

Conf-9410216--6

SANDIA REPORT
SAND94-3101C
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Printed January 1995

DELIBERATE IGNITION OF HYDROGEN-AIR-STEAM MIXTURES UNDER CONDITIONS OF RAPIDLY CONDENSING STEAM

Presented at the 22nd Water Reactor Safety Meeting, Bethesda, MD, October 24-26, 1994

Thomas Blanchat and Douglas Stamps



Sandia National Laboratories
Albuquerque, NM 87185

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Prepared for
Division of Systems Research
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, DC 20555
NRC FIN L2443

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Deliberate Ignition of Hydrogen-Air-Steam Mixtures Under Conditions of Rapidly Condensing Steam*

Thomas K. Blanchat and Douglas W. Stamps
Sandia National Laboratories
Albuquerque, NM 87185

Abstract

A series of experiments was conducted to determine hydrogen combustion behavior under conditions of rapidly condensing steam caused by water sprays. Experiments were conducted in the Surtsey facility under conditions that were nearly prototypical of those that would be expected in a severe accident in the CE System 80+ containment. Mixtures were initially nonflammable owing to dilution by steam. The mixtures were ignited by thermal glow plugs when they became flammable after sufficient steam was removed by condensation caused by water sprays. No detonations or accelerated flame propagation was observed in the Surtsey facility. The combustion mode observed for prototypical mixtures was characterized by multiple deflagrations with relatively small pressure rises. The thermal glow plugs were effective in burning hydrogen safely by igniting the gases as the mixtures became marginally flammable.

Program Description

A hydrogen-air-steam mixture that is initially nonflammable because it is diluted by steam may be rendered flammable when water sprays cause rapid condensation of steam. If the condensation process is sufficiently rapid, then a question arises as to whether a detonation could occur in such mixtures if thermal glow plugs were active during the operation of the water sprays. Detonations may be possible if: (1) ignition by thermal glow plugs is delayed until enough steam has been removed to make the mixture intrinsically detonable or (2) ignition occurs early near the flammability limits but the time for the entire mixture to burn is so long that any unburned mixture can be made intrinsically detonable by the action of the water sprays before the burn is complete.

The effect of water sprays on the combustion of hydrogen-air-steam mixtures was investigated in a 2048-m³ spherical vessel [1]. Unlike the scenario described earlier, however, the water sprays and the combustible gas mixture were at the same temperature so that there was no steam condensation. Since the steam concentration did not change, the chemical sensitivity of the flammable mixture was fixed prior to activation of the igniters. Similar experiments were also performed using water sprays with hydrogen-air mixtures [2-3]. In all of these experiments, the mixtures were flammable prior to the activation of the igniters. These experiments did not address the issue of mixtures becoming more reactive by the removal of steam during the same time the igniters were active. This issue was investigated for hydrogen-air-steam mixtures without water sprays in a 17-liter quasi-spherical vessel [4]. By using water-cooled coils around the outside of the vessel, the steam condensation time could be reduced to less than 10 minutes. However, the condensation

* A final report will be published under SAND94-1676

time can be significantly shorter when the steam is condensed by internal water sprays. Furthermore, the effect of the spray-generated turbulence on the combustion behavior was not investigated. The purpose of the current set of experiments was to investigate the combustion behavior of initially nonflammable mixtures that rapidly became flammable through the action of water sprays when the igniters were activated the entire time.

Depending on the location and the conditions under which hydrogen is released into the containment, the hydrogen may be either stratified or well mixed. Under stratified conditions, the hydrogen is concentrated in the upper part of the containment. If steam were absent, these mixtures would be more sensitive to detonation than if the same quantity of hydrogen was released at a low location and well mixed throughout the entire containment. For the CE System 80+ containment, the well-mixed concentration of hydrogen is approximately 13.6% on a dry basis assuming 100% metal-water reaction of the active cladding [5]. If all of this hydrogen accumulates above the operating deck, the average concentration in that region would be approximately 19.3%. The ability of water sprays to mix stratified mixtures is an important factor in determining the resulting combustion mode.

The combustion mode that results when initially nonflammable mixtures are rendered flammable by rapid condensation of steam caused by water sprays depends on the competition among three processes: (1) the removal of steam by water sprays, (2) the consumption of hydrogen by chemical reaction, and (3) for stratified mixtures, the mixing of hydrogen by water sprays. If the time required to remove steam is slow relative to the other time scales, then combustion can be initiated as the mixture first becomes flammable and a slowly propagating flame will result.

The objectives of this program were developed to cover the conditions of well-mixed or stratified hydrogen-air-steam mixtures prior to the operation of the water sprays. The program objectives were to determine: (1) if detonations or other forms of energetic combustion are possible when originally nonflammable mixtures are rendered flammable by water sprays, (2) the effectiveness of water sprays in mixing hydrogen-stratified mixtures, and (3) the effect of hydrogen stratification on the maximum combustion pressure.

Experiments were conducted in the modified Surtsey vessel shown in Figure 1. The vessel is a domed cylinder 3.6 m in diameter, 5.6 m high, and has a volume of 59.1 m³. The vessel was instrumented with pressure transducers, gas grab sample bottles, and thermocouple rakes. A deliberate ignition system was installed which was composed of actual plant igniters (GM AC7G thermal glow plugs) at three locations. The effect of igniter location was tested using one or more igniters at these three locations. For most tests, the target water spray mass flux was scaled to one spray train at runout flow in the CE System 80+ standard design. Owing to the characteristics of the nozzle used in the tests, the spray mass flux was the same as for the CE System 80+ but the volumetric condensation rate for the tests was 1.63 times the CE System 80+ rate. For these tests, a full-cone spray nozzle (Lechler model 461.148 having a 120° nozzle spray angle) was installed at the top of the dome. When the water spray mass flux was scaled to the operation of both CE System 80+ independent spray trains, a high-capacity full-cone spray nozzle (Lechler model 461.206 having a 90° nozzle spray angle) was installed at the top of the dome.

The experiments were conducted under conditions scaled to be nearly prototypical of those expected in hypothetical severe accidents in the CE System 80+ containment. The quantities that were scaled included the spray mass flux and hydrogen mass flow rates for stratified tests.

Eleven combustion tests were conducted: 8 were well-mixed tests and 3 were stratified tests. The main parameters that were tested in these experiments were hydrogen concentration, steam condensation rate as determined by the water spray flow rate, and igniter location. Table 1 shows the initial conditions of the tests; Table 2 shows the conditions at the first burn; and Table 3 shows the conditions at the end of the tests after the water sprays were turned off. The first test, HIT-1, was performed to collect data and test gas grab sample techniques and igniter design. After test HIT-1, the time to purge gas sample lines was increased and gas sample data were considered more accurate. Results for HIT-7 were influenced by a small leak (~0.003 MPa/hr) in the facility since the test was conducted over a long (23-hour) period. This leak did not have any significant influence on the results of the other tests because the test times were short (~20 minutes). Results from HIT-7 were analyzed assuming the steam was saturated and that the remaining constituents leaked out in proportion to their composition in the facility. A steam condensation experiment was conducted before the combustion tests to determine the steam condensation rate owed to water sprays in a nonflammable helium-air-steam mixture. Three mixing tests were performed at ambient conditions to determine if stratified conditions could be created for the combustion tests and to measure the mixing time with the water sprays.

Results

In all of the combustion tests, one or more relatively slow deflagrations were recorded: no detonations were observed. The combustion behavior of the well-mixed tests can be loosely categorized into one of two types: (1) multiple deflagrations with relatively small pressure rises and (2) a single deflagration with a pressure rise greater than those observed in the multiple deflagrations but lower than the theoretical adiabatic constant volume value. The combustion behavior of the stratified tests was similar to that of the well-mixed tests. This is because the sprays were very effective in mixing the hydrogen before the mixture became flammable.

For mixture compositions that have directionally dependent flammability limits, multiple deflagrations with relatively small pressure rises were observed. Within the range of experimental uncertainty, these mixtures lie below the "nose" of the flammability limits curve. Combustion behavior was not dependent on igniter location. Since these mixtures have directionally dependent flammability limits (upward, horizontal, and downward propagation criteria), the upward propagation criterion was satisfied first and a marginal burn occurred. The combustion was incomplete and the pressure rise was small (1-10 kPa). The partial burn temporarily inerted the mixture, which did not become flammable again until additional steam was condensed by the water sprays. The partial burns were responsible for the multiple deflagrations. This behavior is illustrated in Figure 2, which shows the results of HIT-2, a well-mixed test with 13.5% hydrogen on a dry basis. The pressure rises were larger (up to 30 kPa) in the well-mixed tests with 13.5% hydrogen on a dry basis but having larger steam condensation rates. Multiple deflagrations, however, were again observed. This behavior is illustrated in Figure 3, which shows the results of

HIT-10, a well-mixed test with 13.5% hydrogen on a dry basis and a spray flow rate nominally twice that of test HIT-2.

For mixture compositions that do not have directionally dependent flammability limits, a single deflagration with a pressure rise greater than those observed in the multiple deflagrations but lower than the theoretical adiabatic constant volume value was observed. Once these mixtures were rendered flammable by the water sprays, the flame could propagate in all directions, which consumed nearly all of the hydrogen. This resulted in a single deflagration with a pressure rise closer to the adiabatic constant volume value. The increase in pressure did not achieve peak theoretical values because of heat losses from the relatively slow deflagration near the flammability limit. This behavior is illustrated in Figure 4, which shows the results of HIT-3, a well-mixed test with a stoichiometric mixture (29.5% hydrogen on a dry basis).

Three stratified tests were conducted with different spray flow rates but all having a near stoichiometric mixture in the upper part of the vessel. The target condition for each test was a stratified mixture with 29.5% hydrogen (dry basis) in the upper part of the vessel but having an equivalent well-mixed concentration of 13.5% hydrogen (dry basis). Different spray flow rates were used: 0.0 kg/second, 1.35 kg/second, and 3.17 kg/second. The behavior of these tests was similar to that of the comparable well-mixed tests with 13.5% hydrogen. This behavior is illustrated in Figure 5, which shows the results of HIT-6, a stratified test with approximately 24% hydrogen (dry basis) in the upper part of the vessel. The sprays rapidly mixed the hydrogen in the vessel prior to the mixture becoming flammable so that the hydrogen was essentially uniform at 13.5% (dry basis) before ignition.

The mixing time was determined using newly developed hydrogen microsensors which provided spatially resolved real-time continuous output of hydrogen concentrations. A stratified test was conducted at ambient conditions with approximately 19% hydrogen (dry basis) in the upper part of the vessel. The hydrogen was mixed by water sprays with a flow rate of 1.58 kg/second. As shown in Figure 6, the mixing time was approximately 20 seconds for most of the vessel and less than 1 minute even for the lowest levels.

Conclusions

Multiple deflagrations with relatively small pressure rises or single deflagrations with pressure rises greater than those observed in the multiple deflagrations but lower than the theoretical adiabatic constant volume values were observed when igniters were on during the entire experiment. This is because ignition occurred near the flammability limit and the combustion time was fast relative to the time to condense steam. Detonations or other forms of energetic combustion (flame acceleration or DDT) were not observed in hydrogen-air-steam mixtures which were initially non-flammable owing to steam dilution but were rendered flammable by steam condensation caused by water sprays. Mixtures with hydrogen concentrations above approximately 24% (dry basis) exhibited a single deflagration with a relatively large pressure rise. Such mixtures cannot exist under globally well-mixed conditions in the CE System 80+ or AP600 containments. These containments will have approximately 13.5% hydrogen (dry basis) on a well-mixed basis, assuming a 100% metal-water reaction of the active cladding. If the hydrogen accumulates above the operat-

ing deck level, the concentration could be as high as 19.3% (dry basis) on a well-mixed basis above the floor of the operating deck. Mixtures having 19.3% hydrogen or less exhibited multiple deflagrations with relatively small pressure rises.

Thermal glow plugs functioned as intended: ignition occurred near the flammability limits. The flammability limits for mixtures with water sprays were similar to previously obtained flammability limits for quiescent hydrogen-air-steam mixtures.

Stratified mixtures and well-mixed mixtures yielded similar combustion pressures owing to the effective mixing by water sprays. Stratified hydrogen in the Surtsey vessel was made essentially uniform by the water sprays in less than 1 minute.

Acknowledgments

The support and guidance of the NRC program managers A. Malliakos and C. Tinkler is gratefully acknowledged. The authors would like to thank M. Allen and M. Pilch for their useful discussions during the experimental program. The authors express their gratitude to R. Nichols, the lead mechanical engineer and site manager; to M. Oliver and T. Thornhill, who were the electronics and instrumentation engineers; to J. Ross and T. Covert, who were the mechanical technicians; and R. Hudgens and J. Andazola, who provided technical support during the experiments.

This work was supported by the U.S. Nuclear Regulatory Commission and performed at Sandia National Laboratories, which is operated for the U. S. Department of Energy under Contract No. DE-AC04-94AL85000.

References

1. Ratzel, A. C., "Data Analyses for Nevada Test Site (NTS) Premixed Combustion Tests," Sandia National Laboratories, SAND85-0135, NUREG/CR-4138, Albuquerque, NM, May 1985.
2. Nelson, L. S., "Behavior of Hydrogen Igniters During Operation of Water Sprays in Containment," presented at the Twelfth Water Reactor Safety Research Information Meeting, Gaithersburg, MD, October 22-26, 1983, published as an NRC report, NUREG/CP-0058, Vol. 3, pp. 371-387, Washington, DC, January 1985.
3. Shebeko, Yu. N., Tsarichenko, S. G., Eremenko, O. Ya., Keller, V. D., and Truney, A. V., "Combustion of Lean Hydrogen-Air Mixtures in an Atomized Water Spray," *Fizika Gorenija i Vzryva*, Vol. 26, No. 4, pp. 58-61, July-August 1990.
4. Tamm, H., Ungurian, M., and Kumar, R. K., "Effectiveness of Thermal Ignition Devices in Rich Hydrogen-Air-Steam Mixtures," Electric Power Research Institute report, EPRI NP-5254, AECL-8363, Palo Alto, CA, July 1987.
5. CESSAR Design Certification, Appendix 19.11K, "Assessment of the System 80+ Hydrogen Mitigation System for Application in a Severe Accident Environment," ABB-Combustion, Combustion Engineering, Inc., Windsor Locks, CT, October 1993.

Table 1. Initial conditions in the HIT experiments

		HTT-1	HTT-2	HTT-3	HTT-4	HTT-5	HTT-6	HTT-7	HTT-8	HTT-9	HTT-10	HTT-11
Test Date	1-12-94	1-20-94	1-26-94	2-9-94	2-16-94	2-21-94	4-13-94	4-20-94	5-2-94	5-9-94	5-16-94	
Hydrogen condition wm - well-mixed, strat - stratified	wm	wm	wm	wm	wm	strat	strat	wm	wm	wm	wm	strat
Vessel pressure (MPa)	0.4642	0.4351	0.4686	0.4386	0.4142	0.4552	0.3279	0.4608	0.4040	0.3926	0.4427	
Vessel gas temperature (K)	416	413	412	419	411	416	401	408	405	406	411	
Vessel gas moles (g • moles)	7934	7491	8083	7443	7162	7771	5808	8030	7098	6882	7653	
Gas composition (mole %)	Steam 26.5 Air 4.2 Wet basis-well mixed H ₂ 4.4	69.3 28.0 25.9 11.0 4.4	67.6 28.1 28.1 4.4	63.1 29.3 29.3 7.0	67.5 26.9 26.9 4.2	63.7 36.2 36.2 5.8	68.9 26.1 26.1 8.5	58.0 29.6 29.6 7.5	65.4 49.6 49.6 7.5	62.9 30.6 30.6 4.9	64.5 28.0 28.0 4.5	67.5
Gas composition (mole %)	Steam 86.5 Air 13.5 Dry basis-well mixed H ₂ 13.6	00.0 86.4 70.3 29.7	00.0 86.5 80.7 13.5	00.0 86.5 86.2 19.3	00.0 86.5 86.2 13.5	00.0 86.5 75.3 13.8	00.0 75.3 79.8 24.7	00.0 75.3 79.8 24.7	00.0 79.8 86.2 20.2	00.0 86.2 86.2 20.2	00.0 86.3 86.3 13.8	00.0
Spray nozzle model #461	.148	.148	.148	.148	.148	.148	.148	.148	.148	.148	.148	.148
Spray drop Sauter mean diameter (μ m)	850	850	850	850	850	850	n/a	850	850	850	850	850
Spray flow rate (kg/s)	1.68	1.35	1.61	1.63	1.46	1.48	n/a	1.45	1.45	1.45	1.45	1.45
Spray temperature (K)	332	296	291	289	298	292	n/a	289	289	289	291	291
Igniter temperature (K)	1072	1307	1202	1204	1224	1290	1235	1268	1251	1251	1240	
Igniter location (m) (from floor/from wall)	4.5/1.8 3.7/0.5 1.1/1.8	3.1/1.8 0.6/1.8	3.1/0.5 0.6/1.8	3.1/0.5 1.1/1.8	4.5/1.8 3.7/0.5	3.7/0.5 3.7/0.5	3.7/0.5 3.7/0.5	3.7/0.5 3.7/0.5	3.7/0.5 3.7/0.5	3.7/0.5 3.7/0.5	3.7/0.5 3.7/0.5	
Sursey vessel volume (m ³)												59.1

Gas concentrations at initial conditions are based on the calculated amounts of air and hydrogen gases that were added; steam concentrations are based on the ideal gas law.

Table 2. Conditions at the first burn in the HIT experiments.

	HIT-1	HIT-2	HIT-3	HIT-4	HIT-5	HIT-6	HIT-7	HIT-8	HIT-9	HIT-10	HIT-11
Elapsed time (min)	10.9	9.3	3.0	4.3	5.5	5.8	8.8 hr	4.0	0.8	1.3	1.7
Vessel pressure (MPa)	0.2321	0.2517	0.3382	0.2700	0.2436	0.2682	0.1886	0.3302	0.3093	0.2645	0.2697
Vessel gas temperature (K)	384	385	400	391	386	388	367	391	391	384	385
Vessel gas moles (g • moles)	4299	4650	6018	4904	4486	4909	3653	5997	5625	4899	4976
Gas composition * (mole %)	Steam N ₂ O ₂ H ₂	43.4 38.3 10.3 7.7	47.9 35.2 9.4 7.1	50.5 27.2 7.3 14.7	50.7 33.3 8.9 6.7	42.0 36.5 9.8 11.2	50.8 33.2 8.9 6.7	43.3 38.2 10.3 7.8	53.7 27.2 7.3 11.4	53.3 29.1 7.8 9.4	50.2 33.5 9.0 6.9
Wet basis-well mixed											33.7 9.0 6.8
Gas composition (mole %)	Steam N ₂ O ₂ H ₂	00.0 67.5 18.1 13.5	00.0 54.9 18.1 13.6	00.0 67.5 14.7 29.7	00.0 63.0 18.1 13.5	00.0 67.5 16.9 19.3	00.0 67.3 18.1 13.5	00.0 58.8 15.8 13.8	00.0 62.3 16.7 24.7	00.0 67.3 16.7 20.2	00.0 67.4 18.1 13.8
Dry basis-well mixed											13.7
Pressure increase at 1st burn (kPa)	1	3	604	10	11	13	7	515	164	29	29
Temperature increase at 1st burn (K)	1	1	447	5	7	4	3	457	122	15	10

- Gas concentrations at first burn conditions are based on the assumption that the noncondensable gas moles remain constant at their initial values and steam concentrations are based on the ideal gas law. Wet basis concentrations for HIT-7 were estimated assuming saturated steam conditions. For details on HIT-7, see text in Program Description section. Gas compositions do not include trace air constituents.

Table 3. Final conditions in the HIT experiments

	HIT-1	HIT-2	HIT-3	HIT-4	HIT-5	HIT-6	HIT-7	HIT-8	HIT-9	HIT-10	HIT-11
Elapsed time (min)	18.9	23.7	18.2	19.3	21.5	20.9	23 (fr)	21.7	9.8	9.8	9.9
Vessel pressure (MPa)	0.1697	0.1227	0.1270	0.1261	0.1080	0.1225	0.1083	0.1728	0.1150	0.1198	0.1212
Vessel gas temperature (K)	371	338	346	326	344	338	333	365	330	331	334
Vessel gas moles (g • moles)	3248	2583	2611	2750	2232	2580	2313	3366	2478	2575	2579
Gas composition * (mole %)	Steam	27.9	17.3	32.3	23.4	6.9	19.2	18.4	47.2	21.1	18.2
	N ₂	50.6	63.3	62.7	59.4	73.4	63.2	57.8	48.5	66.1	63.8
Wet basis-well mixed	O ₂	12.3	14.4	3.9	12.3	12.3	12.8	14.8	3.9	9.4	13.3
	H ₂	6.8 ⁺	4.2	0.3	3.8	5.4	4.0	7.8 ⁺	0.2	3.5	3.9
Gas composition (mole %)	Steam	00.0	00.0	00.0	00.0	00.0	00.0	00.0	00.0	00.0	00.0
	N ₂	70.2	76.5	92.5	77.6	78.8	78.2	70.8	91.8	83.8	77.9
Dry basis-well mixed	O ₂	17.1	17.4	5.7	16.0	13.3	15.8	18.1	7.4	12.0	16.3
	H ₂	9.4	5.1	0.5	5.0	5.9	5.0	9.5	0.3	4.4	4.7
Oxygen burned (g • moles)		41	67	338	102	163	107	21	306	205	97
Hydrogen burned (g • moles)		109	220	876	223	381	223	117	680	446	236
2 x O ₂ burned / H ₂ burned		0.75	0.61	0.77	0.91	0.86	0.96	0.37	0.90	0.92	0.82
											0.94

* Gas concentrations at final conditions are based on the gas grab sample results; steam concentrations are based on the ideal gas law. Gas compositions in HIT-1 were influenced by the gas sample procedure (see text in Program Description section for more details). Wet basis concentrations for HIT-7 were estimated assuming saturated steam conditions. Gas compositions do not include trace air constituents.

⁺ Burns were still occurring at the end of the test. They were confined to a region near the igniter and, for HIT-7 in particular, there was no mixing caused by water sprays.

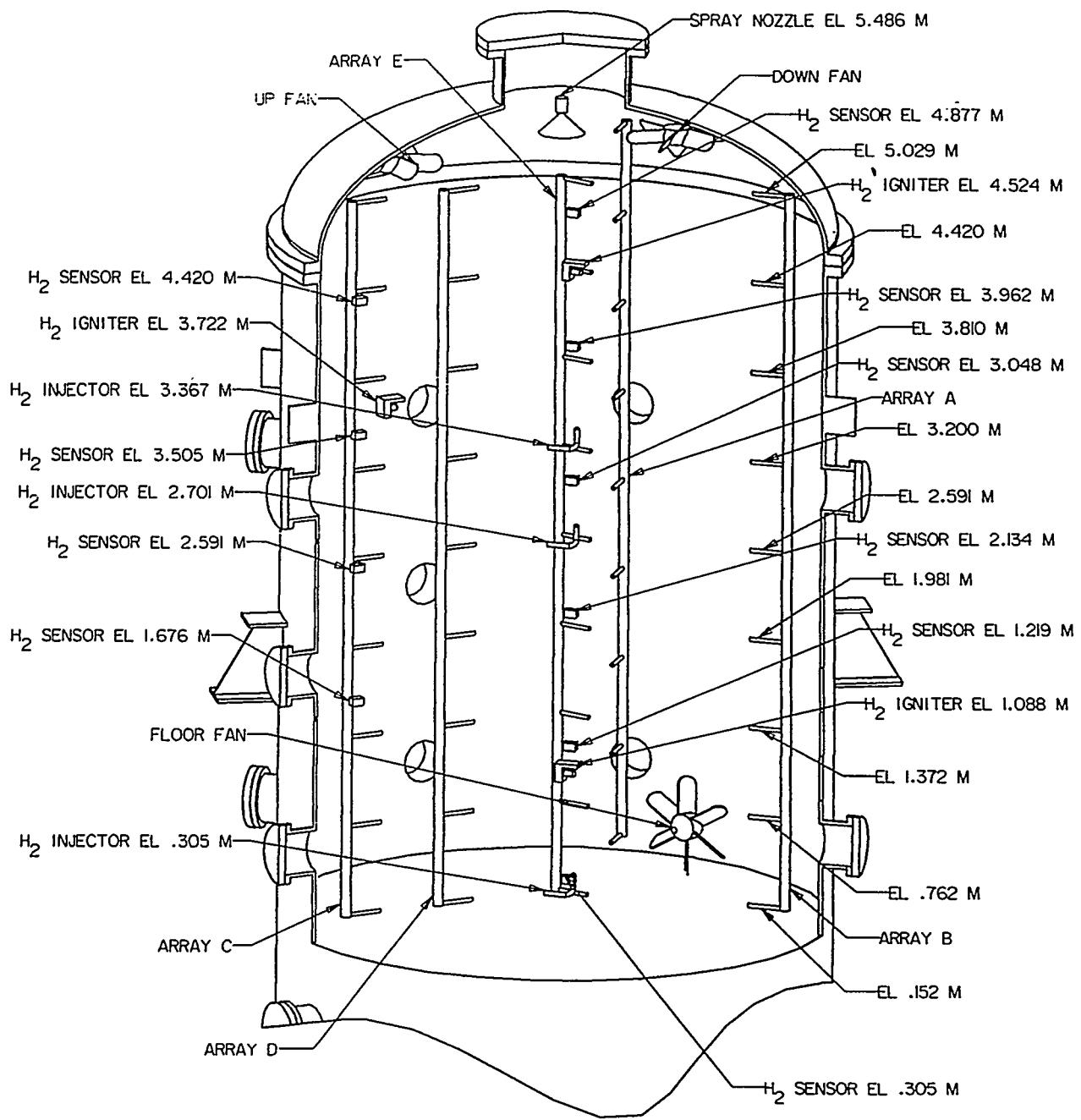


Figure 1. Schematic of the Surtsey facility.

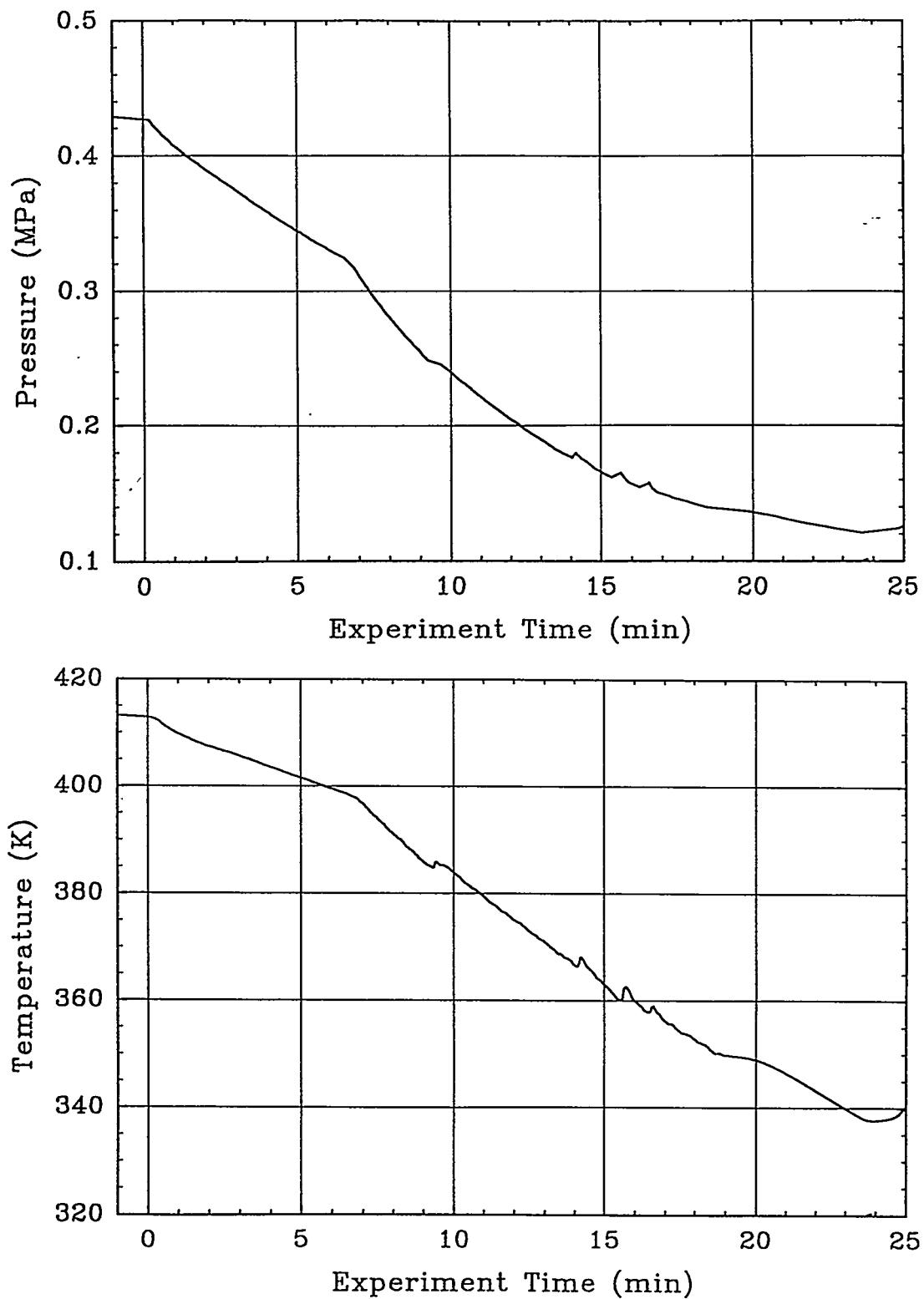


Figure 2. Pressure and temperature from experiment HIT-2.

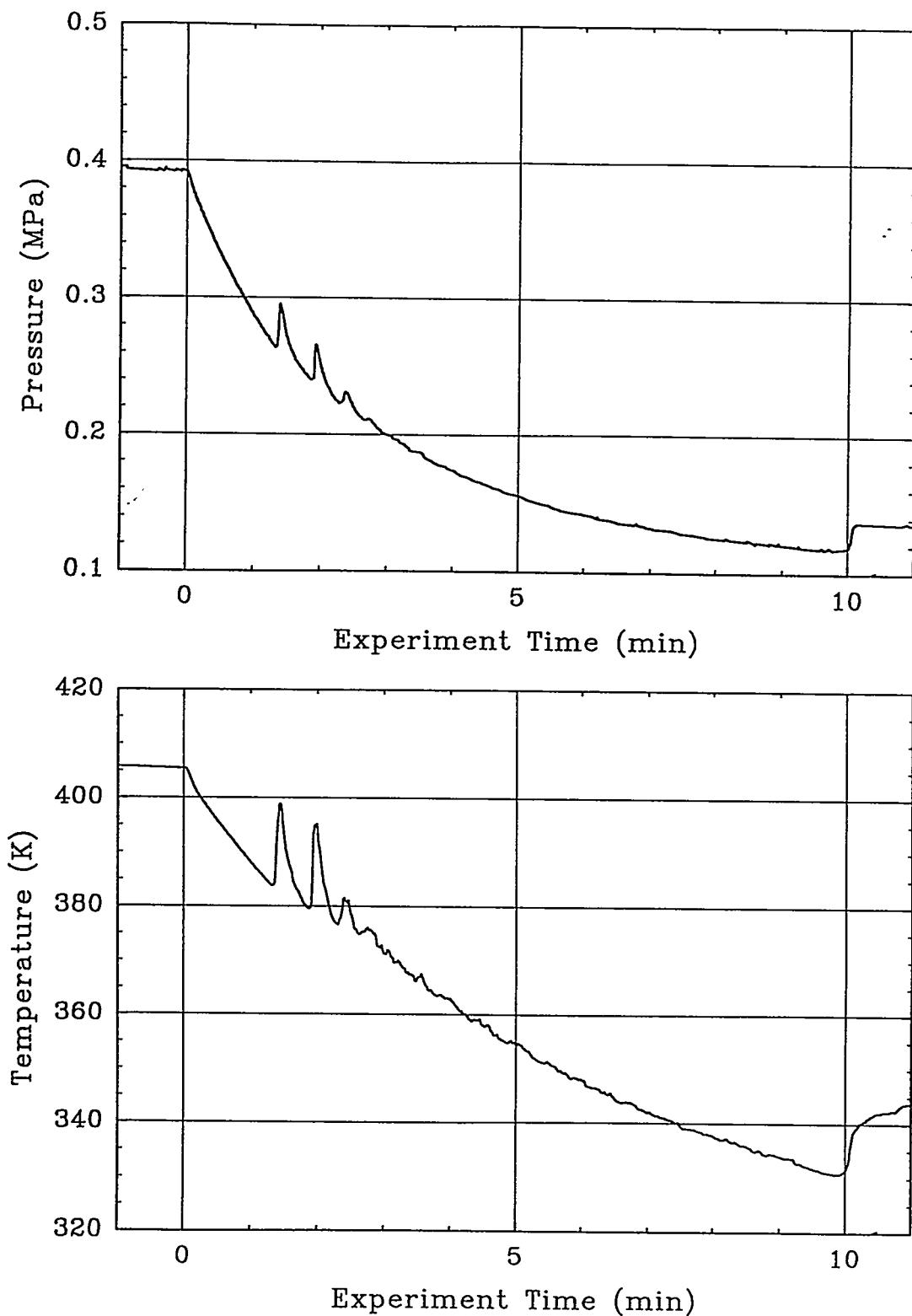


Figure 3. Pressure and temperature from experiment HIT-10.

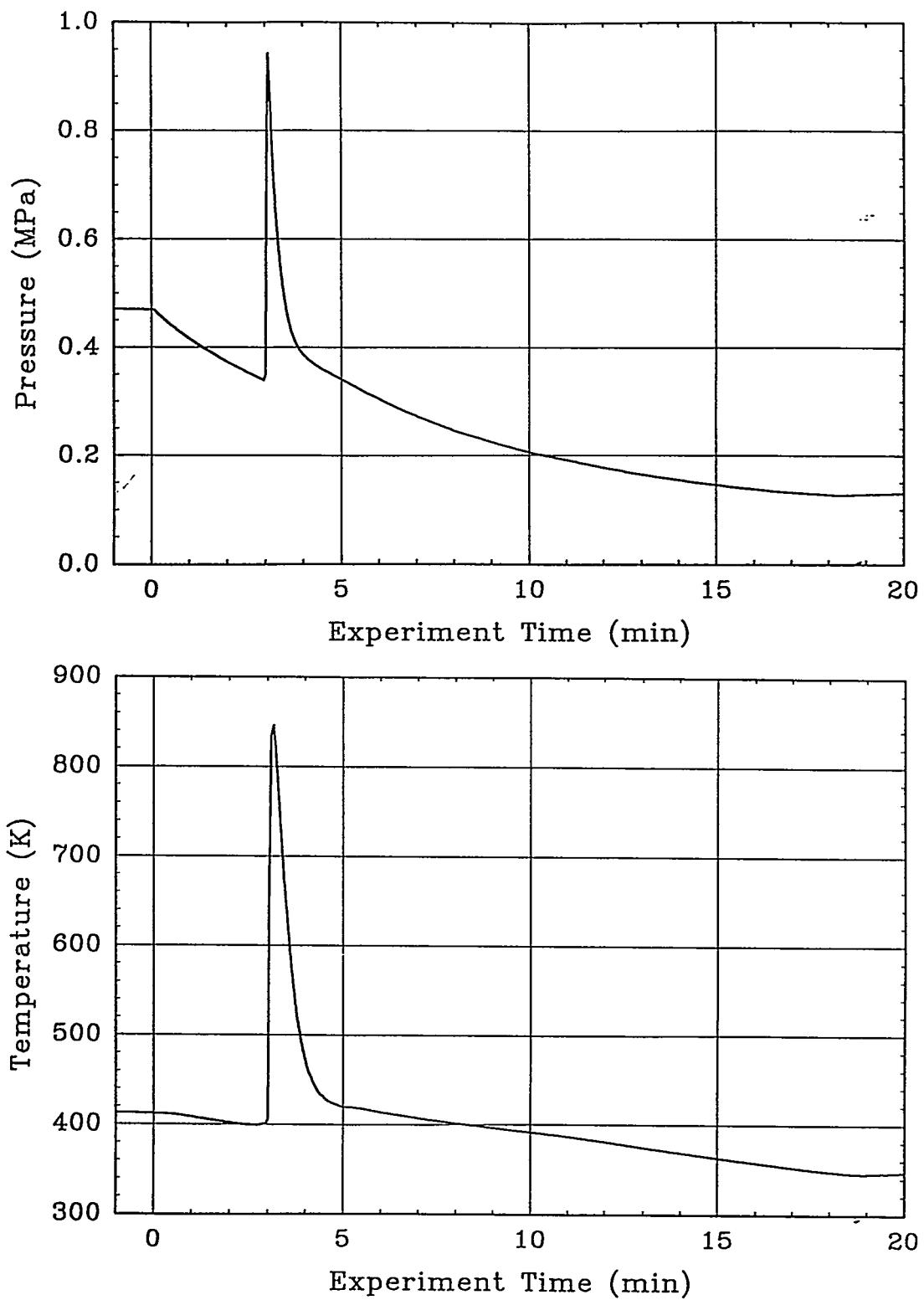


Figure 4. Pressure and temperature from experiment HIT-3.

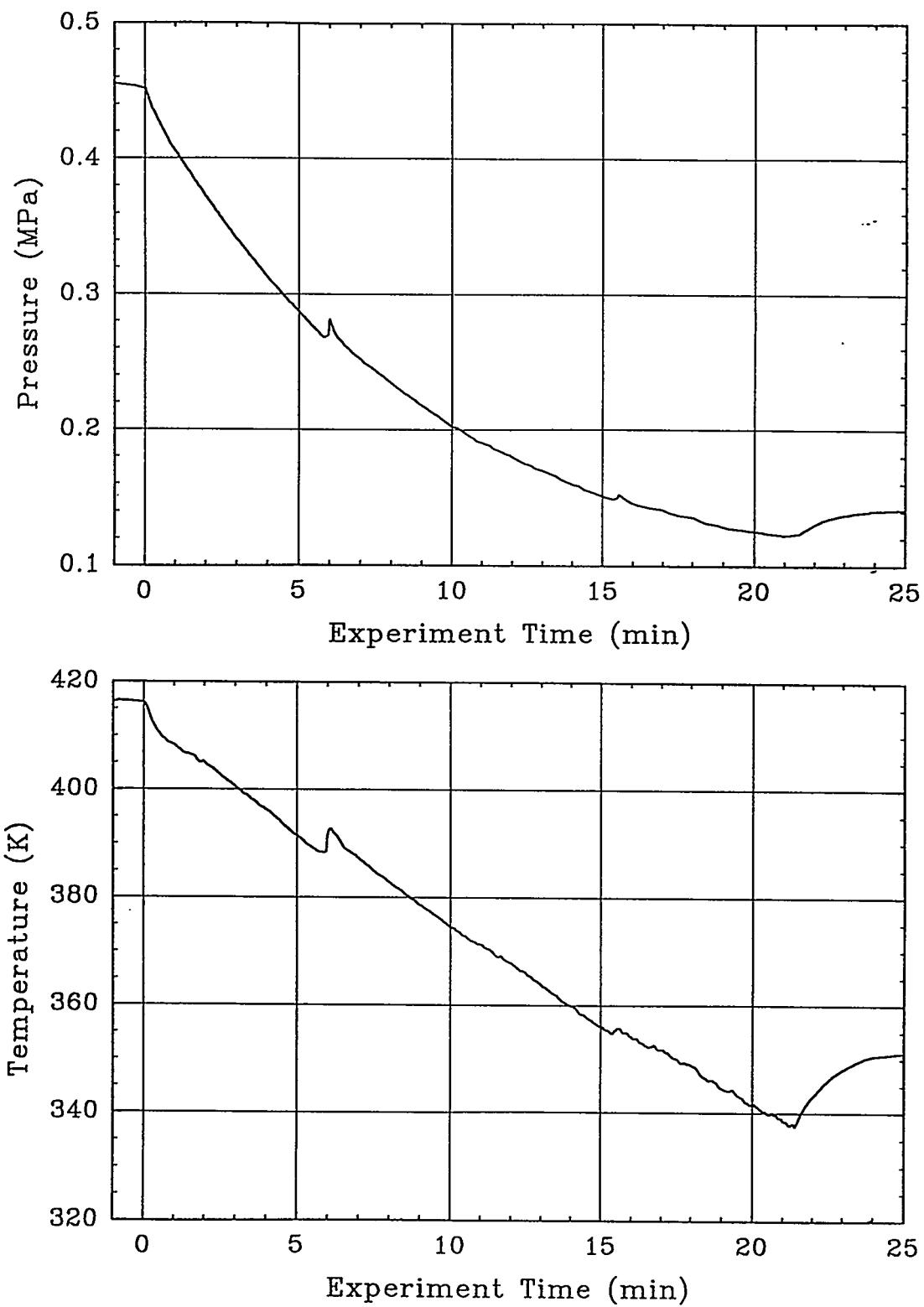


Figure 5. Pressure and temperature from experiment HIT-6.

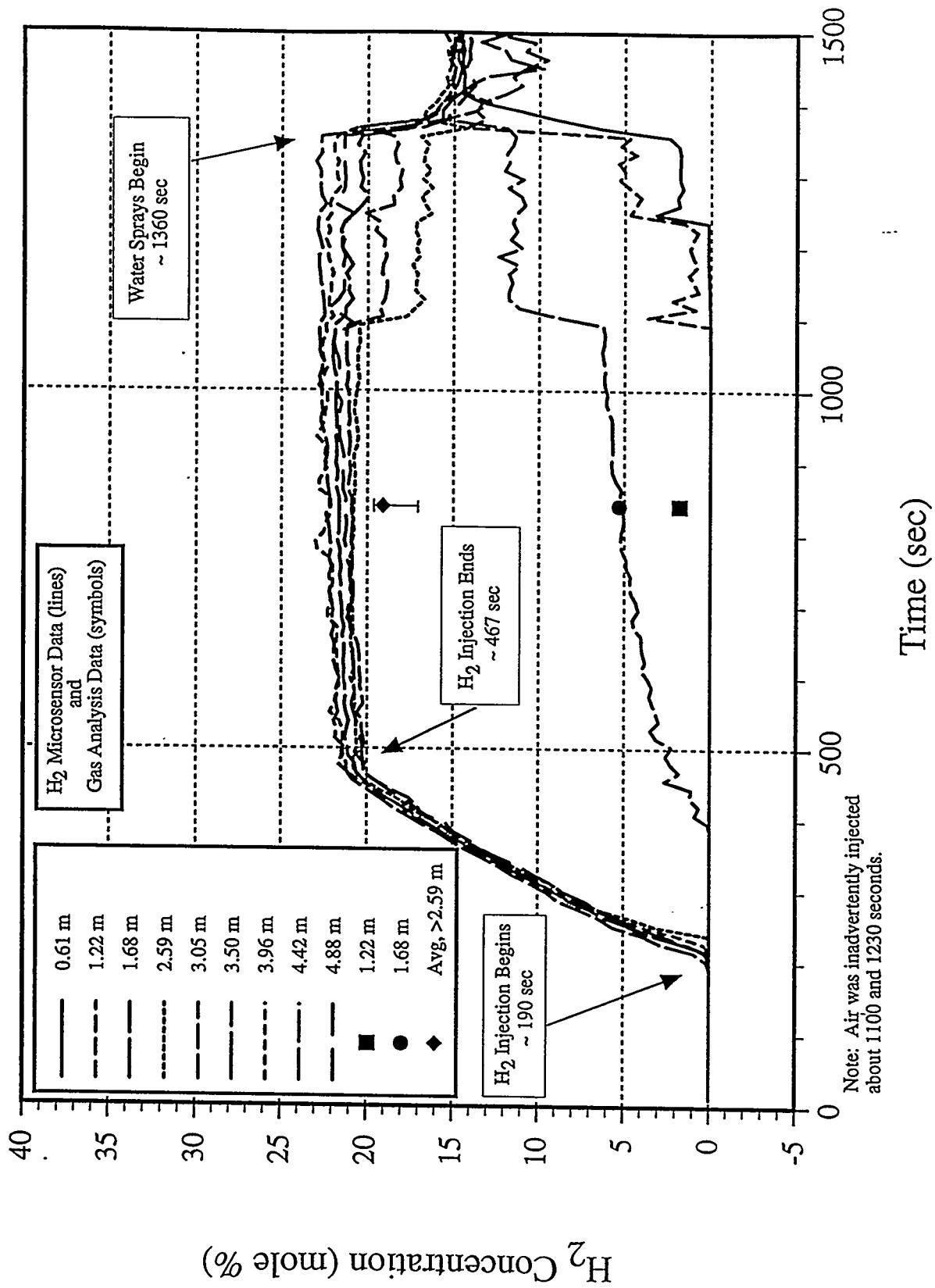


Figure 6. Hydrogen concentration measurements by microsensors and gas grab samples in a stratification test.