
Platinum Catalytic Igniters for Lean Hydrogen-Air Mixtures

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Sandia National Laboratories

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U.S. Nuclear Regulatory
Commission

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ABSTRACT

We have developed a prototype catalytic igniter for lean hydrogen-air mixtures that could have important applications in nuclear reactor safety. The igniter has two useful characteristics related to these applications: it requires no electrical power and it can ignite mixtures as lean as 5.5% hydrogen. The ignition induction time ranges from 20 s to 400 s depending on the hydrogen concentration, gas flow velocity, gas temperature and relative humidity of the gas mixture. Induction times are shorter for mixtures with higher hydrogen concentrations, higher flow velocities, higher gas temperatures and lower relative humidity. The igniter operates successfully under conditions that may be present during a loss-of-coolant accident (LOCA) at a light water nuclear reactor. In the event of a LOCA, large quantities of hydrogen may be produced very rapidly and the catalytic igniter could provide a means of igniting it before dangerously high concentrations are attained; even in the event that electrical power required for conventional igniters is not available. The igniter has not been tested under all possible LOCA conditions. High gas velocities, water spray, steam and iodine-containing compounds may be present during a LOCA and will defeat the prototype igniter. However, shielding and semi-permeable coatings on the igniter could overcome these difficulties. A U. S. Patent has been granted for the catalytic igniters described herein (U. S. Patent No. 4,741,879).

CONTENTS

	<u>Page</u>
Executive Summary.....	1
1. Introduction.....	2
2. Experimental Apparatus and Procedure.....	2
3. Igniter Design Optimization.....	5
4. Performance of Prototype Igniter.....	10
4.1 Hydrogen Concentration.....	10
4.2 Flow Velocity.....	10
4.3 Humidity.....	11
4.4 Temperature.....	11
4.5 Water Spray.....	11
5. Principle of Operation.....	16
6. Expected Operation of Catalytic Igniter in a LOCA.....	18
6.1 Temperature.....	18
6.2 Flow Velocity.....	19
6.3 Hydrogen Concentration.....	19
6.4 Water Spray, Steam, and Fog.....	19
6.5 Contaminated Atmosphere.....	19
7. Further Improvements in Igniter Design.....	20
7.1 Wire.....	20
7.2 Platinum-Coated Substrate.....	20
8. Further Tests.....	20
9. References.....	21
Appendix A - Tests of the Catalytic Igniter in the Fully Instrumented Test System.....	22

LIST OF FIGURES

<u>Figure</u>		<u>Page</u>
1	Apparatus used to test catalytic igniter performance	4
2	Temperature rise of catalytic igniter in a typical test	4
3	First successful catalytic igniter design	6
4	Various prototype catalytic igniter designs	7
5	Optimized catalytic igniter design	8
6	Device to test thermal boosting of wires	8
7	Device to test thermal boosting by substrate	9
8	Effect of gas flow rate on igniter performance	12
9	Effect of humidity on igniter performance	13
10	Effect temperature on igniter performance	14
A1	Pressure rise after ignition by glowplug and catalytic igniter	24

EXECUTIVE SUMMARY

A potentially hazardous hydrogen-air mixture may be produced in the reactor containment of some types of nuclear reactors during a loss-of-coolant accident (LOCA). This mixture is flammable if the hydrogen concentration exceeds 4.1 vol. % and may be explosive if the concentration exceeds 13 vol. %. One way of reducing the risk of explosion or fire damage is to intentionally burn this mixture at sufficiently low hydrogen concentrations (5 - 7 vol. % hydrogen), so that little if any damage to the reactor containment building will occur. Current implementations of this hydrogen mitigation strategy make use of electrically heated glowplugs or coils. However, in the event of a serious accident, the electrical power may be interrupted, disabling these igniters. Thus, nonpowered igniters could provide a valuable safety backup to existing igniter systems.

We have developed a catalytic igniter which can operate under conditions which may prevail during a LOCA and that does not require an external source of power of any kind. The igniter is composed of a catalytic substrate and several platinum wires (0.0123-cm diameter, 4.0-cm long) which project into the unreacted gas. The substrate is an alumina honeycomb (4.4-cm diameter, 3.0-cm high, with 0.2-cm diameter cells) that is coated with high surface area platinum particles to about 1.7 weight % platinum. The igniter operates by catalyzing the exothermic surface reaction between hydrogen and oxygen. If the hydrogen concentration is sufficiently high, heat is generated rapidly enough by the honeycomb to raise the temperature of the wires above 80° C. Above this temperature, the catalytic activity of the wires is sufficient that they warm further -- to the ignition temperature of the mixture (around 585° C).

The atmosphere in the containment structure during a LOCA might be characterized by high gas temperatures, high flow velocities (up to 1000 cm/s), high relative humidity (up to 100%), steam, water spray, the presence of volatile fission products (Cs, I₂, etc.) and CO and CO₂. These conditions may cause igniters to fail, and we have tested the catalytic igniter under some of these.

In our laboratory tests of the catalytic igniter, gas-phase ignition occurs after an induction time (ignition delay time) of 20-400 s, the length of which depends on the hydrogen concentration, gas flow velocity, gas temperature, and the relative humidity. Induction times were measured for hydrogen concentrations in the range of 5.5- 11.0 vol. %, gas flow velocities between 1.7 and 19.5 cm/s, gas temperatures between 20° and 65° C, and relative humidities between 5 and 98%. Induction times are shorter for mixtures with higher hydrogen concentrations, higher flow velocities, higher gas temperatures and lower relative humidity. The igniter successfully ignited static mixtures as lean as 6.5 vol.% H₂, humidified mixtures as lean as 10 vol. % H₂ and humidified flowing mixtures as lean as 6.3 vol. % contained in a 5.6-m³ test vessel. Liquid water defeats the igniter. However, when a wet igniter is dried, it operates normally indicating that liquid water blocks the catalytic sites but does not poison them. The igniter operates repeatedly. Some of the igniters used in this study were cycled tens of times without any sign of reduced performance. This is a desirable characteristic because hydrogen produced during a LOCA may continue to be produced after the first ignition event and repeated ignitions may be required.

We have not tested the catalytic igniter under all possible accident scenarios that may prevail during a LOCA. However, our initial tests indicate that a nonpowered igniter for reactor safety applications is feasible and constitute "proof of principle." Furthermore, we believe that a practical safety device based on our prototype design may be developed with modest additional effort.

1. INTRODUCTION

During a loss-of-coolant accident (LOCA) in a light-water nuclear reactor (LWR), there is the potential for production of large quantities of hydrogen gas due to the reaction of water and steam with the fuel rod cladding¹. The hydrogen production rate and quantity may be such that a flammable mixture could be produced within hours as the hydrogen mixes with the air in the containment building. If this mixture is ignited at sufficiently high hydrogen concentrations, the structural integrity of the containment might be compromised. Serious safety and radiological hazards could result. Therefore, in the event of a LOCA, methods to either make the mixture non-flammable or reduce the hydrogen concentration are needed. One method for reducing the hydrogen concentration is to purposely ignite the mixture at hydrogen levels low enough to prevent serious damage. Although a safe upper limit has not been established for the hydrogen concentration, it is less than 13%, the concentration above which a transition from normal burning to detonation could occur. The deliberate-ignition approach has already been implemented at several LWR sites by using electrically heated glowplugs located at various positions within the containment building. One disadvantage of glowplugs, however, is that they require a continuous source of electrical power which may be lost during a serious accident. And, even with battery backup (which has not been fielded), an accident may cut the power cables to the igniters. Thus, a device that ignites lean hydrogen-air mixtures in a safe range of hydrogen concentrations (5-7%) and does not require an external source of power could have important applications as a nuclear reactor safety device.

The conditions inside the containment building in the initial stages of a LOCA may include a wide temperature range (0 - 200° C), wide velocity range (0 - 1,000 cm/s)^{2,3}, high humidity, the presence of steam, water spray, carbon monoxide, and iodine- and cesium-containing compounds. The igniter must operate under these conditions and have the capability of repeated operation in the event of repeated hydrogen buildup.

We report here the successful development of a prototype catalytic igniter which has some of the operating characteristics desired of a non-powered igniter. We present the results of experiments aimed at optimizing the igniter performance, and typical performance characteristics of the prototype device. Finally, we present a discussion of expected igniter performance in a LOCA environment, additional tests that should be made and a discussion of those design parameters which should be considered in future igniter designs. An appendix is included that gives the test results for the ignition of large static mixtures.

2. EXPERIMENTAL APPARATUS AND PROCEDURE

Several catalytic igniters were constructed and then tested in the apparatus shown schematically in Figure 1. The ignition delay time, that is, the time required for the igniter to ignite the mixture, was taken as a measure of the igniter's performance. Ignition was detected by a sudden drop in the temperature of the catalytic module as indicated by an attached thermocouple or infrared detector. The test apparatus consisted of a water-cooled burner to which was attached a quartz chimney. The premixed hydrogen-air mixture was directed through the burner surface into the chimney and onto the igniter. The burner was not operated as a burner *per se* but as a stop for the flame front which propagates away from the igniter toward the burner surface when ignition occurs.

The hydrogen concentration was controlled by regulating the flow of air and hydrogen supplied to the burner. Tylan mass flow meters were used to monitor the mass flows of hydrogen and air. The meters were calibrated by setting the flow control valve for a specific flow rate while the flow was directed through a wet test meter which measured the total volume of gas delivered. The time required for the delivery of a specific volume was then measured. The volume delivered at standard conditions was calculated and divided by the elapsed time to give the flow rate at standard temperature and pressure. The meter was then adjusted to read this value. The estimated absolute accuracy of the measured flow is $\pm 3\%$.

The lowest ignitable hydrogen concentration is apparatus dependent. Thus, it was desirable to determine the lowest hydrogen concentrations that could be ignited in our apparatus before testing the catalytic igniter. This was done using a spark igniter and a gas flow velocity of 5.7 cm/s. The leanest mixture which could be ignited in our test apparatus was 5.1% for upward flame propagation (burner in inverted configuration) and 9.3% for downward flame propagation (normal burner configuration). This compares with 4.1% and 9.0%, respectively, for measurements made in a standard apparatus with static premixed gases.⁴

The temperature of the igniter honeycomb was monitored quantitatively with a chromel/alumel thermocouple and qualitatively with an infrared sensitive detector. Because the infrared energy emitted by a hot body is proportional to its temperature to the fourth power, the infrared detector is most sensitive to high temperatures and produces a large output change for a small change in temperature. Because of this, the infrared detector provided a good indication of the time of ignition by a sudden drop in the detector output. When gas-phase ignition occurred, the temperature of the igniter dropped because the flame front moved away from the igniter to the burner surface. The rapid response time of the infrared detector aided the determination of when ignition occurred.

In a typical test sequence, the igniter to be tested was placed in the chimney with the valve to the vent open and the valve to the burner closed. Next, the flow rates of hydrogen and air were adjusted to produce the desired hydrogen concentration. To initiate the test, the valve to the burner was opened quickly, and at the same time the valve to the vent was closed. After an induction time of 30 to 400 s, the catalytic igniter ignited the hydrogen-air mixture and the flame front traveled from the point of ignition to the burner surface where it was stabilized. As soon as ignition was detected by a sudden drop in the temperature of the module, the positions of the valves were reversed to extinguish the flame. For safety, the apparatus was located within the flow field of a hood which was vented to the outside so that any unburned hydrogen was diluted to an unignitable concentration and removed from the laboratory.

A trace of the temperature of the igniter honeycomb as measured with the thermocouple is shown in Figure 2 for a typical experiment. The infrared detector signal is also shown in Figure 2. The temperature drop after ignition is clearly evident. In addition to the substrate temperature measurements, thermocouple temperature measurements were also made in the region a few millimeters above the catalytic substrate. For these measurements, ignition of the gas-phase mixture was indicated by a rise in temperature.

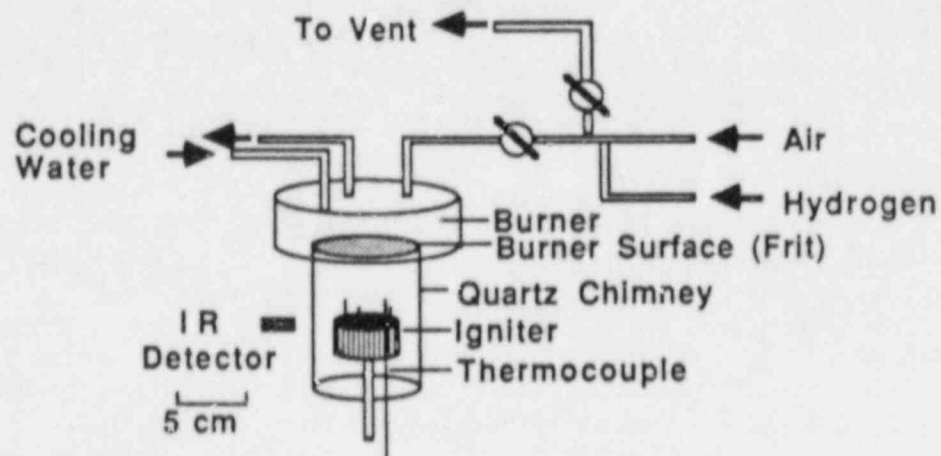


Figure 1. Apparatus to test catalytic igniter performance (inverted configuration).

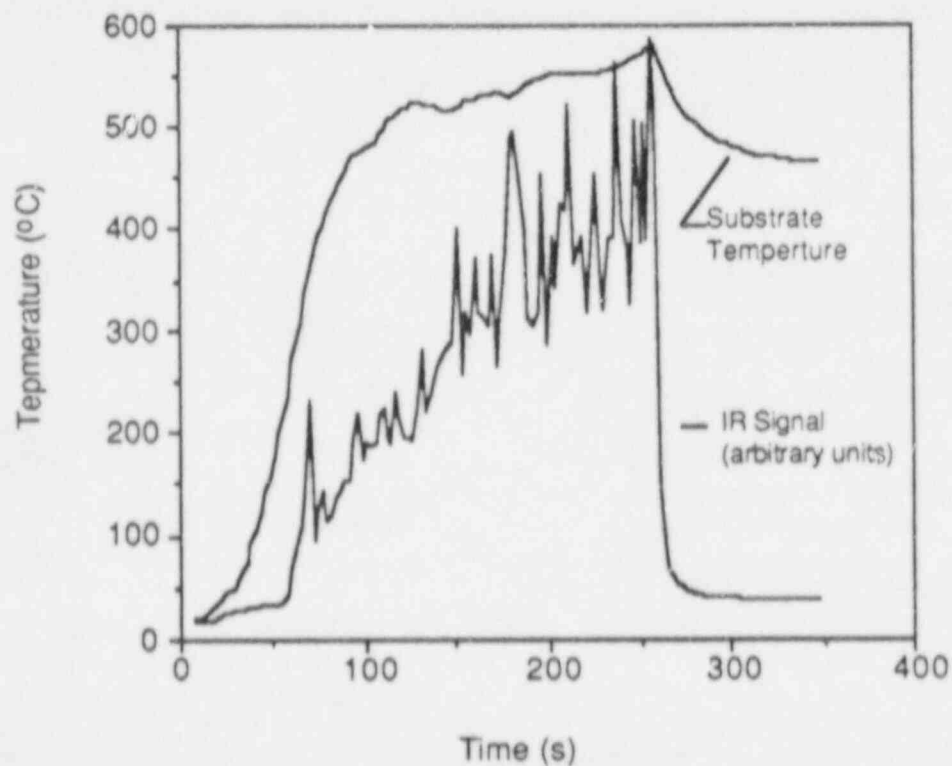


Figure 2. Results of a typical igniter test. Upper trace is the temperature of the platinum-coated substrate as measured by a chromel/alumel thermocouple. Lower trace is the signal from the infrared (IR) detector which detects the infrared radiation emitted from the catalytic substrate. The response time of the IR detector is more rapid than that of the thermocouple.

3. IGNITER DESIGN OPTIMIZATION

A schematic diagram of the first successful catalytic igniter is shown in Figure 3. It consists of a platinum coated honeycomb and platinum coil which are instrumented with a platinum/platinum - 13% rhodium thermocouple. Six other designs were tested and are shown in Figure 4. The figure caption indicates the conditions for which gas-phase ignition was achieved. The design which gave the best performance, that is, ignited the leanest mixtures, is shown in Figure 5.

We observed that for all of the successful igniter designs, the honeycomb first warmed, then the wires warmed and finally the wires glowed red hot and caused ignition. Small areas on the upstream surface of the substrate also glowed, but they were much less bright and more red than the wires. This observation provided qualitative evidence that the wires reached higher temperatures than the honeycomb and that ignition occurred near the wires and not near the honeycomb.

From our preliminary tests, we determined that both a substrate coated with high surface area platinum and platinum wires are necessary for the device to ignite very lean mixtures. That both are needed to cause ignition indicates that there exists some interaction between them. This could be mechanical (e.g., disruption of the flow around the igniter), chemical (e.g., one igniter part may supply a necessary chemical species to the other), or thermal (e.g., one igniter part may provide a temperature boost to the other).

The results of two separate experiments showed that the mode of interaction is primarily thermal. The apparatus used for the first experiment is shown in Figure 6. A coil of 0.0127 - cm diameter Pt wire was used as an electrical resistance heater to raise the temperature of the 0.0726 - cm diameter Pt wire whose catalytic ignition properties were to be determined. The Pt heater assembly and one end of the Pt catalyst wire below the heater coil were carefully sealed in Pyrex glass so the heater would not accidentally cause catalytic ignition of the hydrogen-air mixture. A chromel/alumel thermocouple was chosen to monitor the wire temperature and was attached to the Pt catalytic wire above the glass seal. Chromel/alumel rather than a platinum/platinum-rhodium thermocouple was used to avoid catalytic ignition from the thermocouple. To initiate the experiment, the platinum wire was heated electrically to a temperature high enough so that the chemical surface reaction would sustain itself without further electrical heating. At this point the electrical supply to the heater coil was disconnected. When the hydrogen concentration was high enough, the temperature of the wire continued to rise until gas-phase ignition occurred. It was found that only a small temperature boost above room temperature was needed to cause the surface reaction on the wire to accelerate and ultimately ignite the gas-phase mixture. When heated to 80° C, the wire positioned in a vertical direction ignited mixtures as lean as 8.0% hydrogen. This compares to 8.5% for a horizontal wire. Even less boosting was required for a vertical, 0.0127-cm diameter wire; heating to only 60° C was sufficient.

In the second experiment, the temperature boost was provided by the catalytic honeycomb as shown in Figure 7. One end of a 0.0726-cm diameter platinum wire was fastened to an alumina rod, and the other end was bent so that it could touch the substrate but not be fastened to it. If the distance between the wire and the platinum-coated honeycomb was more than 0.1 cm, then the platinum wire would not heat much above room temperature, and ignition would not occur even though the substrate heated to the

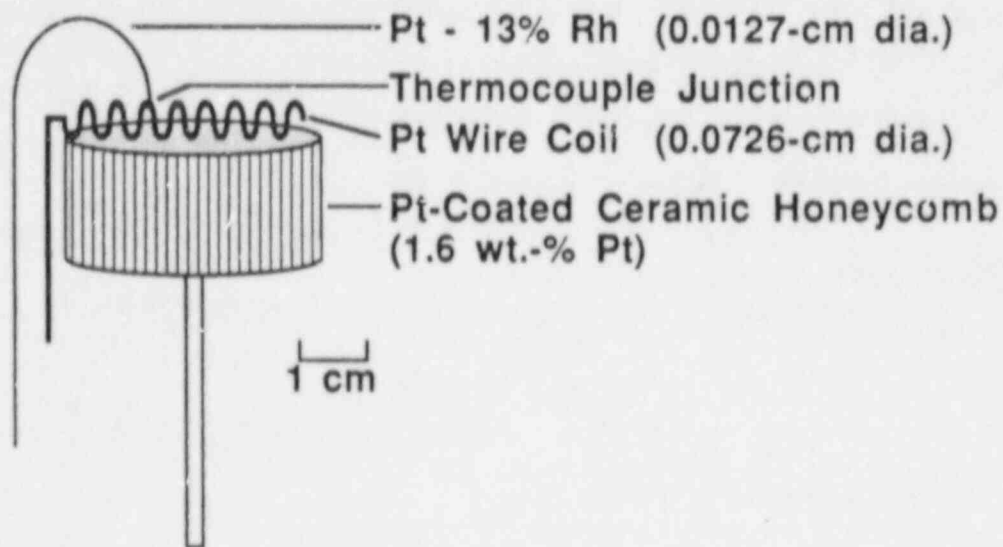


Figure 3. First successful catalytic igniter design. Composed of platinum-coated honeycomb, platinum wire coil and platinum - 13% rhodium wire. The temperature of the platinum wire coil was measured by the thermocouple junction formed by the platinum - 13% rhodium wire welded to the platinum wire coil.

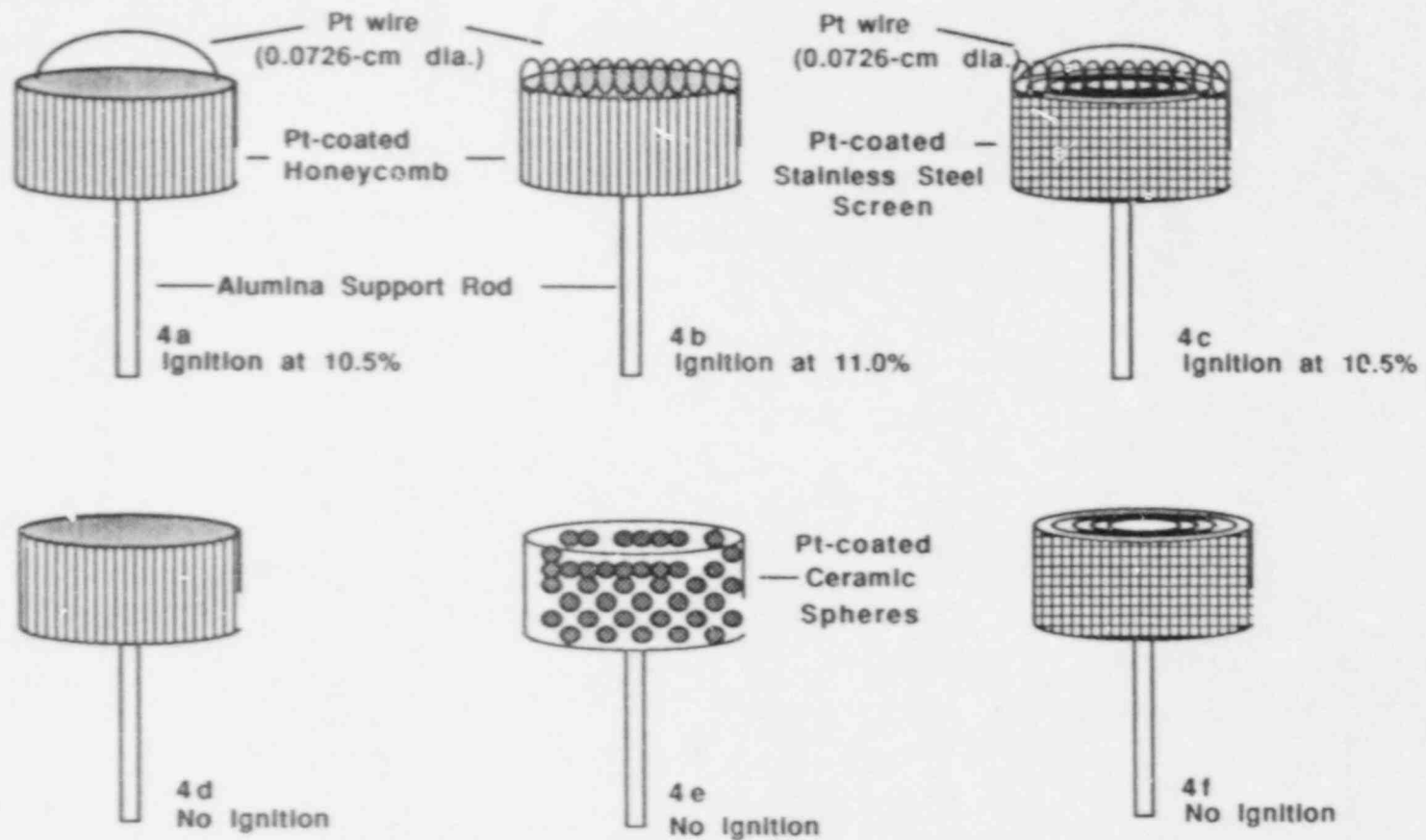


Figure 4. Catalytic igniter designs. Some designs worked, others did not. The hydrogen concentration at which the mixture was ignited is indicated near each figure.

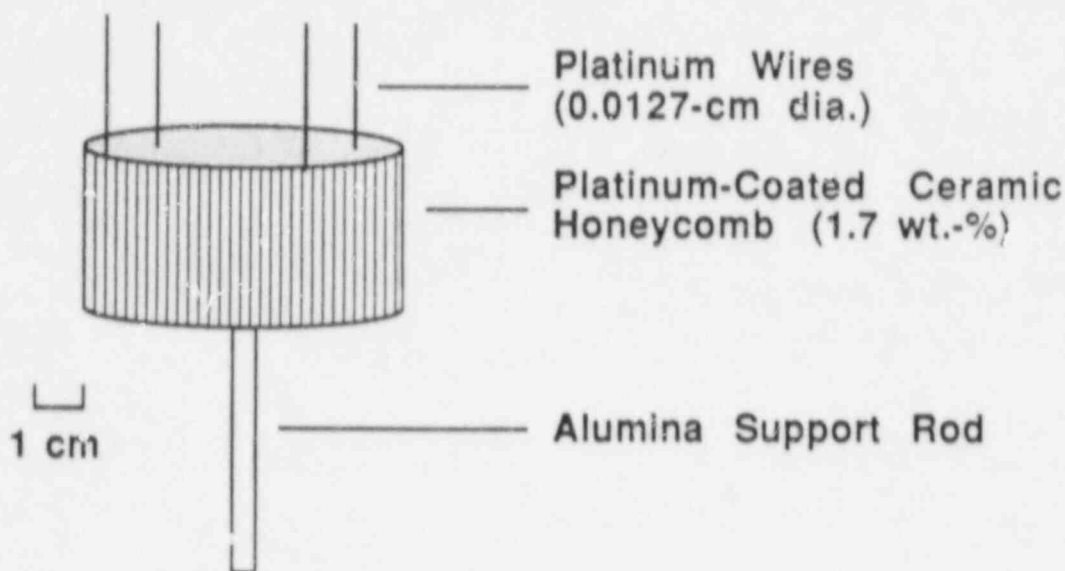


Figure 5. Schematic diagram of optimized catalytic igniter.

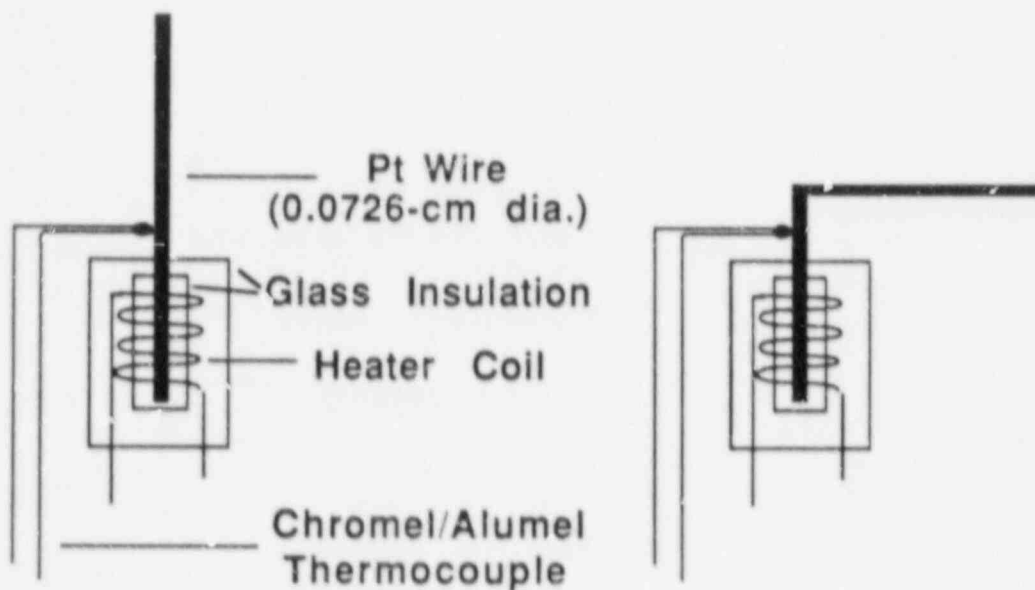


Figure 6. Device to electrically heat platinum wire to test the effects of thermal boosting. Ignition occurs at 8.0% hydrogen with a thermal boost to 82°C when the wire is oriented vertically. Ignition occurs at 8.5% hydrogen with a thermal boost to 80°C for the wire oriented horizontally. No ignition occurs without heating.

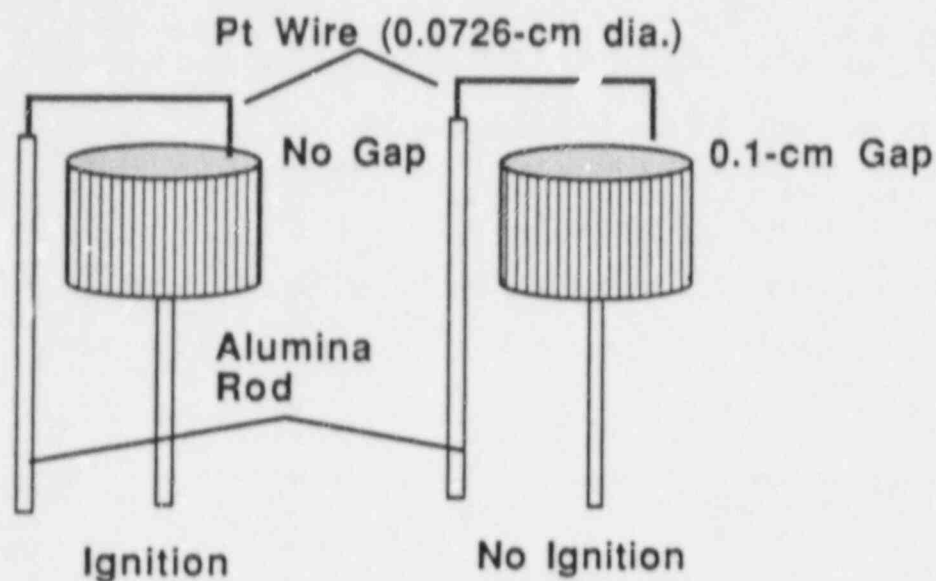


Figure 7. Device to test the effect of thermal boosting provided by the platinum-coated honeycomb. Ignition only occurs when there is thermal contact between the honeycomb and the platinum wire.

Table 1

Results of Optimizing Wire Diameter and Length.^a

Wire Diameter (cm)	Wire Length (cm)	Minimum Hydrogen Con. Needed for Ignition (%)
0.0127	1	6.1
0.0127	2	5.5
0.0127	4	5.5
0.0254	2	5.5
0.0492	2	7.5
0.0726	2	8.5

^aLong thin wires work best.

usual temperature. If the wire touched the substrate then ignition occurred as usual, indicating that the wire must be thermally boosted by the substrate.

To test the importance of wire position, the wires were hung below the substrate with the burner in the inverted configuration. In this position, the wires were downstream from the substrate and the igniter failed to ignite an 11% hydrogen mixture. Presumably, leaner mixtures could not have been ignited either.

From the foregoing experiments it is clear that (1) both the platinum-coated substrate and the platinum wire are necessary, (2) the primary effect of the substrate is to provide a temperature boost to the platinum wire, (3) vertical positioning of the platinum wire is more effective than horizontal, (4) straight wires are better than coils and (5) the wires must project away from the substrate into the unreacted mixture (upstream or to the side).

At this point experiments were performed to optimize the wire diameter and length. Wire lengths of 1, 2, and 4 cm, and wire diameters of 0.0127, 0.0254 and 0.0726 cm were tested. The results, which are summarized in Table 1, showed that long, thin wires worked best. This concluded our initial efforts to optimize the igniter design.

4. PERFORMANCE OF PROTOTYPE IGNITER

The optimized prototype igniter shown in Figure 5 was evaluated in terms of its response to hydrogen concentration, gas flow velocity, gas temperature, gas humidity and water spray. The ignition delay time was taken as a measure of the igniter's performance. The results are summarized below.

4.1 Hydrogen Concentration

For the inverted burner configuration (upward flame propagation) and a gas flow velocity of 2.8 cm/s, the leanest mixture which was tested, 5.5%, was successfully ignited. The leanest ignitable concentration for downward flame propagation was 11.6%. Hydrogen concentrations lower than 11.6% were ignited (as lean as 9.0%), but the flame stabilized on the catalytic substrate or moved upward to the edge of the chimney and extinguished. Only at concentrations of 11.6% or higher would the flame front propagate downward and stabilize on the burner surface. Because of this, we suspect that the 11.6% limit is strongly dependent on the gas flow velocity. However, we did not perform further tests to determine the interaction of gas velocity and concentration for downward flame propagation.

Results of the hydrogen concentration tests indicate that the catalytic igniter can successfully ignite very lean hydrogen-air mixtures. These are within the concentration range of interest in nuclear reactor safety (4 - 10%). In fact, the leanest mixtures ignitable with the catalytic igniter are only slightly more rich than the leanest mixtures ignitable with a spark in the case of upward flame propagation.

4.2 Flow Velocity

The effects of flow velocity for flows of 1.7, 2.8, 5.7, 8.6, 14.5 and 19.5 cm/s were examined for concentrations in the range of 5.5 to 11.5% with the gas mixture at room temperature (22° C). The ignition induction time (i.e., the time between the first exposure of the igniter to the hydrogen mixture and the time of the gas-phase ignition) was taken as an indication of the effectiveness of the igniter. Shorter times indicated

better performance. The results shown in Figure 8 indicate that the effects of flow rate are greatest at low hydrogen concentrations making the leaner hydrogen mixtures harder to ignite at slow flow velocities. This implies that static gas mixtures might be the most difficult to ignite so ignition tests with zero gas velocity were performed with collaborators at Sandia National Laboratories, Albuquerque, and are reported in the Appendix A. These tests showed that the catalytic igniter successfully ignited dry static hydrogen-air mixtures as lean as 6.5% hydrogen and humidified mixtures as lean as 10%.

4.3 Humidity

The effects of humidity were tested by adding water vapor to the hydrogen-air mixture. This was accomplished by bubbling the mixture through a heated water bath. The bath consisted of a 5-cm diameter, 20-cm long, copper cylinder filled with copper turnings and water. To prevent condensation of the water, the entire apparatus was heated to a temperature 2 - 20° C higher than the water bath depending on the desired relative humidity. This provided a humidified mixture at the temperature of the apparatus but at a dew point (saturated vapor temperature) equal to the temperature of the water bath. The relative humidity was computed from psychrometric tables taking the temperature of the apparatus as the dry bulb temperature and the temperature of the water bath as the wet bulb temperature. The relative humidity calculated in this way is only approximate because the gas flow velocities in the apparatus are much lower than those typically used to make psychrometric humidity measurements. This means that the relative humidity computed for the apparatus may be slightly higher than the actual humidity.

The results showing the effect of humidity are given in Figure 9 and Table 2. They indicate that humidified mixtures are more difficult to ignite and that the increase in the ignition delay is about a factor of three from the low humidity case to the 100% relative humidity case. Thus, the effect of high humidity is to delay the ignition but not to prevent it.

4.4 Temperature

The effect of gas temperature was investigated with the same apparatus used for the humidity studies except that there was no water in the bubbler and the entire apparatus was thermostated to a uniform temperature to within $\pm 2^\circ$ C. The results are shown in Figure 10. For the temperature interval tested, the data can be approximated by a straight line.

4.5 Water Spray

Fine water droplets were misted onto the igniter until its mass increased by about 10% (5 g of water). The igniter failed to warm even when exposed to a 11% hydrogen-air mixture. After the liquid water had evaporated, the igniter operated normally as before.

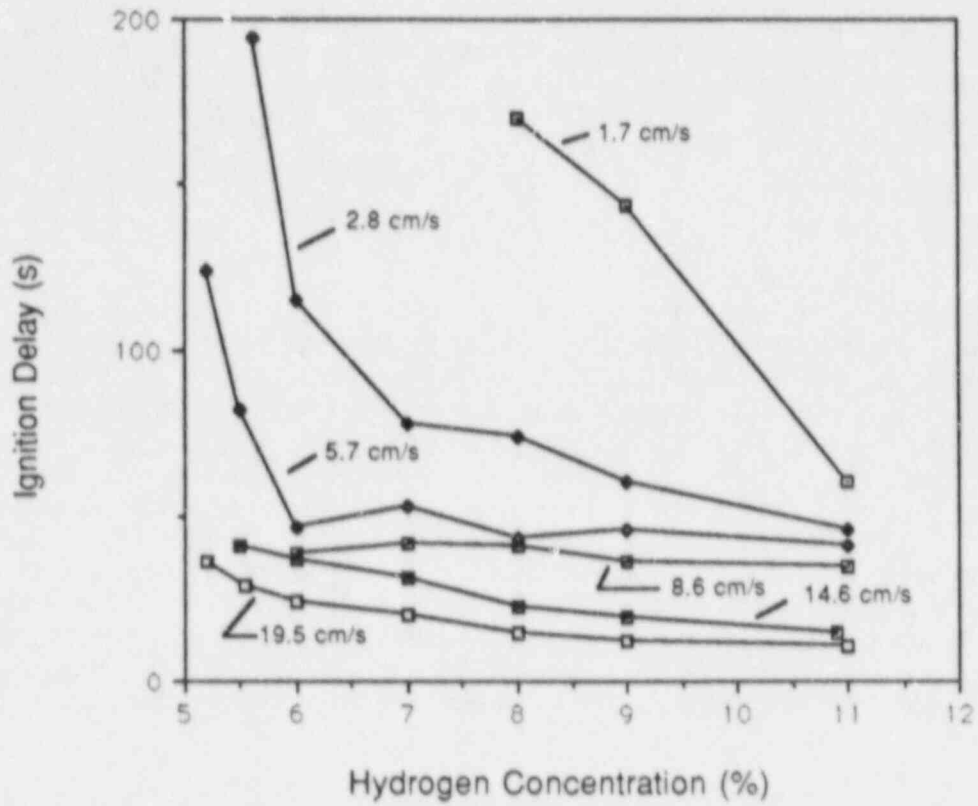


Figure 8. Effect of gas flow rate on igniter performance. Mixtures with low hydrogen concentrations and low velocities are most difficult to ignite. Gas temperature was 23°C and the relative humidity was less than 5%.

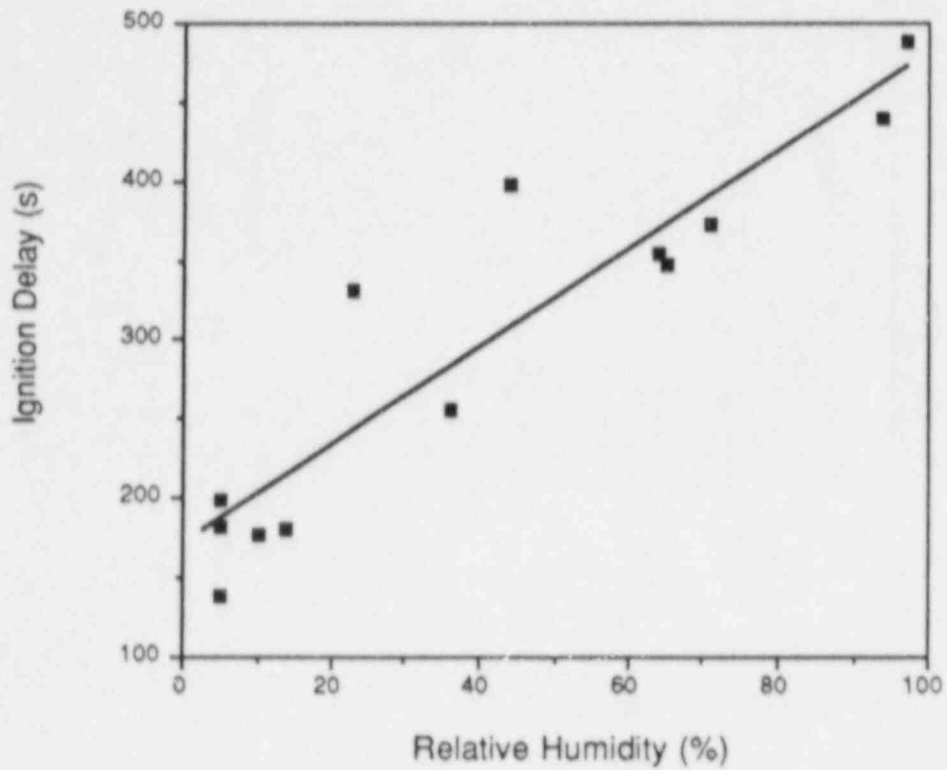


Figure 9. Effect of humidity on igniter performance. Humidified mixtures are more difficult to ignite than dry mixtures. The gas flow velocity was 2.8 cm/sec and the hydrogen concentration was 8%. The temperature of the gas was varied to achieve the desired relative humidity (see text).

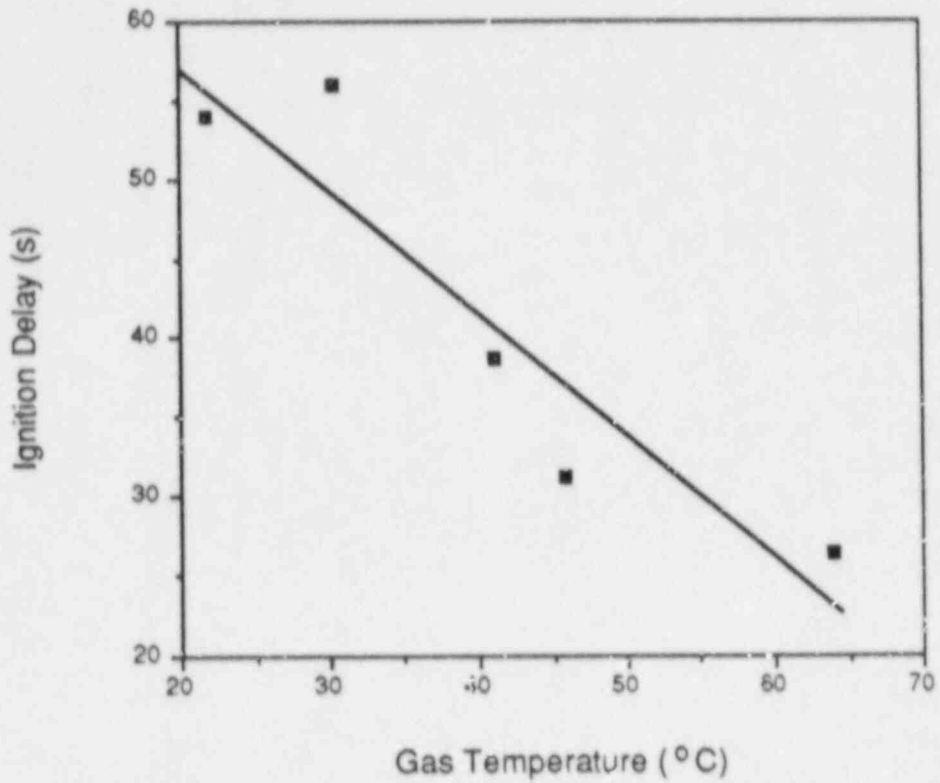


Figure 10. Effect of gas temperature on igniter performance. Warmer gas mixtures are easier to ignite. The flow velocity was 2.8 cm/sec, the hydrogen concentration was 8% and the relative humidity was less than 5%.

Table 2.

Effects of Flow Rate

Flow Velocity (cm/s)	Hydrogen Concentration (%)	Ignition Delay (s)
1.7	8.0	170
	9.0	144
	11.0	60
2.8	5.6	194
	6.0	115
	7.0	78
	8.0	74
	9.0	60
	11.0	46
5.6	5.5	199
	6.0	38
	7.0	41
	8.0	36
	11.0	35
8.6	5.2	124
	5.5	82
	6.0	47
	7.0	53
	8.0	43
	9.0	46
	11.0	41
14.6	5.5	41
	6.0	37
	7.0	31
	8.0	23
	9.0	19
	10.9	14
19.5	5.2	36
	5.6	29
	6.0	24
	7.0	20
	8.0	14
	9.0	12
	11.0	11

5. PRINCIPLE OF OPERATION

In order to fully optimize the igniter and to predict its behavior under a variety of conditions that may exist during a LOCA, it is important to understand its principle of operation.

Heating of both the platinum-coated substrate and the platinum wire depends on the catalytic reaction of hydrogen with oxygen on the platinum surface. The catalytic reaction occurs in the same way on the surface of the platinum-coated honeycomb and the surface of the wire. On a per-unit-area basis, the reaction rate for the wire and substrate is about the same.^{7,8} The primary difference is that the effective surface area of the platinum on the substrate is enormous relative to the area of the wire. Because of its high effective surface area, the substrate warms spontaneously in much leaner mixtures than does the wire.

The ignition sequence may be outlined as follows. Many of the processes proceed concurrently so the ordering is somewhat arbitrary and the exact details of the surface reaction mechanism are not fully known.⁷

1. Diffusion of hydrogen and oxygen to the platinum surface.
2. Adsorption of hydrogen and oxygen on the surface. (Since the igniter is stored in air, the hydrogen is actually adsorbed on a surface preadsorbed with oxygen).
3. Dissociation of hydrogen on the surface.⁷
4. Reaction of H with O or O₂ on surface to produce OH and heat.^{7,8}
5. Reaction of H or OH with OH on surface to produce H₂O and heat.^{7,8}
6. Desorption of H₂O from surface⁷ and the associated loss of heat from the surface.
7. Diffusion of hot H₂O from surface.
8. Acceleration of surface reaction rate due to surface heating.^{7,8}
9. Heat transfer from the catalytic substrate to the platinum wire and initiation of processes 4 - 7 on the wire surface.
10. Catalytic ignition of surface reaction on substrate (defined as the condition when the surface reaction rate is limited only by the diffusion rate of reactants to the surface). For lean static mixtures, catalytic ignition may occur at relatively low temperatures (100 - 300° C).⁹
11. Heating of the gases surrounding the igniter by conduction, convection, and radiation.
12. Initiation of catalytic ignition on wire surface.

13. Heat transfer from wires to substrate.
14. Ignition of gas-phase mixture near wire.
15. Propagation of the flame front away from the igniter.
16. Cooling of the igniter surface due to reduced supply of reactants to the surface.

Several steps in the process are worthy of further comment.

Step 1 for the wires, diffusion of reactants to the surface, is affected by the wire diameter. This can be understood in terms of a boundary layer surrounding the catalytic surface, defined as the region near the surface where the reactants are depleted relative to their concentrations in the bulk. Since the diffusion rate is proportional to the concentration gradient, and the gradient is larger near the surface of an object with a smaller radius than one with a larger radius, the diffusion rate near the surface of a small diameter wire is more rapid than for a large diameter wire. We believe that this is why the smaller diameter wires are capable of igniting leaner mixtures than larger diameter ones. It is important to realize that this argument implies that the thickness of the boundary layer is comparable to or thicker than the diameter of the wire (0.02 cm for these experiments). At low velocities or static conditions, mass transport is primarily by diffusion. At moderate velocities, convective transport occurs, but associated with it is convective heat loss. This loss reduces the wire temperature and thus the reaction rate. That we observe shorter ignition delay times with increased gas velocity indicates that the increased transport associated with higher velocities more than compensates for the effects of convective heat loss for the flow velocity used in our tests (below 20 cm/s).

That mass diffusion is important to the operation of the wires is indicated by model calculations performed by Schefer¹⁰ that ignored mass diffusion. The effect of mass diffusion is eliminated if one assumes that the concentrations at the surface are the same as they are in the bulk gas phase (i.e., no boundary layer exists). Then, according to the model calculations, large diameter wires should heat to a higher temperature than small diameter wires. This is contrary to our findings; and we, therefore, conclude that diffusion effects are important under our test conditions. It should be noted that Schefer's results may predict the correct dependence on wire diameter for very high gas velocities, in which case, diffusion is less important than convective transport. In this regard, our studies show that higher flow velocity gas streams are easier to ignite than lower velocity streams. This suggests that for the range of velocities studied, heating of the wire surface is strongly influenced by the transport of reactants and products to and from the surface.

In Step 7 water vapor desorbs from the platinum surface. Studies of the adsorption of water on platinum under ultra-high vacuum conditions show that the binding energy of water to platinum is quite low (12 kJ/mol).⁷ Further, there is no appreciable absorption at temperatures above 220 K.¹¹ This means that water vapor does not act as a poison toward the platinum and that once the water has formed on the surface, it should desorb easily if the igniter is at room temperature or above. This is consistent with our observations that the igniter can be cycled repeatedly. However, an igniter wet with liquid water will function normally only after it has been dried. This suggests that a film of liquid water blocks virtually all of the catalytic sites and/or inhibits the diffusion of reactants to the catalytic sites.

The effects of humidity on ignition induction time can be understood in terms of the ability of the gas-phase to take up water vapor produced from the surface reaction. If diffusion of water away from the platinum surface is the limiting step in determining the overall reaction rate, then the ignition induction time should be dependent on the ability of the gas-phase to take up water, i.e., relative humidity. We have observed a close-to-linear dependence (Figure 9). Water vapor should also affect the rate of diffusion of reactants to the surface, but this effect should be much smaller than the ability of the gas-phase to carry away the reaction products (water vapor). This is because the diffusion rate of oxygen and hydrogen through air compared with their diffusion rate through air saturated with water vapor should be nearly the same. Thus, the effects of water are to reduce the available reactants by dilution and to retard the loss of water vapor from the platinum surface. Both effects lengthen the ignition induction time.

6. EXPECTED PERFORMANCE OF CATALYTIC IGNITER IN A LOCA

6.1 Temperature

The initial temperature of the igniter strongly influences the ignition delay time. Lower temperatures give longer delays. We have not established the lowest temperature at which the igniter is operable. It has operated with initial temperatures as low as 18° C, although it is conceivable that temperatures as low as 0° C might be encountered in an ice-condensing type containment. The catalytic reaction of hydrogen with oxygen proceeds at 200 K (-73° C) and is not limited by water desorption at this temperature⁷. However, the rate of reaction may not be high enough to provide the necessary thermal boost for the igniter to operate normally. Thus, additional tests are needed if operation below room temperature is important.

At the other temperature extreme, our tests show that the igniter operates repeatedly up to the ignition temperature of hydrogen-air mixtures (585° C). Operation to the melting point of platinum (1772° C) is likely but of little consequence because ordinary metal surfaces will cause ignition around 585° C.

An important related question is whether or not the igniter will survive the ignition event it causes. In our laboratory tests, the catalytic substrate and wires were heated for several minutes to incandescence during studies with the richer (11% hydrogen) mixtures. We estimated the highest temperature achieved to be near 800° C. The igniter performance was not degraded by these high temperatures and the igniters operated repeatedly. In a LOCA environment, the flame front will propagate away from the igniter once ignition occurs, thereby reducing the exposure time of the igniter to extreme temperatures. This is because the hydrogen is depleted rapidly behind the flame front. The coatings used on the "wet proofed" catalytic substrates limit their maximum operating temperature to 250° C. Thus, a "wet proofed" igniter using currently available substrates might have a limited life. For example, if the hydrogen concentration is 5% or lower, and the substrate has a high platinum loading, then the substrate may heat above 250° C; but the mixture is too lean to be ignited by the platinum wires so the substrate "cooks" until it is destroyed. This problem can be overcome by optimizing the platinum loading of the substrate, but repeatable operation is still an unknown.

6.2 Flow Velocity

The flow velocity in the containment may be in the range of 0 - 10 m/s.^{2,3} Our tests show that the igniter operates successfully in the range 0 - 20 cm/s with higher velocity mixtures being easier to ignite. At some unknown velocity, further increases will not appreciably increase the supply of reactants to the surface but will increase the convective heat losses. At this point, higher gas velocities will produce longer ignition delay times, not shorter, and could defeat the igniter. We have not determined this critical velocity. However, if high velocities are anticipated, shields could provide areas of sufficiently low gas velocity to permit normal igniter operation.

6.3 Hydrogen Concentration

For any igniter to be an effective safety device, the time required for it to cause ignition must be short relative to the time that the hydrogen concentration builds to levels unsafe for intentional ignition. The longest ignition delay times for the catalytic igniter are roughly 400 s. This is short relative to the time expected for the hydrogen concentration to reach levels unsafe for ignition in the containment during a LOCA. In the case of Three Mile Island - Unit 2, the hydrogen concentration reached ignitable levels in a about 6 hours after turbine trip.¹²

6.4 Water Spray, Steam, and Fog

Our tests showed that liquid water on the surface of a room temperature igniter will prevent it from heating. This is a serious problem if water spray is used in the containment or if large amounts of steam are generated during the LOCA. One solution is to use a "wet proof" catalytic substrate. These substrates have been developed by Atomic Energy of Canada Limited (AECL) at Chalk River.¹³ AECL has fabricated substrates whose catalytic activities meet or exceed that of the substrate incorporated in our prototype igniter. One potential problem, however, is that they may be damaged at temperatures above 250° C.

6.5 Contaminated Atmosphere

If the igniter catalyst is poisoned, then the igniter will not function. During a LOCA, the atmosphere in the reactor containment may contain significant concentrations of gas-phase fission products such as iodine or cesium. Other gases, such as, carbon monoxide may also be present. The ability of these species to poison the platinum catalyst and defeat the igniter is generally not known. Tests will have to be performed to obtain a definitive answer. However, it is known that carbon monoxide is not a poison and is oxidized catalytically to carbon dioxide in the presence of oxygen on a platinum surface. Methyl iodide may be present during a LOCA, and it is known that methyl iodide reduces the catalytic activity of platinum if the concentration exceeds 0.1 ppm and will reversibly deactivate the catalyst at concentrations of 20 ppm. Catalytic activity is regained by heating the catalyst to 150° C with hydrogen concentrations above 6% and without methyl iodide present¹⁴.

The wet proof coating on the AECL catalysts might be impermeable to methyl iodide and other molecules containing large nuclei on the basis of size exclusion or polarizability. This could allow the igniter to operate normally in the presence of high levels of atmospheric contamination. This possibility of course must be tested.

7. FURTHER IMPROVEMENTS IN IGNITER DESIGN

There are several design parameters associated with the catalytic igniter that we have not optimized but which should be considered in future designs. These are discussed below.

7.1 Wire

The number of wires and their distribution on the surface of the substrate should be optimized. We have studied only designs with four wires placed around the edge of the substrate. Also, plating the wires with platinum black, a high surface area platinum coating, should be considered. This type of coating should increase the reaction rate on the surface of the wires without appreciably affecting its heat transport characteristics.

7.2 Platinum-Coated Substrate

We used only platinum-coated, ceramic honeycomb substrates with 1.6-1.7 weight % platinum. Catalytic substrates with higher platinum loading are available and should be tested. Since the transport of reactants to the substrate is affected by diffusion, the cell size of the honeycomb should be optimized along with the diameter and thickness of the honeycomb itself. The upstream face of the honeycomb (face closest to the burner) in our experiments warmed first and reached higher temperatures. This suggests that the optimum geometry for the honeycomb might have a larger, more open, or graded cell size and that the substrate could be thinner than the one we used. In addition, the honeycomb we used was deactivated by liquid water. Researchers at Atomic Energy of Canada at Chalk River¹³ have developed a "wet proofed" platinum/Teflon-coated catalytic substrate. Our preliminary tests using this substrate indicate that it will provide the thermal boost needed to heat the wires but that it is not quite as effective as the honeycomb for very lean mixtures (below 10% hydrogen). However, in some applications, the added benefit of the wet proofing may outweigh the lower catalytic activity of the substrate.

8. FURTHER TESTS

We have presented test results for a prototype non-powered igniter for nuclear reactor safety applications. It possesses two essential features of a usable igniter; it requires no power and it ignites very lean hydrogen-air mixtures. However, before the usefulness of the device as a replacement or supplement for existing electrically heated igniters can be established, further tests are required. These tests should include the environmental factors indicated in the Introduction. Specifically, further tests should examine the effects of steam, high flow velocities (up to 10 m/s), fog, water spray, and catalytic poisons. The results of these tests will clearly dictate the nature of further improvements.

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Appendix A

Tests of the Catalytic Igniter in the Fully Instrumented Test System (FITS)

L. S. Nelson, Kenneth P. Guay and L. R. Thorne

The laboratory test of the nonpowered catalytic igniter showed that the ignition delay time increases markedly for gas velocities below 10 cm/s. Unfortunately, the laboratory test apparatus was not capable of making tests below 1 cm/s to determine whether the catalytic igniter would ignite static hydrogen-air mixtures. However, because the velocity of the hydrogen-air mixture in the containment might be zero, or very low, under some circumstances that might occur during a loss of coolant accident (LOCA), it is important to test the igniter under similar flow conditions. Furthermore, it is important to make these tests in a moderately large volume to more nearly duplicate the conditions inside a nuclear reactor containment.

The Fully Instrumented Test System (FITS) at Sandia Albuquerque provides an opportunity to make the desired tests. The FITS system consists of a large (5.6 m³) cylindrical pressure vessel that is instrumented with a series of thermocouples and pressure gages placed at various positions along the vertical centerline of the vessel. Additional thermocouples are located in other positions as well. A computer-controlled data acquisition system logs the elapsed time and the temperature and pressure data.

A typical test proceeds by first purging the vessel with air, then adding enough hydrogen to bring the final mixture to the desired hydrogen concentration. This mixture is then stirred with a pneumatic fan for 10 min to ensure uniform concentration throughout the vessel. The mixture is then allowed to stand for 10 min to let any wind currents abate. At this point the igniter can be tested. The catalytic igniter is advanced by a pneumatically-driven piston from an argon-filled side arm on the vessel, through a ball valve, to near the center of the vessel. The argon in the side arm prevents the igniter from contacting the hydrogen-air mixture and warming prior to the desired start of the test. After a certain length of time (ignition delay time), ignition occurs and the temperature and pressure rises caused by the burning hydrogen are recorded by the computer-controlled data acquisition system. In some of the catalytic igniter tests, if the igniter did not operate within 10 min, the fans were turned on once again to produce low velocity flow across the igniter which helped the igniter to function. Tests were also made using glowplug and spark igniters in order to make comparisons with the catalytic igniter results. The same test procedure is followed for these igniters except that ignition was controlled by the operator by electrically energizing the igniter, and the igniters were located inside the tank from the beginning of the experiment rather than being moved into the tank using the side arm.

Ignition of dry and humidified mixtures was tested. Humidified mixtures were produced using a droplet mister located near the top of the vessel that generated micron-size drops. The mister was operated during the time the pneumatic fan was on and provided thorough humidification of close to 100% relative humidity.

The catalytic igniter was tested under four different conditions, with a wet or dry hydrogen-air mixture and with either 10% hydrogen or 6.5% hydrogen. The results of these tests are given in Table A1. Although there is considerable variability in the ignition delay times, the catalytic igniter successfully ignited wet 10% mixtures and dry 6.5% mixtures. The single wet 6.5% mixture tested required the fan to be turned on to

provide flow across the igniter. The pressure and temperature rises inside the vessel after ignition by the catalytic igniter were similar to those caused by glowplug and spark ignition. See, for example, Figure A1. The fraction of total hydrogen burned was also similar as indicated in Table A1.

The implications of these tests for reactor safety are that the nonpowered catalytic igniter is capable of igniting lean, static hydrogen air mixtures with hydrogen concentrations as low as 6.5% and humidified static mixtures with hydrogen concentrations as low as 10%. Humidified mixtures with hydrogen concentrations below 10% may need to be flowing before the catalytic igniter is effective. Nevertheless, ignition by the catalytic igniter is just as effective at reducing the hydrogen concentration as ignition by a spark or glowplug, but the catalytic igniter has the advantage that no electrical power is required for it to operate.

Table A1.

Results for the FITS tests. Data for glowplug and spark ignition are included for comparison with the catalytic igniter results. Two catalytic igniters of the same design were tested, CI A and CI B.

Test	Device	%Hydrogen Pre-Test	%Hydrogen Post-Test	Humidity	Fans On (s)	Ignition Delay (s)
1.	CI A	(10%) ^a	-----	DRY	300	305
2.	CI B	9.70%	0.02%	DRY	NO	76
3.	CI A	9.62%	0.02%	WET	NO	172
4.	CI B ^b	6.14%	2.35%	DRY	600	900
5.	CI B	6.16%	4.14%	DRY	NO	405
6.	CI B	6.16%	3.04%	DRY	NO	10
7.	CI B	(6%)		DRY	NO	10
8.	CI B	6.27%	2.11%	WET	600	665
9.	Glowplug	9.51%	0.02%	DRY	NO	15
10.	Glowplug	9.59%	0.02%	DRY	NO	15
11.	Spark	6.13%	4.65%	DRY	NO	1
12.	Spark	6.21%	4.51%	DRY	NO	1

^a Values in parentheses are estimated initial concentrations based on the volume of hydrogen supplied to the test tank.

^b Catalytic igniter rotated 90% from normal orientation.

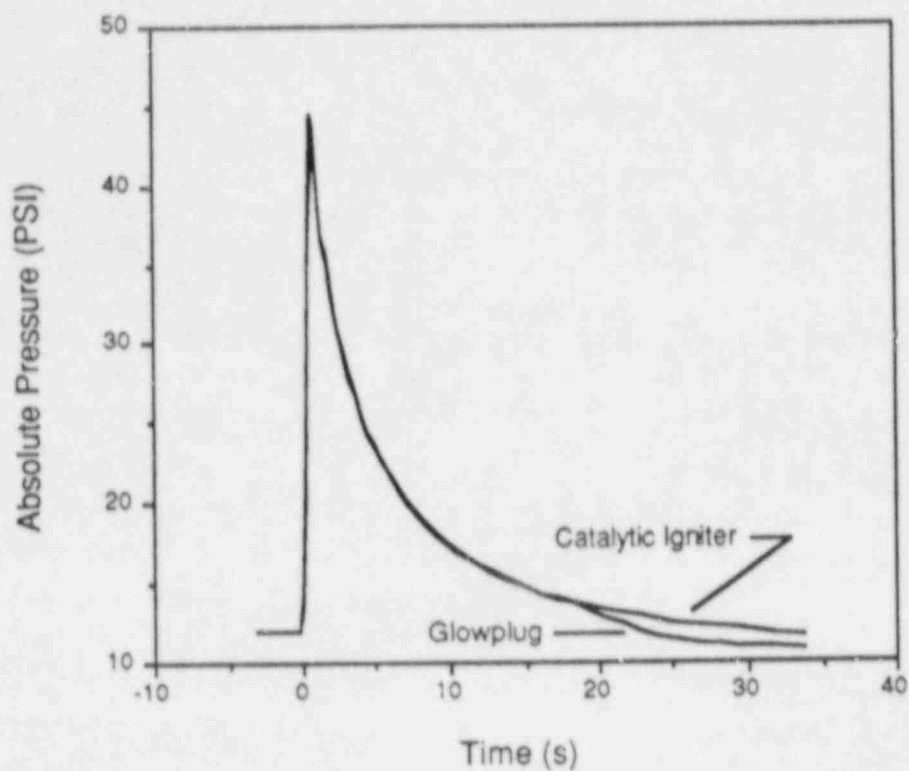


Figure A1. Pressure rise after ignition of 10% hydrogen mixtures in air. The pressure rise after ignition is virtually identical for ignition caused by either the glowplug or the catalytic igniter.

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We have developed a prototype catalytic igniter for lean hydrogen-air mixtures that could have important applications in nuclear reactor safety. The igniter has two useful characteristics related to these applications: it requires no electrical power and it can ignite mixtures as lean as 5.5% hydrogen. The ignition induction time ranges from 20 s to 400 s depending on the hydrogen concentration, gas flow velocity, gas temperature and relative humidity of the gas mixture. Induction times are shorter for mixtures with higher hydrogen concentrations, higher flow velocities, higher gas temperatures and lower relative humidity. The igniter operates successfully under conditions that may be present during a loss-of-coolant accident (LOCA) at a light water nuclear reactor. In the event of a LOCA, large quantities of hydrogen may be produced very rapidly and the catalytic igniter could provide a means of igniting it before dangerously high concentrations are attained; even in the event that electrical power required for conventional igniters is not available. The igniter has not been tested under all possible LOCA conditions. High gas velocities, water spray, steam and iodine-containing compounds may be present during a LOCA and will defeat the prototype igniter. However, shielding and semipermeable coatings on the igniter could overcome these difficulties.

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