shortcomings and/or detractors. For example, "inerting" is clearly adequate to protect against combustion; however, it does impede (if not degrade) operations, and thus it has a negative impact on inspection and maintenance. The igniters, on the other hand, besides the question of availability (i.e., power source in a station blackout) have raised issues of location and reliability. Even a rather extreme position of adverse consequences has been voiced (Oppenheim, 1988). Even though the primary role of hydrogen is in combustion events, containment loading from its partial pressure may also be significant, especially for small, and therefore already inerted, containments.

8.2 Detailed Description of Concerns

For the basic loading mechanism, the "problem" is straightforward. It can be computed simply by heating up the containment atmosphere by the reaction heat of the available hydro-[illegible] This is because once initiated a major combustion event would be expected to proceed to completion, and do so fast enough [illegible] can be expected to be negligible. As an example of this approach for a large PWR containment (say, Zion) burning, at once, the hydrogen from oxidation of 100% of the core zircalloy, would produce a $\Delta P/P_0$ of $\sim$1.44, where P_0 is the initial pressure before combustion.

Unfortunately, this simple treatment is marred by two significant complications. The first and most important one is that under certain conditions a combustion front can accelerate into supersonic speeds yielding a detonation. Although the total energy release is the same, a detonation entails very significant local dynamic behavior characterized by very high pressures, which would be further augmented by reflection off the containment walls. The reflected wave is a multiple of the incoming one, and this multiplication factor increases beyond 2 (valid for accoustic waves) to larger numbers with increasing the mach number of the incident shock. Under these conditions, structural evaluations must account, in general, for transient loading as well as for multidimensional effects. The deflagration-to-detonation transition (or DDT) is not completely understood yet, thus it is not exactly predictable; however, it is known to relate to the stiochiometry (i.e., the composition) and the level of turbulence (i.e., the geometry). As a result, one needs to consider, besides the total quantity of hydrogen needed for the simple treatment, also its spatial distribution, which leads in turn to the time-history of release and containment atmosphere motions. In particular, one looks for the extent of hydrogen maldistribution due to stratification—the stratification mechanism exists because of the lower density of hydrogen (compared to the other constituents of the containment atmosphere) and it can potentially be found even in complex interactions with condensation-driven currents (Travis and Theofanous, 1987). [[One also can examine the interior layout of the containment building for potentially contributing factors, e.g., small tightly arranged compartments having constricted compartment-to-compartment openings, or long duct-like enclosures having little or not transverse venting.]] Thus, the first complication consists, in fact, of three topical problem areas, namely: metal-water reaction history in a degrading core and associated hydrogen release evolution; containment currents, condensation, and resulting hydrogen distribution (or maldistribution); and deflagration-to-detonation transition criteria, including related igniter aspects.

The second complication arises only if one's strategy is to back off from the Grade A-type treatment of normal combustion events. For non-energetic releases of hydrogen, hydrogen concentrations in the containment build up rather slowly; thus, with igniters operating the burning will be slow, such that containment heat sinks *count*. Accordingly, one now needs to be concerned about hydrogen release rates, ignition events (i.e., reaching appropriate conditions), and heat losses.

as already noted, in such cases, the partial pressure of hydrogen could become significant in producing a direct loading mechanism.

8.2.1 Specific Consideration of the AP600

It will be recalled from Chapter 3 that the AP600 containment is of the "large–dry" type, it has no internal sprays, and it is to be equipped with igniters (or recombiners). Also, from Chapter 4, we recall that "high pressure scenarios" can be made, and hence considered, of negligible frequency. Under low pressure we expect that hydrogen production and release will be gradual (during the in-vessel core melt progression) and rather unlikely to approach 100% reaction of the available zirconium (14.6 tons). Further oxidation reactions are possible in the reactor cavity (which would be flooded, as noted in Chapter 3), and these may not be gradual; however, as discussed in Appendix B, we believe it will be possible to demonstrate that for the AP600 vessel breach (leading to ex-vessel sequences) is "physically unreasonable." This allows consideration of the hydrogen challenge from an in-vessel release perspective only, a gradual one, which in a Grade A approach will be taken to involve 320 kg-moles of hydrogen, i.e., a 100% conversion.

Let us consider a saturated containment atmosphere at 100 °C, as is typical of large dry containments after the discharge of the contents of the primary system. The air partial pressure is raised (by the heating) to 1.27 bar, the 320 kg-moles of hydrogen exert a partial pressure of 0.2 bar, and together with the steam partial pressure, we obtain a total containment pressure of 2.47 bar (36 psia). The hydrogen mole fraction in this atmosphere is 8%, and it would increase to an absolute upper value of 13.6% if all the steam was to condense. The AP600 design approach is to burn the hydrogen as it is being produced, thus preventing the buildup of concentrations significantly greater than the lower flamability limits (~4 to 5%). Note that this also eliminates the possibility of significant (in the DDT context) stratification concerns. An initial perspective of resulting temperature and pressure increases can be obtained from an adiabatic combustion, which can be approximated for the range of conditions of interest by

$$\frac{\Delta P}{P_o} = \frac{\Delta T}{T_o} \simeq \frac{\eta_h \Delta H_r}{(\eta_s + \eta_a + \eta_h)\bar{c}_p \bar{T}_o} \sim 20 \frac{\eta_h}{\eta_s + \eta_a + \eta_h} \tag{8.1}$$

where ΔP and ΔT are pressure and temperatures increases; P_o and T_o are the initial pressure and temperature, respectively, and η_h, η_s and η_a are the kg-moles of hydrogen, steam, and air in the containment. For example, consider the case at the flammability limit: from an initial atmosphere at $P_o = 2.47$ bar and $T_o = 373$ K, burning of 160 kg-moles of hydrogen (i.e., at 4% mole fraction) would produce final pressures and temperatures of ~4.45 bar (65.3 psia) and 671 K, respectively. With a design pressure of 4 bar (60 psia), such pressurization amounts to an [illegible]

and penetration seals would clearly need a careful consideration. Even at the upper limit (320 kg-moles) the pressure rise to ~6.4 bar (94.4 psia) would not be expected (based on current experience) to yield failures at any significant rate (low end of the fragility curve), although it must be emphasized that such an event is not physically possible in the AP600 design.

8.2.2 Specific Consideration of the SBWR

The SBWR is inerted, so the role of hydrogen is limited to that of a non-condensible. We saw in the AP600 that this role is negligible; however, here we have only 16.6% of the containment-free volume and double the quantity of zirconium. Oxidation of all zirconium would produce 640 kg-moles of hydrogen and a partial pressure in the containment (say, at 313 K) of 2 bar (30 psia). In relation to the design pressure of 4.76 bar (70 psia) this does not appear to present a significant concern. This conclusion may not be as straightforward in extended accident scenarios that produce elevated pool temperatures followed by a rapid, massive steam generation event (i.e., an ex-vessel fuel-coolant interaction); however, we believe it should be easy to show that the passive features of the SBWR together with a solid approach to preventing and managing ATWS (see Chapter 6) make such circumstances extremely unlikely, i.e., excludable in terms of their low frequency. Also, the energetics/coolability approach discussed in Chapters 7 and 9 is consistent with excluding massive fuel-coolant interaction events. From another perspective, we need to be concerned about the potential role of hydrogen in interfering with the function of the isolation condensers. Finally, it should be mentioned that the SBWR design currently includes safety-related igniters for design basis control of deinerting expected to occur within 72 hours due to radiolytic decomposition.

8.3 Current Approaches to Resolution or Mitigation

Besides the well-known approach of inerting, the principal efforts toward hydrogen control have been in the quest of reliable igniter or catalytic recombiner systems.

The German (Siemens) work on spark igniters is also particularly noteworthy (Heck and Hill, 1992). From a radically different perspective, another effort, also in Germany (KfK), is aiming to design detonation-resistant pressure boundaries in large dry containments (Kuczera, 1992). A brief review and comment on these efforts is provided in what follows.

[[A variety of designs have been considered for catalytic recombiners (Rhode et al., 1991), in which buoyancy flow is driven between vertical catalytic panels by the heat of reaction. The Siemens design discussed further below employs flat plates on which catalyst material is deposited. Another design, in which panels consist of cartridges containing ceramic pellets coated with catalyst material, for the AP600 and SBWR (Wolff, 1993).]]

[illegible]

approach to hydrogen control is two-pronged, involving spark igniters and recombiners. The recombiners are of relatively low capacity (~3.6 kg-moles/hr for a 1.5 x 1.4 x 0.3 m panel at a containment pressure of 2.6 bar and a hydrogen concentration of 4%) and intended to moderate the rates of hydrogen concentration buildup in the region below the lower flammability limits. Both the recombiner and the igniters are passive, requiring no external actuation or source of power. The igniters employ two diverse principles: one based on accumulation of heat of recombination on catalytic foils, the other producing high voltage discharges (sparks), automatically initiated after sensing increased pressures or temperatures. In the course of qualification testing, one set of batteries was found to allow up to 28 days of continuous operation. The recombiner design employs catalytic panels, and buoyancy flow driven by the heat of reaction. Both igniters and recombiners were subjected to extensive functional tests at the Siemens test facility in Karlstein, and to more integral-type qualification in HDR and in the model containment of Battelle Institute. Key conclusions can be listed as follows:

1. For Igniters
   - Resistance to thermal aging, vibration, and radiation
   - Adequate performance under thermodynamic loads in the presence of airborne impurities and mythyl-iodine load.
   - Ignition occurs almost immediately after ignitable gas mixtures have formed (including steam deinerting)
   - Ignition not effective for steam concentration higher than 45%
2. For Recombiners
   - Recombiner capacity increases significantly with hydrogen concentration.
   - Recombiners are also effective under steam inerting.
   - Recombiners promote mixing of the atmosphere.

In summary, Siemens believes that these devices are now fully qualified and ready to use; they estimate that a 1300 MWe PWR large dry containment would require 40 recombiners and 150 igniters.

In another version of these recombiners designed by the GRS, the catalytic plates are kept in an inerted box (to avoid contamination), and they are supposed to automatically unfold on the occurrence of an accident; in this unfolded position, the plates are enveloped by filters, to protect against aerosols.

[illegible] 1991). In our view, the information available to us (i.e., openly available) on this two-pronged approach and the particulars of the performance of each prong are promising and worthy of serious consideration.

The KfK approach (Kuczera, 1992), on the other hand, assumes that ignition does not occur until all possible zirconium and some of the iron have been oxidized to produce 1300 and 400 kg of H_2, respectively, which increased by another 300 kg ("for conservative reasons") yields a quantity of 2000 kg to be reckoned with. Depending on the quantity of steam available, this corresponds to hydrogen concentrations in the 16 to 20% range, i.e., presenting significant deflagration loads as well as detonation potential. For example, from an initial pressure of 3 bar and use of Eq. (8.1), we obtain a final pressure of 12.6 bar. KfK reports a potential pressure rise in the 5 to 15 bar range, as well as a value, limited only by the amount of oxygen available, of 17 bar. As seen in Figure 8.1, the conditions in the above example (16% hydrogen, ~35% steam) is quite close to the detonation zone, as it is understood today, and in the context of this investigation (i.e., what has been postulated already), DDT is clearly envisioned. Peak detonation loads of 105 bar, with an impulse of 60 kPa·s over one-third of the upper containment surface (about 1400 m^2) have been obtained from 1-D (conservative) calculations. The work is continuing along the lines of refining these loads (3-D calculations) and the associated design of the I-beam spacers, supporting the containment steel shell against the reinforced concrete (2 meters thick) of the containment structure.

This is, in our view, a radical new approach to containment design with advantages and disadvantages that may be difficult to judge at such an early stage of the design. Accordingly, at this time, all we can do is view this effort cautiously. It is important to acknowledge, however, that the basic premise of it—that all (or the vast majority of) the 150 independent igniters (the Siemens recommendation noted earlier) fail to perform their function—is arbitrary. [[The earlier EPRI work with thermal igniters shows that relatively few preactivated igniters are necessary to control hydrogen during continuous injection (Haugh et al., 1991).]] Eventually, the choice will come down to the cost and any safety drawbacks weighted against this arbitrary safety margin. As a technical issue, we would like to mention that so far the thermal loads and their accommodation remain unknown, if not understated. For example, the extreme case considered above would produce a containment atmosphere heatup to 1566 K, clearly a situation deserving a most thorough consideration.

8.4 Proposed Approach and Quantification Needs

It appears quite feasible that the hydrogen concern be addressed by Grade A-type approaches in both designs. The SBWR containment is to be inerted, and the role of hydrogen as a noncondensible (a source of pressure) can be readily accommodated in the absence of major steaming events (see also Chapters 7 and 9). On the other hand, hydrogen could interfere with [illegible] it is addressed in the design and confirmatory experiments are planned. [[Catalytic recombiners are also under consideration for the SBWR.]] The AP600 containment is to employ igniters or catalytic recombiners, and again, it is straightforward in showing that hydrogen combustion loads are readily accommodated. There are no internal sprays, nor can we expect energetic releases of hydrogen; thus, we need not be concerned about rapid deinerting events, or stratified situations (that is, the intense natural circulation currents promoted by external cooling of the containment shell should easily and completely disperse the slowly released hydrogen). We agree that this igniter-based approach is sound, and we appreciate the advantages of using passive igniters of two different operation principles, as in Siemens. Also, we believe the further potential improvement by the use of recombiners alone, or in combination with igniters, merits careful consideration. [[As noted by Dr. Leaver, the US ALWR Program Requirements Document concludes that adequate hydrogen protection can be provided by recombiners only, and Prof. Mayinger indicates that the German Reactor Safety Commission decided to recommend the use of recombiners only for hydrogen management in existing German PWRs.]] Future needs are limited, therefore, to establishing the reliability of these equipment. As noted above, Siemens has met its major objective in this regard, but a wider and diverse qualification basis may be appropriate to meet the screening frequency goals in this important area.

8.5 Conclusions and Recommendations

The hydrogen control/mitigation approach adopted in both passive advanced designs is solidly based. For the AP600, we recommend a thorough investigation of existing igniters/recombiners and of the supporting data base, a selection, and further work on qualification, as needed, to meet the reliability goals necessary to make concentrations significantly in excess of 4% physically unreasonable [[see also Reference 1]]. For the SBWR, the role of hydrogen as a noncondensible is dual. As potentially interfering with the function of the isolation condensers, this role is being quantified. As a pressurizing agent it can be significant only in conjunction with fuel-coolant interactions, but within the coolability/energetics approach discussed in Chapters 7 and 9, this role does not appear to be significant.

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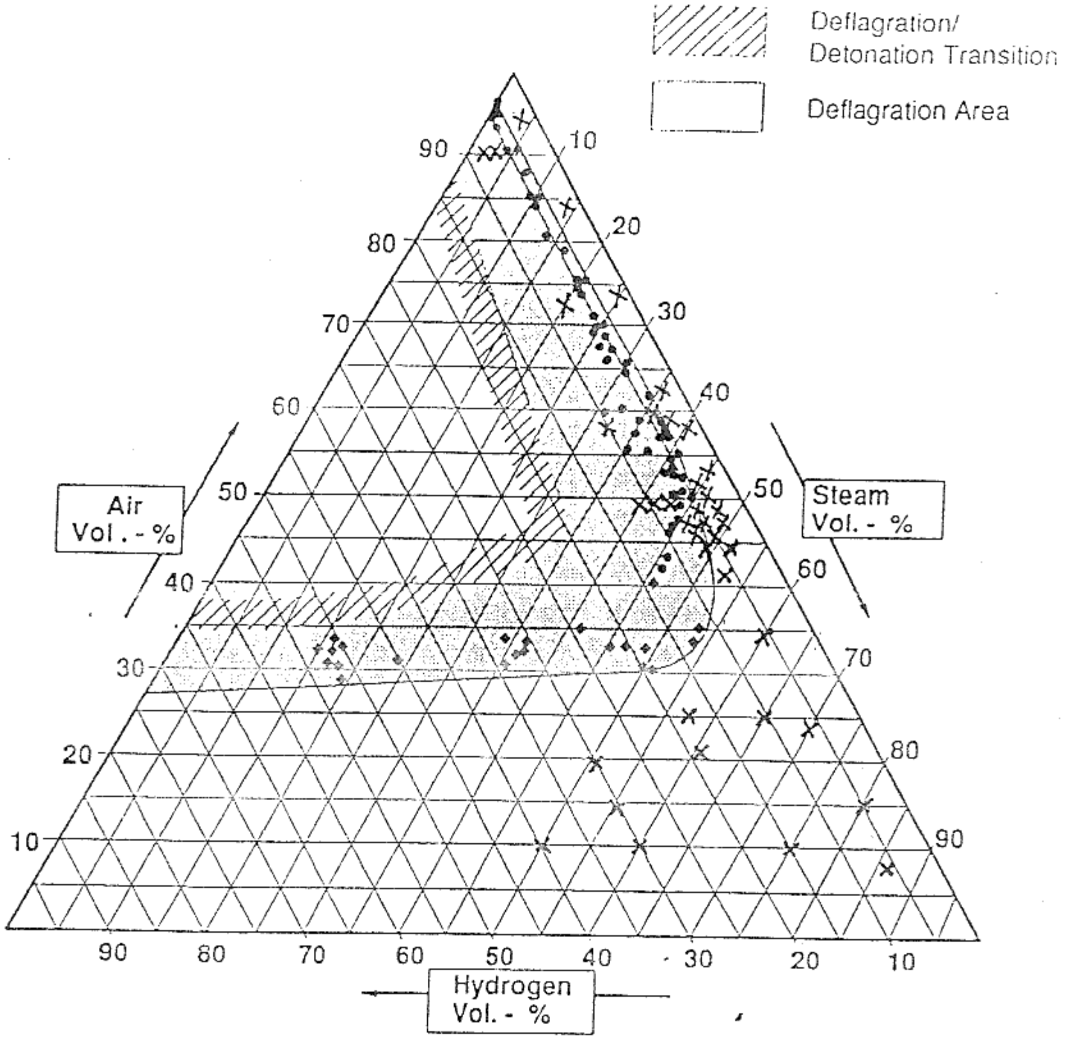
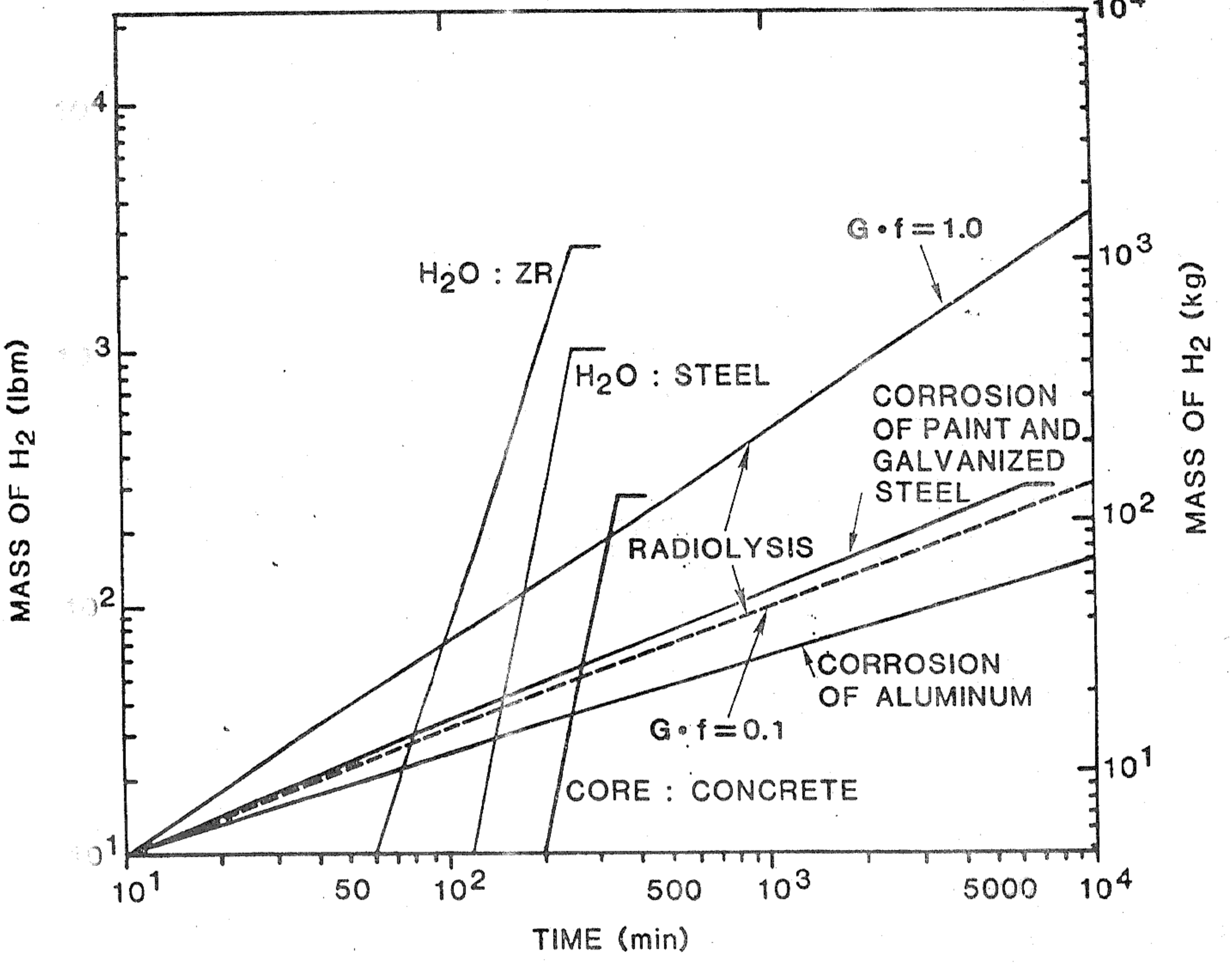


◆ ● Performed Ignitions with Catalytic and Spark Igniters
× Successful Tests with Catalytic Recombiners

HYPOTHETICAL H_2 PRODUCTION



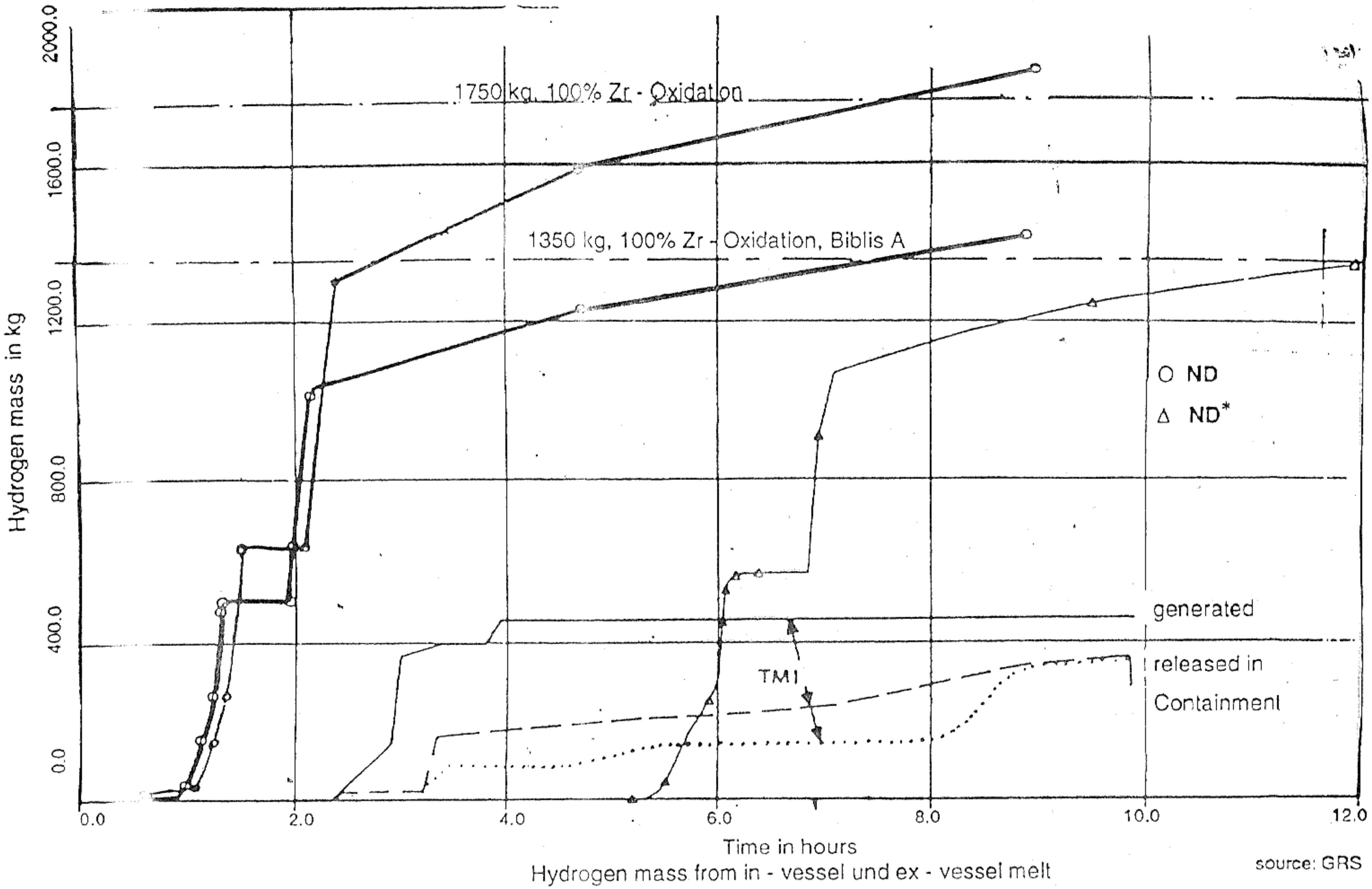
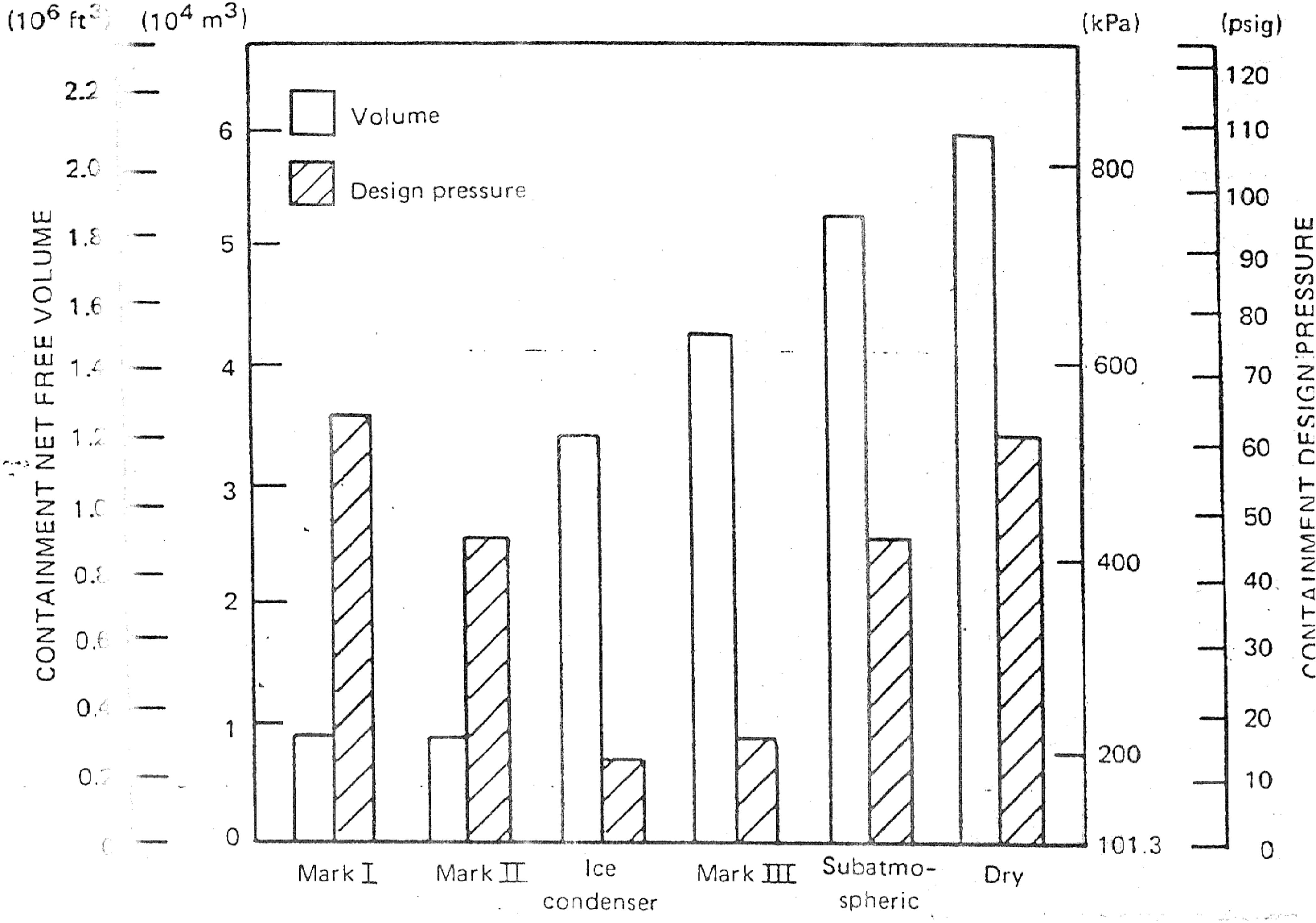
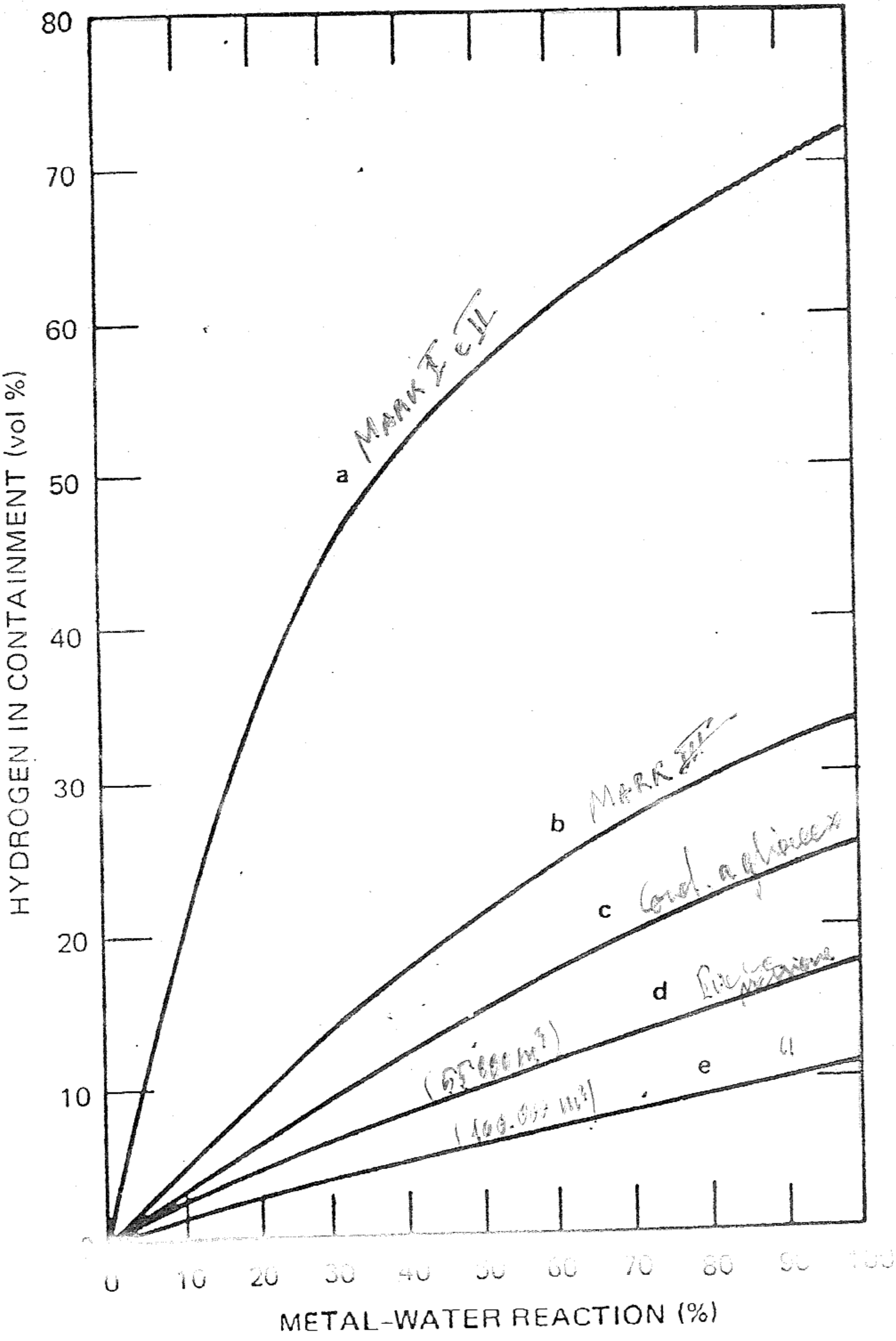


Fig. 1: EPR, Basic assumptions concerning the H_2 - situation with severe accidents/
Time History of H_2 - release in the containment derived from German Risk Study, Phase B

S/ KWU
E443
Hill/ 8.92
07-006-I



CONTAINMENT NET FREE VOLUME
(10^6 ft^3)
(10^4 m^3)
2.2
2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
6
5
4
3
2
1
0
Volume
Design pressure
Mark I
Mark II
Ice condenser
Mark III
Subatmo-spheric
Dry
(kPa)
800
600
400
200
101.3
(psig)
120
110
100
90
80
70
60
50
40
30
20
10
0
CONTAINMENT DESIGN PRESSURE


HYDROGEN IN CONTAINMENT (vol %)
METAL-WATER REACTION (%)
0
10
20
30
40
50
60
70
80
90
100
a
b
c
d
e
MARK I e II
MARK III

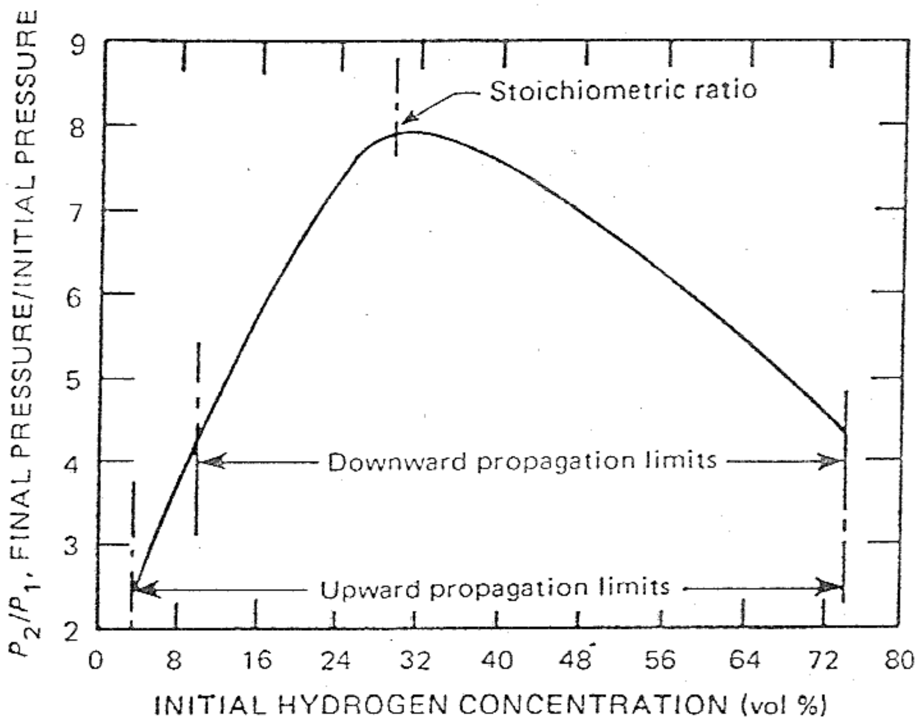


Fig. 3 **Theoretical adiabatic, constant-volume combustion pressure of hydrogen-air mixtures.** Initial conditions: T_1 = 298 K 25°C); P_1 = 101 kPa (1 atm); 100% relative humidity.

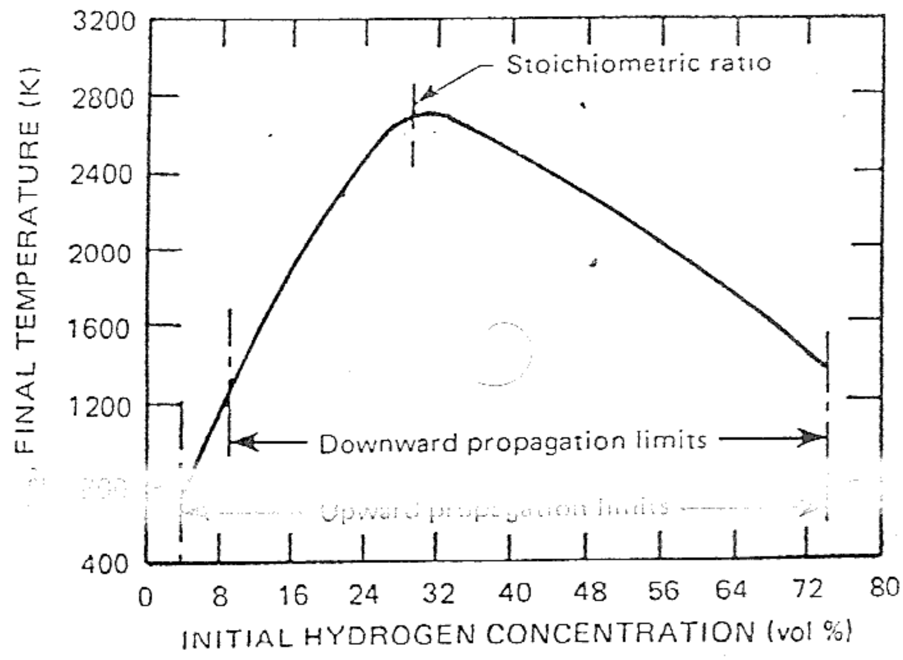


Fig. 4 **Theoretical adiabatic, constant-volume combustion temperatures for hydrogen-air mixtures. Initial conditions:** T_1 =

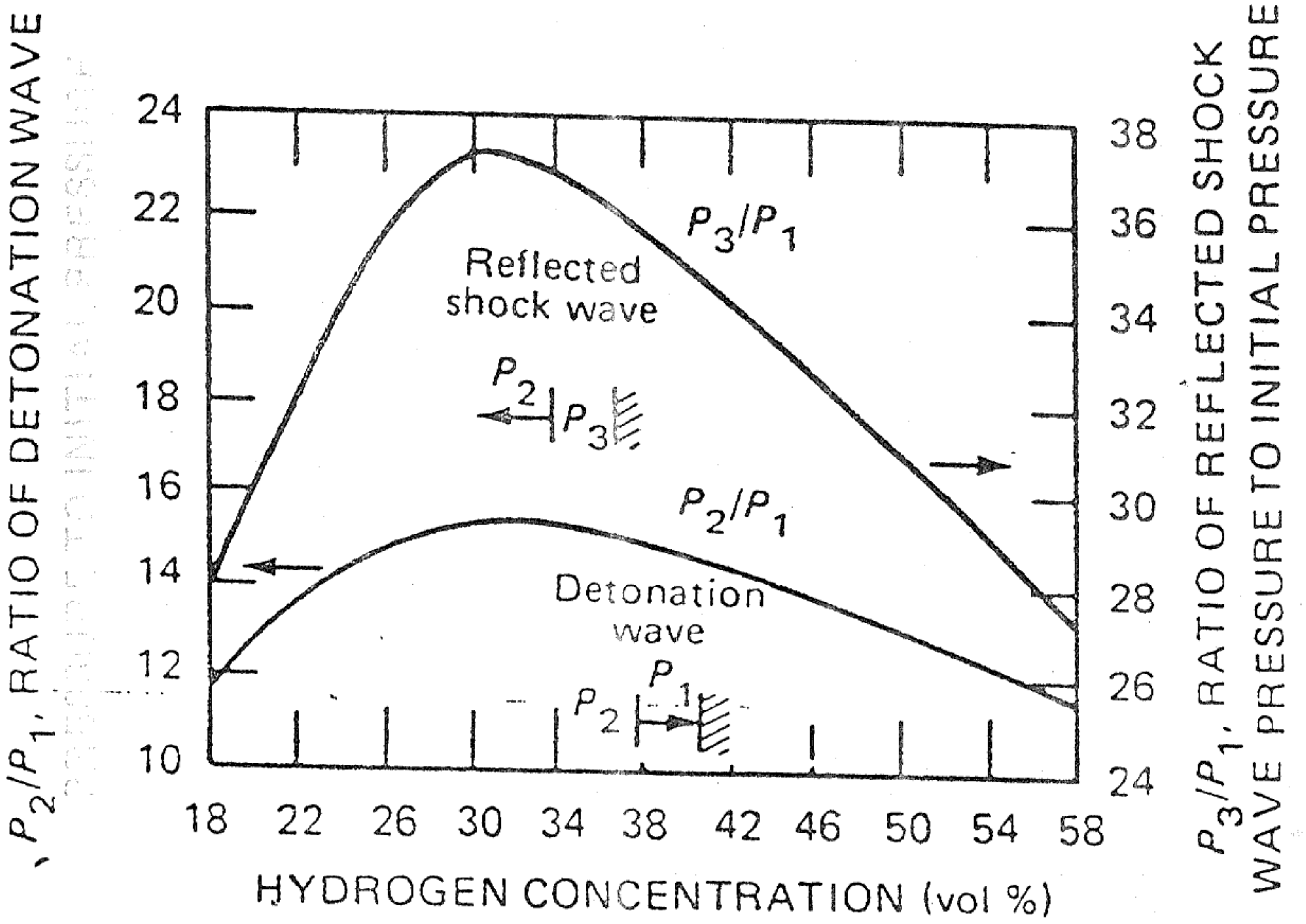


Fig. Theoretical detonation pressure and normally reflected detonation pressure for hydrogen–air mixtures. Initial conditions: T_1, 298 K (536°R); P_1, 101 kPa (1 atm); and 100% relative humidity.