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Results of Testing Large Hydrogen Generation Units to Inflate Lighter than Air (LTA) Vehicles

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ADMINISTRATIVE INFORMATION

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EXECUTIVE SUMMARY

OBJECTIVE

This report describes testing of hydrogen generation units capable of producing 330 cubic feet (ft^3) of hydrogen gas. For the tests conducted, cobalt chloride ($CoCl_2$) catalyzed sodium borohydride ($NaBH_4$):water (H_2O) chemistry was used to generate the hydrogen gas. Three configurations of the hydrogen gas generation units were tested. These tests were conducted under pressure at ambient temperatures of 0, 15, and 25 degrees Celsius (°C). A temperature control algorithm was developed for this system to automatically control the hydrogen gas generation reactions.

RESULTS

The results of these experiments showed that 330 ft^3 of dry hydrogen gas could be generated in a controlled manner.

The hydrogen generation reactor was modular and consisted of either two or three chambers. In these tests, the upper chamber housed the water while the lower chamber contained the $NaBH_4$ and $CoCl_2$. Either a thermocouple or a thermistor in the reagents was used to monitor and control the reaction. When producing 330 standard cubic feet (SCF) of hydrogen gas (H_2), the reagent bath from the lower chamber did rise into the chamber above, despite the fact that the reaction occurred under pressure.

Test Requirements

If no cooling coils were present in that chamber, thermal runaway occurred as the hottest zone of the reaction was at the air/reagent interface. Consequently, reaction control required configurations that assured the cooling coils would be in contact with the air/reagent interface. A temperature profile was imposed as part of the reaction control algorithm. This temperature profile ramped the control temperature from 42 to 57 °C at 1 °C increments every 2 minutes. A derivative control component was used in addition to the linear step-wise ramp. With the use of a control thermistor, which can sense small changes in temperature, derivative control smoothed the flow rate. This temperature profile worked for ambient operating temperatures of 0, 15, and 25 °C. For reactions that were under control, the temperature of the gas exiting the hydrogen generation unit was below 40 °C.

Outcome

Test results showed that the relative humidity of the exiting gas was significantly below that of ambient indicating that the gas was dry.

RECOMMENDATIONS

For future testing, use of hydrogen gas to inflate an aerostat needs to be demonstrated.

ABBREVIATIONS AND ACRONYMS

Co_2B	Cobalt Boride
$CoCl_2$	Cobalt Chloride
Cu	Copper
DC	Direct Current
ft	Feet
ft^3	Cubic Feet
gal	Gallon
H_2	Hydrogen Gas
H_2O	Water
ID	Inner Diameter
in	Inch
LTA	Lighter than Air
min	Minute
$NaBH_4$	Sodium Borohydride
$NaBO_2$	Sodium Metaborate
N_2	Nitrogen
PD	Proportional and Derivative
psi	Pounds per Square Inch
SCF	Standard Cubic Feet
T	Temperature
T_C	Temperature of Controlling Thermocouple
T_i	Temperature of Thermocouple i Where i = 1–6, Exit
T_{TH}	Temperature of Controlling Thermistor
°C	Degrees Celsius
%	Percent

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1. INTRODUCTION

1.1 OVERVIEW

As manufacturing processes using helium have increased worldwide, so has the demand for helium. Since helium is a non-renewable resource, this increased use in manufacturing has led to dwindling supplies of helium worldwide and a corresponding increase in cost. At the same time the use of lighter than air (LTA) vehicles, such as aerostats for military and commercial applications, has increased. Traditionally these vehicles have been inflated using helium. However, with the rising cost and scarcity of helium, the use of hydrogen gas to inflate LTA vehicles has been gaining greater acceptance. This report focuses on the generation of hydrogen gas as an alternative to helium as a lighter than air gas source.

1.2 HYDROGEN BENEFITS

Besides cost, there are other advantages of hydrogen over helium. One is that hydrogen provides more lift than helium. Consequently, larger payloads can be deployed using hydrogen to inflate LTA vehicles.

1.2.1 Advantages of hydrogen metal hydride

Unlike helium, hydrogen can be stored in a chemically dense form as a metal hydride. The addition of water, and in some cases, a catalyst, to the metal hydride starts the hydrogen generation process. Storing hydrogen as a metal hydride results in a long shelf-life. This is not true of compressed hydrogen or helium in tanks. These tanks leak considerably with time.

1.3 TEST SUMMARY

Prior to tests run for this report, experiments were conducted that used the Cobalt Chloride ($CoCl_2$) catalyzed sodium borohydride ($NaBH_4$):water (H_2O) hydrolysis to generate hydrogen gas [1]. The purpose of these experiments was to determine the feasibility of generating hydrogen gas to inflate LTA vehicles in a controlled manner. In the reaction vessel, copper cooling coils were immersed in the reagents, and the water flowing through the coils was used to remove heat generated during the reaction. The reaction temperature needed to be kept between 50 and 60 degrees Celsius (°C) to minimize water vapor in the gas stream and prevent thermal damage to the LTA vehicle. The experiments were conducted at ambient and cold temperatures while under pressure. When using $CoCl_2$ as a catalyst, we found that either distilled or deionized water should be used in order to complete the reaction in a reasonable time. Much longer reaction times occurred when tap water or sea water were used. It is believed that the chloramine in tap water and dissolved organics in sea water form complexes with the catalyst, impeding its ability to catalyze the reaction between $NaBH_4$ and H_2O. It was found that the minimum ratio of H_2O:$NaBH_4$ that could be tolerated was 4.6:1. Lower H_2O:$NaBH_4$ ratios resulted in a viscous sludge due to the precipitation of sodium metaborate ($NaBO_2$) hydrate crystals, causing foaming and reaction temperatures approaching 90 °C. The reaction rate could be controlled by either varying the cooling water flow rate or by switching the cooling on or off upon reaching a set temperature. Thermal imaging showed that the reacting solution temperature was inhomogeneous and hottest near the air/reagent interface. Consequently, placement of the monitoring/controlling thermocouple influenced control of the reaction. Experiments conducted under pressure showed that pressure suppressed foaming.

1.3.1 Test goals

Our goal was to use chemically generated hydrogen gas to inflate an aerostat. To this end, reactors capable of generating up to 330 standard cubic feet (SCF) of hydrogen gas were fabricated for our tests. The lessons learned as a result of the earlier laboratory experiments were applied in testing the larger units. The results of these tests are summarized in this report in Sections 3–5.

2. EXPERIMENTAL DETAILS

2.1 CHEMICALS

Sodium borohydride (GFS Chemicals), anhydrous cobalt (II) chloride (Sigma-Aldrich), and distilled water (Arrowhead) were used as received.

2.2 LARGE HYDROGEN GAS GENERATOR CONFIGURATIONS

Figure 1 shows schematics of the hydrogen gas generator configurations that were tested. The following design features applied to all testing conducted:

- The large hydrogen gas generator has a modular design, which allowed us to test three configurations shown in Sections 2.2.1, 2.2.2 and 2.2.3.
- The components were fabricated from aluminum.
- After fabrication, the components were given a Teflon impregnated, hard anodic coating.
- O-rings and clamps between the chambers of each unit provided a leak-tight seal.
- The bottom of the lower chamber, for all three configurations tested, was rounded instead of flat. An earlier investigation conducted by Ferreira et al., [2] showed that reactor bottom shape greatly affected hydrogen generation. Ferreira found that a conical bottom shape greatly enhanced the reaction rate and practically eliminated the reaction induction time.
- In the three configurations tested there was a burst disc (Fike) rated at 275 pounds per square inch (psi) to prevent the gas generation unit from over-pressurizing during the reaction.

2.2.1 Two Chambers with One Set of Cooling Coils

Figure 1a shows a schematic of the simplest configuration used. In this configuration, there were two chambers. The smaller, upper chamber housed the water used to react with the sodium borohydride. The maximum volume of water contained in the upper chamber was 4.4 gallons. The larger, lower chamber housed the cooling coils as well as a perforated basket containing the $NaBH_4$. During assembly, the $CoCl_2$ was placed on the bottom of the lower module. $NaBH_4$ was poured over it. Use of a metal screen to separate the cobalt chloride from the $NaBH_4$ was explored. It was found that the metal screen impeded the reaction between the $CoCl_2$ and the $NaBH_4$ to form the CoB_2 catalyst. As a result, not all of the $CoCl_2$ was converted to the CoB_2 catalyst, which affected the reaction rate. For this reason, the metal screen to separate the $CoCl_2$ and $NaBH_4$ was not used in subsequent tests. A perforated basket and bottom plate were put into place in the lower chamber. This basket was filled with the remaining $NaBH_4$. A trigger assembly provided a water tight seal between the two chambers. When activated, the trigger assembly opened and allowed the water from the upper chamber to enter the lower chamber.

2.2.2 Three Chambers with Two Set of Cooling Coils

A schematic of the second configuration used is shown in Figure 1b. In this configuration, there were three chambers. The smaller, upper chamber housed the water (a maximum of 4.4 gallons) used to react with the $NaBH_4$. Cooling coils were present in the middle and lower chambers. The gap separating the two sets of coils was 4 inches. The lower chamber housed the perforated basket that held the solid reagents ($CoCl_2$ and $NaBH_4$) in place. There was an opening between the middle and lower chambers. This opening allowed reagents to enter the middle chamber during the reaction. As in the previous configuration, a water tight trigger assembly separated the upper and middle chambers. When activated, the trigger assembly opened and allowed water from the upper chamber to enter the lower chambers.

2.2.3 Two Chambers with Two Sets of Cooling Coils

Figure 1c shows a schematic of the third configuration that was tested. There were two chambers in this configuration. Cooling coils were present in both chambers. The gap between the two sets of coils was 2.5 inches. The upper chamber housed the water used to react with the $NaBH_4$. The lower chamber housed the solid reagents, which were not confined in a perforated basket. As before, a trigger assembly separated the two chambers. When activated, the trigger assembly opened allowing the water from the upper chamber to enter the lower chamber.

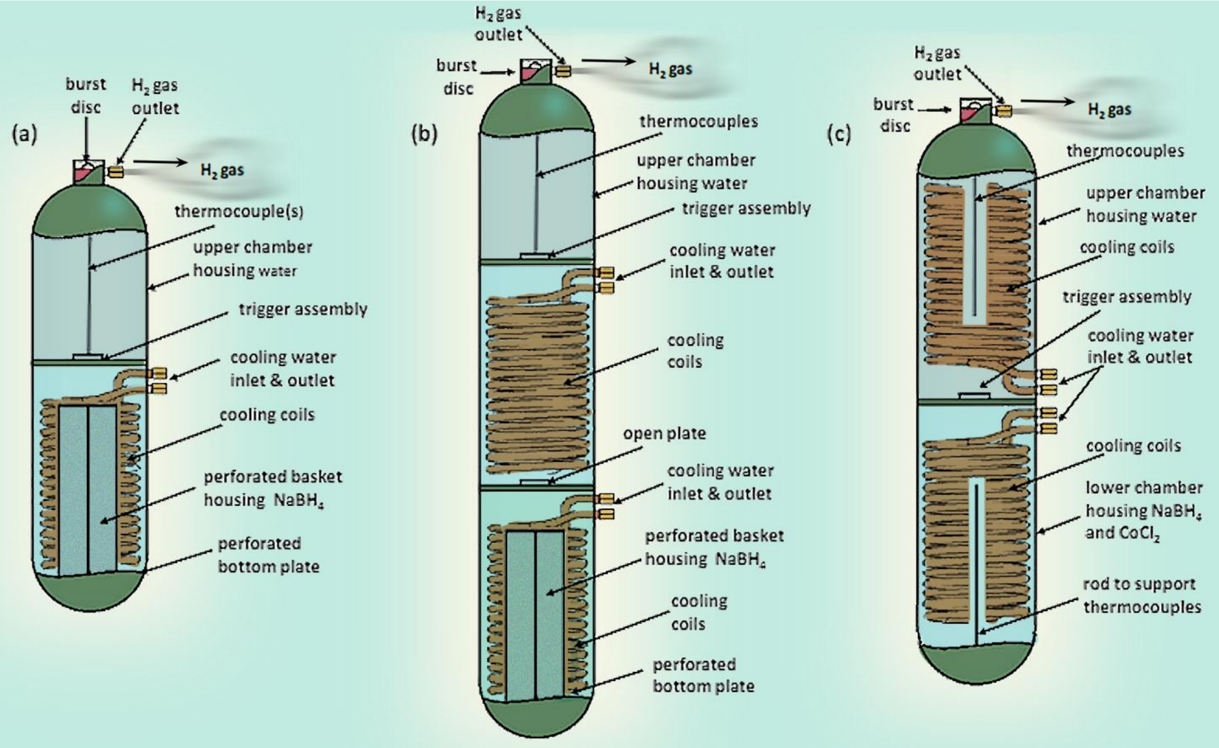

Figure 1. Schematics of (a) the two chamber hydrogen gas (H_2) gas generation reactor that used one set of cooling coils, (b) the three chamber H_2 gas generation reactor that used two sets of cooling coils, and (c) the two chamber H_2 gas generation reactor that used two sets of cooling coils.

2.3 HYDROGEN GAS GENERATION PROCEDURE

Unless otherwise indicated, the ratio of water to sodium borohydride used in these tests was 4.6:1. Each configuration of the hydrogen generation reactor had the capacity to generate 330 SCF of hydrogen gas, requiring 3.6 kg of sodium borohydride and 4.4 gallons of water. The amount of cobalt chloride used in these tests was in percent by weight relative to the sodium borohydride. Tests using both ambient temperature water and chilled water were conducted.

Figure 2 shows a schematic of the experimental set-up used to test the modules. Prior to reaction start, the H_2 reactor soaked in a 110-gallon (gal) drum of water used to establish the initial in situ reactor temperature. This bath also served as back-up cooling in the event the primary reactor cooling system failed. A submersible, centrifugal direct current (DC) well pump (Sun Pumps, model SCS 43-70-105 BL), immersed in a 300 gallon cooling water supply reservoir, provided cooling water to the generator cooling coils. The water flow through each coil was between 2.2–2.6 gal/minute (min). The outlets of the cooling coils discharged via hoses to a large discharge reservoir to collect the water exiting the cooling system. The H_2 gas outlet discharged gas into a 0.5 inch inner diameter, 400-feet (ft) hose rated for 4300 psi (8M2T Megaflex). Approximately 300 feet of this hose was immersed in a water-filled pool to provide cooling. Prior to testing, the hose was charged with nitrogen gas to flush-out oxygen and check for leaks. The reactor was operated at either ambient or colder temperatures. Ice was added to the 110-gal drum, the cooling water supply reservoir, and the H_2 gas discharge hose pool when the test plan dictated temperature conditions lower than ambient.

Depending on the generator configuration, four T-type thermocouples (Omega™) were installed at various locations inside the middle and lower chambers to monitor the reaction. Placement of the thermocouples, relative to the perforated bottom plate, are indicated in the figure captions and tables. In the upper chamber there were additional T-type thermocouples placed 17 inches and 11 inches from the top of the module. For the configurations shown in Figures 1a, and 1b, the 17-inch thermocouple was at the trigger assembly. A pressure transducer (Omega™ model: PX835-300GI) and T-type thermocouple were placed just downstream of the H_2 gas outlet to measure the pressure and temperature of the H_2 gas as it entered the 0.5 inch, 400-ft hose. At the exit of this hose, a mass gas flow meter (Alicat Scientific) was used to measure the gas flow rate, gas pressure, and temperature of the gas. A data logger (Omega™, model: OM-EL-USB-2-LCD) placed at the hose exit, recorded temperature and relative humidity. All data acquisition was under computer control. A 100-psi pressure relief valve (Circle Seal Controls 5-80-A-3MP-100), at the exit of the Alicat gas flow meter, kept the reagents under pressure to suppress foaming.

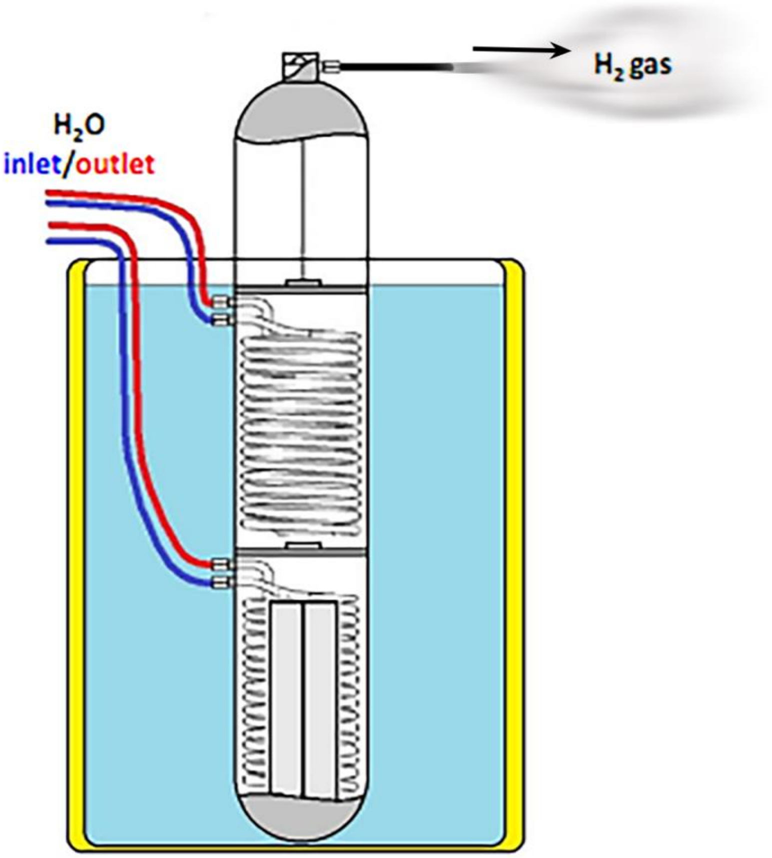

Figure 2. Schematic of the experimental configuration used to test the modules.

Once the H_2 gas reactor, the cooling water supply reservoir, and the gas exit hose pool reached the desired operating temperature, the reaction was initiated by an electronic signal that triggered the release of water from the upper chamber of the H_2 gas generation reactor into the lower chamber(s). This water reacted with the sodium borohydride and cobalt chloride to generate H_2 gas. A thermocouple in the lower chamber was used to control the reaction. This thermocouple is indicated in the figure captions and tables. When the control thermocouple reached a set temperature, the pump would force water through the cooling coils. This set temperature could be changed manually during the course of the reaction.

Once the reaction reached completion, hot water was used to flush spent reagents out of the reactor vessel. The water purge was sustained until clear water exited the unit. The unit was then filled with water and 2 gallons of vinegar. After 5 minutes, nitrogen gas was used to flush the vinegar-water mixture out of the reactor. Water was again flushed through the unit and then forced out using pressurized nitrogen gas. The reactor was then taken to a laboratory for disassembly, cleaning, and inspection.

3. RESULTS OBTAINED USING THE TWO CHAMBER, ONE SET OF COOLING COILS CONFIGURATION

Initial tests used enough sodium borohydride to generate 165 and 247.5 cubic feet (ft^3) of hydrogen gas. The ratio of $H_2O:NaBH_4$ for the 165 and 247.5-ft^3 H_2 generation tests were 9.3:1 and 6.2:1, respectively. The total amount of water used in each of these experiments was 4.4 gal. Results of these experiments are summarized in Table 1 (Run 1 and Run 2) and Figures 3 and 4. The results were similar to that observed in the laboratory experiments [1].

Table 1. Summary of hydrogen generation runs using the two chamber, one set of cooling coils configuration shown in Figure 1a.

Two Chamber Run Number	SCF of H_2 gas generated[a]	Ratio H_2O to $NaBH_4$	Percent (%) $CoCl_2$ catalyst[b]	T of water baths (°C)[c]	Height of Control Thermocouple (inches)[d]	Set T (°C)	Time to Initial Set T (minutes)	Reaction Time (minutes)
1	165 (Figure 3)	9.3:1	3.25	15	12	50	5.6	18
2	247.5 (Figure 4)	6.2:1	2.75	15	12	40, then step to 45, 50	2.6	32.2
3	330 (Figure 5)	4.6:1	1.25	15	12	40	10.1	disc burst
4	330	4.6:1	0.50	15	12	37, then step to 40	95	200
5	247.5	4.6:1	0.90	15	10.5	33, step to 35,37,40,45, 50,70, 60	8	168
6	247.5 (Figure 6)	4.6:1	1.25	15	10.5	33 step to 35, 37, 38, 39,40,41, 42,43, 45,59,70	6.7	120
7	247.5	4.6:1	1.25	15	10.5	33, step to 35, 37, 40, 45,47,50,52, 70	8	91

a. Figure corresponding to run is indicated.

b. Percent $CoCl_2$ is relative to the mass of $NaBH_4$.

c. The hydrogen generation reactor, cooling water and hose are in drums or containers maintained at a given temperature.

d. Height of the control thermocouple is relative to the perforated bottom plate, Figure 1a.

Table 1. Summary of hydrogen generation runs using the two chamber, one set of cooling coils configuration shown in Figure 1a. (Continued)

Two Chamber Run Number	SCF of H_2 gas generated[a]	Ratio H_2O to $NaBH_4$	% $CoCl_2$ catalyst[b]	T of water baths (°C)[c]	Height of Control Thermocouple (inches)[d]	Set T (°C)	Time to Initial Set T (minutes)	Reaction Time (minutes)
8	165	4.6:1	1.25	15	4	45, step to 50	14.5	68
9	165 (Figure 7)	4.6:1	2.00	15	4	45, step to 48, 50	6	31
10	214.5	4.6:1	2.125	15	4	50	8.5	32

a. Figure corresponding to run is indicated.

b. Percent $CoCl_2$ is relative to the mass of $NaBH_4$.

c. The hydrogen generation reactor, cooling water and hose are in drums or containers maintained at a given temperature.

d. Height of the control thermocouple is relative to the perforated bottom plate, Figure 1a.

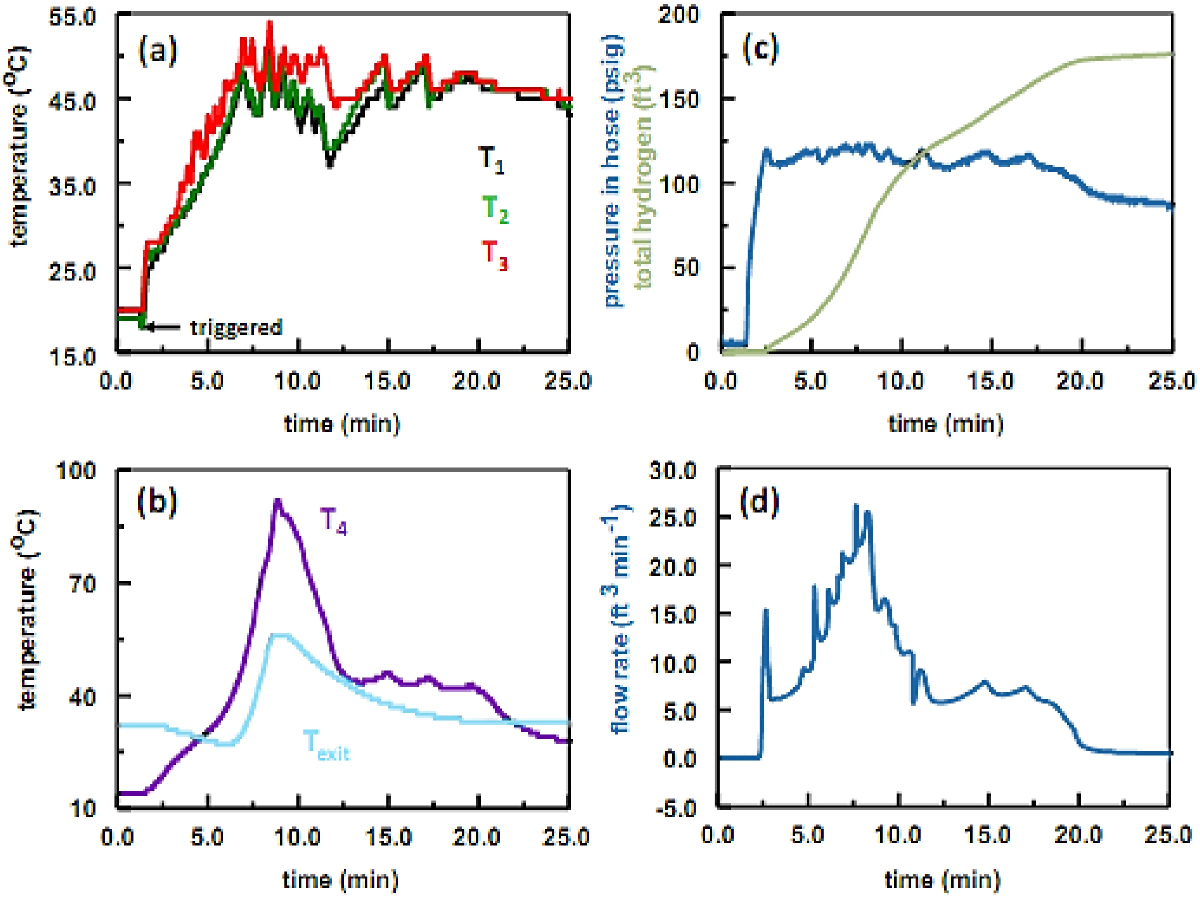

Figure 3. Results measured as a function of time for the 165 SCF H_2 gas generation, 9.3:1 H_2O:$NaBH_4$ ratio run using the two chamber, one set of cooling coils configuration, Figure 1a. The test was carried out at a temperature of 15 °C. The catalyst was 3.25% $CoCl_2$ relative to $NaBH_4$. (a) Temperatures measured in the lower chamber where T_1, T_2, and T_3 were placed 4, 8, and 12 inches relative to the perforated bottom plate, Figure 1a. Thermocouple T_3 was used to control the reaction. (b) Temperatures measured in the upper chamber at the trigger assembly, T_4, and the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure in the hose and total hydrogen gas. (d) Measured flow rate.

For the 165-ft^3 H_2 gas generation, a 9.3:1 H_2O:$NaBH_4$ ratio was run at 15 °C. The thermocouple used to control the reaction, T_3, was placed 12 inches above the perforated bottom plate, Figure 1a, close to the air/water interface, which earlier laboratory experiments showed was the hottest region of the reaction [1]. Thermocouples T_1, T_2, and T_3 were in the lower chamber and were in contact with the reagents throughout the course of the experiment. Thermocouple T_4 was in the upper chamber near the trigger assembly. After triggering, it took 5.6 minutes for the reaction to reach the set temperature of 50 °C, Figure 3a, at which time the flow of water through the cooling coils was activated. Thermocouple T_4 reached a maximum temperature of 91 °C, Figure 3b. Given that the unit was running at 50% of its capacity, T_4 recorded the temperature of the H_2 gas above the reagents.

The thermocouple used to measure the hydrogen gas exit temperature, T_{exit}, was approximately 25 inches downstream of T_4. As shown in Figure 3b, the temperature of the hydrogen gas cooled significantly from a maximum 91 °C to 56 °C. By the time the gas flowed through 400 feet of hose, volume as a function of time. The plot of total H_2 gas volume is nonlinear, indicating that gas production was not homogeneous. Figure 3d is a plot of flow rate as a function of time. The spike in flow rate at 2.7 minutes was primarily due to the nitrogen (N_2) gas exiting the hose. The Alicat is a mass flow meter and N_2 (in the hose to displace combustible oxygen) is significantly more massive than H_2 gas. Formation of the cobalt boride (Co_2B) catalyst also contributes to this spike, as was demonstrated in the tests run in the laboratory [1]. As shown in Figure 3c, the maximum flow rate coincides with the maximum temperature recorded by thermocouple T4. The total reaction time for the 165-ft^3 H_2 gas generation (9.3:1 H_2O:$NaBH_4$ ratio) run was 18 minutes. The sensor at the hose exit showed that the relative humidity of the gas exiting the hose was 20–46% (ambient humidity on the day of the test was 73%), indicating that the hydrogen gas was dry.

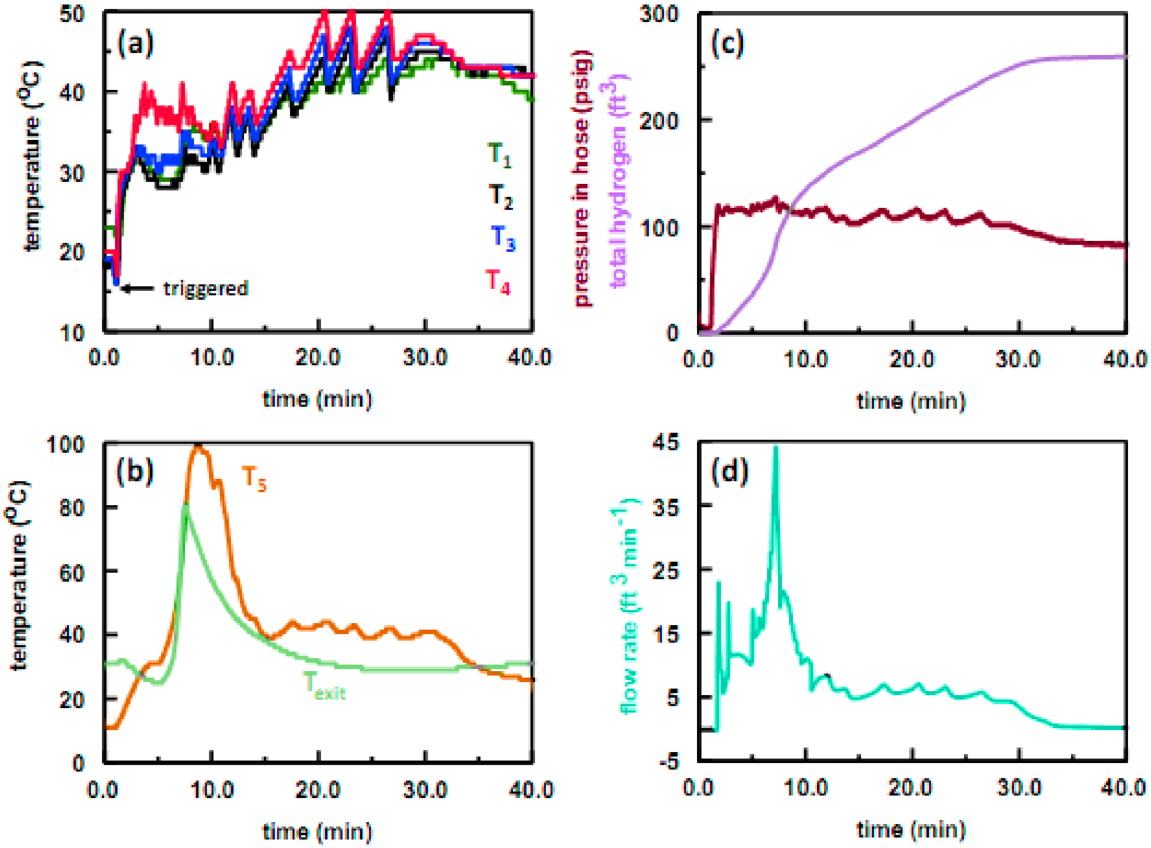

Figure 4. Results measured as a function of time for the 247.5 SCF H_2 gas generation, 6.2:1 H_2O:$NaBH_4$ ratio run using the two chamber, one set of cooling coils configuration, Figure 1a. The test was carried out at a temperature of 15 °C. The catalyst was 2.75% $CoCl_2$ relative to $NaBH_4$. (a) Temperatures measured in the lower chamber where T_1, T_2, T_3, and T_4 were placed 0, 4, 8, and 12 inches relative to the perforated bottom plate, Figure 1a. Thermocouple T_4 was used to control the reaction. (b) Temperatures measured in the upper chamber at the trigger assembly, T_5; and the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure in the hose and total hydrogen gas. (d) Measured flow rate.

In the case of the 247.5-ft^3 H_2 gas generation test, the ratio of H_2O:$NaBH_4$ was 6.2:1 and run at an ambient temperature of 15 °C. Thermocouple T_4, was placed 12 inches above the perforated bottom plate (Figure 1a) and used for control. The placement of the thermocouples is described in the caption of Figure 4. Because of the lower H_2O:$NaBH_4$ ratio and lessons learned from our previous laboratory experiments, the amount of catalyst was reduced to 2.75%. The initial set temperature to activate cooling was 40 °C. After triggering, it took 2.6 minutes for the reaction to reach the set temperature of 40 °C, Figure 4a. The temperature of the thermocouple at the trigger assembly, T_5, peaked at 100 °C, Figure 4b. The temperature of the H_2 gas exiting the hydrogen generation unit, T_{exit}, peaked at 80 °C, Figure 4b. However, the temperature of the gas exiting the 400-ft hose ranged between 30 and 40 °C. The pressure stayed steady at 100 psig. Total hydrogen gas production was nonlinear, Figure 4c, indicating that the production of hydrogen gas was not uniform. The temperature spikes observed for thermocouples T_5 and T_{exit} coincided with a spike in the H_2 gas flow rate, Figure 4d.

At 13.3 minutes into the reaction, when the temperatures recorded by thermocouples T_5 and T_{exit} had come down, the set temperature was manually increased to 45 °C. After one saw tooth, when no increase in temperature measured by thermocouple T_5 had occurred, the set temperature was increased to 50 °C. The total reaction time was 32.2 minutes. At the 37-minutes mark, T_1 is observed to cool faster than T_2–T_4. This is because thermocouple T_1 is closer to the bottom of the hydrogen generation unit, which is in direct contact with the exterior cold water in the 110-gal reactor immersion drum. The sensor at the hose exit showed that the relative humidity of the exiting gas was 20–40% (ambient humidity on the day of the test was 62%) indicating that the hydrogen gas was dry.

A 330 SCF H_2 gas generation run at 15 °C was then conducted using a 4.6:1 H_2O:NaBH4 ratio. The amount of catalyst used in this run was 1.25% and thermocouple T_4, placed 12 inches above the perforated bottom plate (Figure 1a), was used for control. Results of Run 3 are summarized in Table 1 and Figure 5. The temperature to activate cooling was set at 40 °C. As shown in Figure 5a, it took 10.1 minutes for the reaction to reach the set temperature, at which point cooling was activated. At the 11-minutes mark, the temperatures recorded by thermocouples T_5 and T_{exit} began to dramatically increase, Figure 5b. The temperature of the H_2 gas exiting the hydrogen generation unit greatly increased, as did the pressure of the gas entering the hose, Figure 5c. The build-up of pressure caused the reactor vessel burst disc to rupture at the 12.1-minutes mark spewing reagent and relieving the pressure. With no pressure to suppress foaming, reagent continued to discharge from the ruptured burst disk. At the 12.4-minutes mark, thermocouple T_{exit} peaked at 170 °C as hot reagent flowed into the gas exit hose. Figure 5d shows plots of flow rate and total hydrogen gas as a function of time. The spike in flow rate at 3.8 minutes was due primarily to the N_2 gas exiting the hose as described previously. Formation of the Co_2B catalyst also contributed to this spike, as was demonstrated in the laboratory [1]. The thermocouple that was used to measure the temperature of the hydrogen gas exiting the hydrogen generation unit, T_{exit}, was approximately 25 inches downstream from T_5. The increase in flow rate coincided with the temperature increases measured by both T_5 and T_{exit}. The flow rate pegged the meter at 44 SCF per minute, which is the maximum capacity of the Alicat device. When the burst disc ruptured, the reaction was approximately 21% complete.

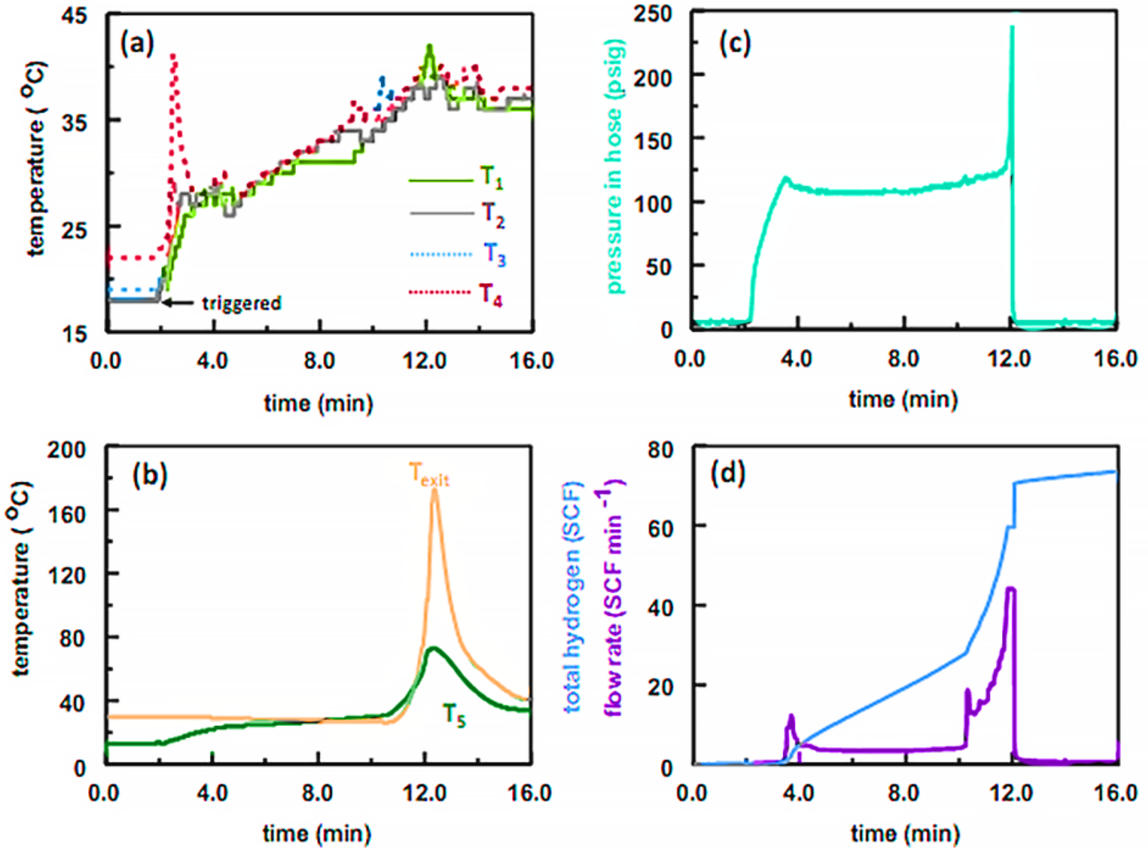

Figure 5. Results measured as a function of time for the 330 SCF H_2 gas generation, 4.6:1 H_2O:$NaBH_4$ ratio run using the configuration shown in Figure 1a. The test was carried out at a temperature of 15 °C. The catalyst was 1.25% CoCl2 relative to $NaBH_4$. (a) Temperatures measured in the lower chamber where T_1, T_2, T_3, and T_4 were placed 0, 4, 8, and 12 inches relative to the perforated bottom plate, Figure 1a. Thermocouple T_4 was used to control the reaction. (b) Temperatures measured in the upper chamber at the trigger assembly, T_5, and the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure into the hose. (d) Measured flow rate and total hydrogen gas.

The 330 SCF H_2 gas generation run at 15 °C using a 4.6:1 H_2O:$NaBH_4$ ratio was repeated in Run 4, Table 1. However, less catalyst was used in this Run (0.5% $CoCl_2$) and the temperature set point was lowered to 37 °C. As the data summarized in Table 1 indicates, it took significantly longer to reach the set temperature and longer for the reaction to come to completion. While the thermocouples in the lower chamber stayed at or below the set temperature, the thermocouple in the upper chamber at the trigger assembly, T_5, recorded two heat bursts. One heat burst occurred when the set temperature of 37 °C was reached on thermocouple T4. The maximum temperature reached during this burst was 74 °C on thermocouple T5. This heat burst lasted 30 minutes. The second heat burst occurred 11 minutes after the first heat burst ended. This heat burst lasted 60 minutes and the maximum temperature measured was 61 °C. The gas flow rate peaked at 15 SCF per minute during the first heat burst. The total hydrogen volume as a function of time was nonlinear.

The results of the two 330 SCF runs using a 4.6:1 H_2O:$NaBH_4$ ratio clearly show that the reagents were getting into the upper chamber during the reaction and that the air/reagent interface was the hottest region. It was also the region not in contact with the cooling coils. Follow on tests reduced the volume of hydrogen gas generated while maintaining the 4.6:1 H2O:NaBH4 ratio, so that the reagents remained in the lower chamber and in contact with the cooling coils. Table 1, Run 5, summarizes a test conducted at 15 °C ambient temperature to generate 247.5 SCF of hydrogen gas. The amount of catalyst used was 0.90%. Thermocouple T4, 10.5 inches above the bottom of the perforated basket, was used for control. The initial set point temperature to initiate cooling was 33 °C. It took 8 minutes to reach the set temperature. The temperature was kept at the 33 °C set point for 100 minutes. During this time, the temperature of the thermocouple in the upper chamber near the trigger assembly, T5, recorded temperatures between 20 and 30 °C. The hydrogen gas flow rate was a constant 1.38 SCF per minute. After 100 minutes, when the reaction was approximately 60% complete, we began increasing the temperature set point. As the set point increased, the gas flow rate increased. The temperature measured by T5 never exceeded the temperatures measured in the lower chamber. At 152 minutes into the reaction, the temperature at T4 dropped below temperatures at T1–T3 (0, 4, and 8 inches relative to the bottom of the basket), indicating that T4 was no longer in contact with the reagents. The reaction came to completion after 168 minutes.

The next test to generate 247.5 ft3 of hydrogen gas, given 15 °C ambient conditions, was conducted using 1.25% catalyst. Thermocouple T4, positioned 10.5 inches above the base of the basket, was used for control. Results are summarized in Table 1, Run 6, and Figure 6. The initial set temperature to activate cooling was 33 °C. It took 6.7 minutes for the reaction to reach this temperature, Figure 6a. The set point temperature was manually increased multiple times throughout the course of the reaction. The temperatures recorded by thermocouples T1–T5 stayed at or below the set temperature as indicated in Figures 6a and 6b. At approximately 90 minutes into the reaction, temperatures measured by thermocouples T1 and T4 dropped dramatically below those measured by thermocouples T2 and T3. These four thermocouples were all in the lower chamber. T1–T4 were positioned 0, 4, 8, and 10.5 inches, respectively, relative to the bottom of the basket. The T4 temperature decrease indicates that this thermocouple was no longer in contact with the reagents. Thermocouple T1 had to be in contact with the reagents, however it was closer to the bottom of the hydrogen generation unit and thus closer to the cold water present in the 110-gal water bath in which the unit was immersed. This explains why the temperature recorded by T1 was less than that of T2 and T3. Figure 6b shows that the temperature of the H_2 gas exiting the hydrogen generation unit, T_{exit}, was between 25 °C and 32 °C. The reaction came to completion after 120 minutes. Figure 6c shows the gas hose pressure and total hydrogen volume as a function of time. The plot of total hydrogen is nearly linear. The flow rate as a function of time is shown in Figure 6d. Flow rate was observed to increase whenever the set temperature was increased. The relative humidity during the reaction varied between 30% and 50%. Ambient humidity was 57%. This run was repeated with results summarized in Table 1, Run 7. To speed up the reaction, the time intervals between set temperatures were shortened. Run 7 took less time to reach completion, otherwise, the temperature, flow rate, hose pressure, and total hydrogen profiles were similar to those of Run 6.

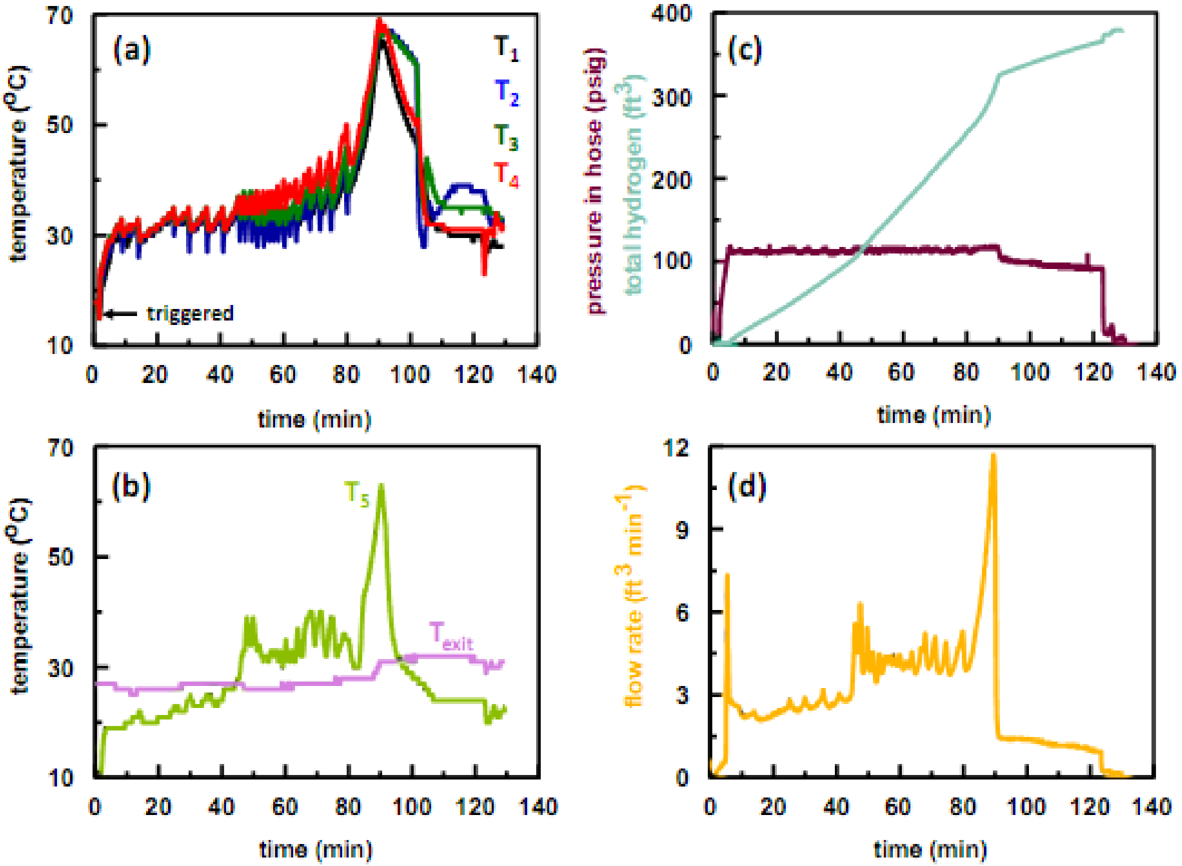

Figure 6. Results measured as a function of time for the 247.5 SCF H_2 gas generation, 4.6:1 H_2O:$NaBH_4$ ratio run using the two chamber, one set of cooling coils configuration, Figure 1a. The test was carried out at a temperature of 15 °C. The catalyst was 1.25% $CoCl_2$ relative to $NaBH_4$. (a) Temperatures measured in the lower chamber where T_1, T_2, T_3, and T_4 were placed 0, 4, 8, and 10.5 inches relative to the perforated bottom plate, Figure 1a. Thermocouple T_4 was used to control the reaction. (b) Temperatures measured in the upper chamber at the trigger assembly, T_5, and the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure in the hose and total hydrogen gas. (d) Measured flow rate.

Runs 8 and 9 were conducted to generate 165 ft^3 of hydrogen gas using the 4.6:1.0 H_2O:$NaBH_4$ ratio. Thermocouple T_2, which was 4 inches above the bottom of the basket, was used for control as this thermocouple would stay immersed in the reagents for the duration of the run. Run 8 used 1.25% catalyst. Results are summarized in Table 1. The reaction was well controlled though the duration was considered excessive. In Run 9, the catalyst was increased to 2%. Results are summarized in Table 1 and Figure 7. The thermocouples in the lower chamber, T_1–T_4, were placed 0, 4, 10.5, and 8 inches above the bottom of the basket, respectively. There were two thermocouples in the upper chamber. Thermocouple T_5 was at the trigger assembly and T_6 was 6 inches above T_5. Figure 7a shows plots of the temperatures measured by thermocouples T_1–T_4 as a function of time. It took 6 minutes for the reaction to reach the set temperature of 50 °C. Thermocouples T_1, T_2, and T_4 track one another; while thermocouple T_3 recorded four heat bursts. Given its position in the lower chamber, thermocouple T_3 was closest to the air/reagent interface, which was the hottest region of the reaction. When the hydrogen gas reached the upper chamber, the temperature of the gas was 18–35 °C, Figure 7b. The temperature of the gas exiting the hydrogen generation unit, T_{exit}, was 20–30 °C. Figure 7c shows the pressure in the hose and the total hydrogen volume as a function of time. The total hydrogen profile is nearly linear. The hydrogen gas flow rate as a function of time is shown in Figure 7d.

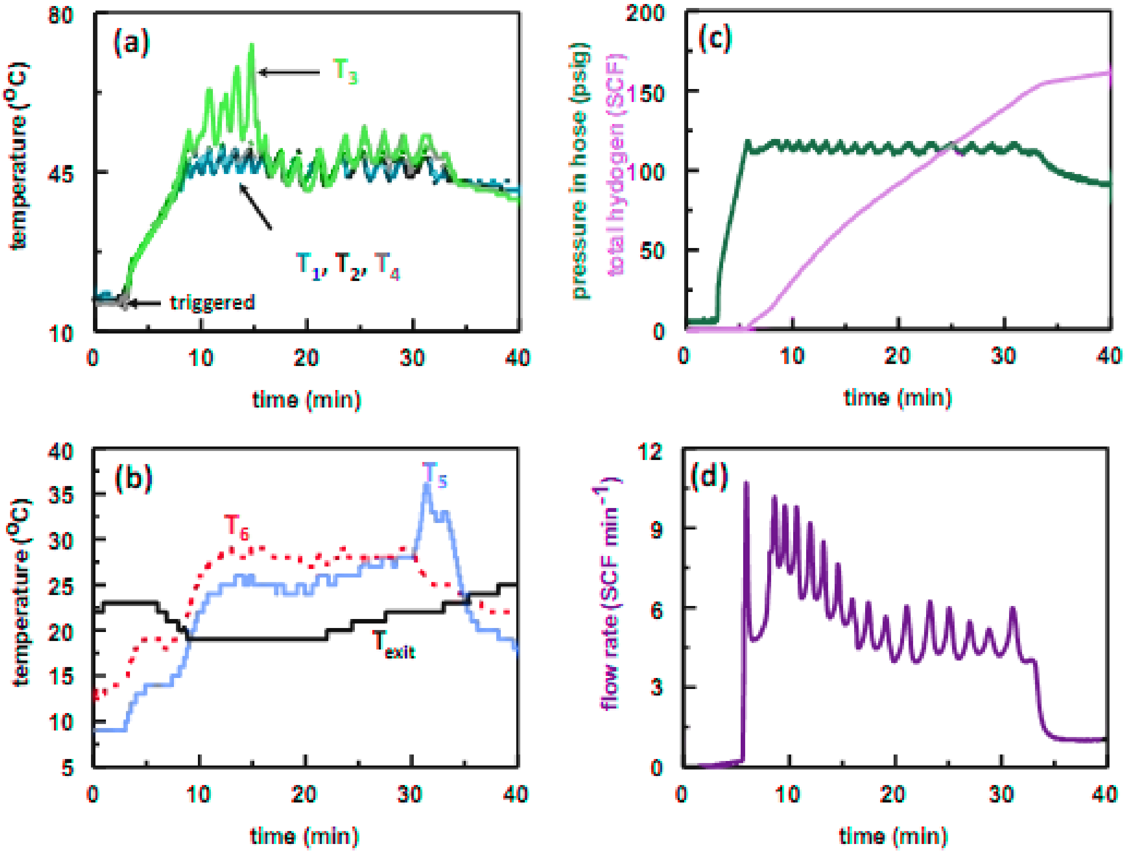

Figure 7. Results measured as a function of time for the 165 SCF H_2 gas generation, 4.6:1 H_2O:$NaBH_4$ ratio run using the configuration shown in Figure 1a. The test was carried out at a temperature of 15 °C. The catalyst was 2.00% $CoCl_2$ relative to $NaBH_4$. (a) Temperatures measured in the lower chamber where T_1, T_2, T_3, and T_4 were placed 0, 4, 10.5, and 8 inches relative to the perforated bottom plate, Figure 1a. Thermocouple T_2 was used to control the reaction. (b) Temperatures measured in the upper chamber at the trigger assembly, T_5, and 6 inches above the trigger assembly, T_6, and the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure into the hose and total hydrogen gas. (d) Measured flow rate.

Run 10 was conducted to generate 214.5 SCF of hydrogen gas (65% of capacity). The results are summarized in Table 1. The temperature, flow rate, hose pressure, and total hydrogen profiles were similar to Run 1 and Run 2 described earlier.

The results using the configuration shown in Figure 1a indicate that cooling coils need to be present at or near the reagent/air interface to generate 330 SCF of hydrogen gas in a reasonable amount of time without thermal runaway. To this end, subsequent testing used the configurations shown in Figures 1b and 1c.

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4. RESULTS OBTAINED USING THE THREE CHAMBER, TWO SETS OF COOLING COILS CONFIGURATION

In the tests using the configuration shown in Figure 1b, the influence of cooling water temperature was investigated. In addition, the influence of various control temperature profiles was also investigated. Results are summarized in Table 2. Note that the run numbers in the left hand column of Table 2 start with Run 1. That is, the run numbering has been reinitialized for the testing covered by Table 2. The reader is cautioned not to confuse the runs in Table 2 with those in Table 1.

Figure 8 summarizes a test that was conducted at 15 °C using 2.76 % $CoCl_2$. Thermocouple T_1, positioned 10 inches above the base of the basket, was used for control. The placement of the thermocouples in the lower and middle modules relative to the bottom of the basket are indicated in Table 2. In the upper chamber, thermocouple T_5 was at the trigger assembly and thermocouple T_6 was 6 inches above T_5. A thermocouple, T_{exit}, measured the temperature of the gas entering the hose. Figure 8a shows plots of the temperatures measured by thermocouples T_1–T_4 as a function of time. Recorded temperatures overshot the initial set temperature of 30 °C, peaking at 44 °C before cooling brought the reaction under control. At the 17-minutes mark, the set temperature was manually increased to speed up the reaction. As shown in Figure 8a, thermocouples T_1–T_3 track one another. At the 40-minutes mark, thermocouples T_2 and T_3 begin to diverge from T_1 and record lower temperatures than T_1. Thermocouple T_4 records a lower temperature than thermocouples T_1–T_3 until the 30-minutes mark when all four thermocouples track one another. The reaction reached completion at the 55-minutes mark after which the temperature recorded by thermocouple T_1 remained constant while the temperatures measured by thermocouples T_2–T_4 decreased rapidly. These results indicate that thermocouples T_1–T_3 are in contact with the reagents until the 40-minutes mark, at which time only thermocouple T_1 maintains contact. At the 30-minutes mark, the reagents are in contact with thermocouple T_4. To verify that the reagents had reached the middle chamber in contact with thermocouple T_3, a permanent marker was used to draw a vertical line on the inside wall of the middle chamber. This line was used to determine the maximum level the reagents reached inside the middle chamber. The line was faded where it was in contact with the reagents. The maximum reagent level corresponded to the position of T_3.

Figure 8b shows the temperatures of T_5, T_6, and T_{exit}, as a function of time. The plots show that the temperature of the gas inside the hydrogen generation unit was between 20 and 44 °C while the temperature of the gas entering the hose was between 15 and 35 °C. The plot of total H_2 volume as a function of time is nearly linear, Figure 8c. Figure 8d shows that the H_2 gas flow rate increased as the set temperature increased.

The initial overshoot in temperature that was observed in Run 1 (of Table 2) indicated that there was too much catalyst. Five runs were then conducted using less catalyst (2.25%) and an ambient operational temperature of 15 °C. That is, the cooling water reservoir and external bath temperature were cooled to 15 °C prior to triggering the reaction. In these runs, the thermocouple used to control the reaction, temperature control (T_C), was 6 inches above the base of the basket. This change was made to ensure that the control thermocouple was always in contact with the reagents. The placement of the thermocouples in the lower and middle modules relative to the bottom of the basket are indicated in Table 2. The temperature profiles used to control the reaction are also summarized in Table 2.

Table 2. Summary of hydrogen generation runs using the three chamber, two sets of cooling coils configuration shown in Figure 1b. In these runs, 330 SCF of hydrogen gas was generated and the $H_2O:NaBH_4$ ratio was 4.6:1. Run numbers here represent different tests than those in Table 1.

Three Chamber Run Number	% $CoCl_2$ catalyst[a]	T of Water Baths (°C)[b]	Height of Control T_C or T_{TH} (in)[c]	Height of T_1-T_4 (in)[c]	Set T (°C)
1	2.76 (Figure 8)	15	$T_C = 10$	$T_1 = 16$ $T_2 = 18$ $T_3 = 22$	30, then step to 32, 34, 36, 38, 40, 42, 44, 46, 48, 50, 55
2	2.25 (Figure 9)	15	$T_C = 6$	$T_1 = 16$ $T_2 = 18$ $T_3 = 22$	45, then step to 55 when 13.7 minutes into the reaction
3	2.25 (Figure 10)	15	$T_C = 6$	$T_1 = 16$ $T_2 = 18$ $T_3 = 22$	50, then step to 55 when 10.3 minutes into the reaction
4	2.25 (Figure 11)	15	$T_C = 6$	$T_1 = 16$ $T_2 = 18$ $T_3 = 22$	Ramp 42 to 57 with no derivative
5	2.25 (Figure 12)	15	$T_C = 6$	$T_1 = -1.1$ $T_2 = 16$ $T_3 = 22$	Ramp 42 to 57 with derivative
6	2.25	15	$T_C = 6$	$T_1 = 3$ $T_2 = 16$ $T_3 = 18$	Ramp 42 to 57 with derivative
7	2.25	25	$T_C = 6$	$T_1 = 10$ $T_2 = 16$ $T_3 = 18$	45 step to 50 when 11.7 minutes into reaction, 55 when 21.4 minutes into reaction

a. Percent $CoCl_2$ is relative to the mass of $NaBH_4$. Figure numbers associated with each run are indicated.

b. The hydrogen generation reactor, cooling water, and gas hose were immersed in water at the temperatures indicated.

c. Height of the control thermocouple (T_c) or thermistor (T_{TH}) is relative to the perforated bottom plate, Figure 1b.

Table 2. Summary of hydrogen generation runs using the three chamber, two sets of cooling coils configuration shown in Figure 1b. In these runs, 330 SCF of hydrogen gas was generated and the $H_2O:NaBH_4$ ratio was 4.6:1. (Continued)

Three Chamber Run Number	% $CoCl_2$ Catalyst[a]	T of Water Baths (°C)[b]	Height of Control T_C or T_{TH} (in)[c]	Height of T_1-T_4 (in)[c]	Set T (°C)
8	2.25 (Figure 13)	25	$T_C = 6$	$T_1 = -1.1$ $T_2 = 16$ $T_3 = 22$	Ramp 42 to 57 with derivative
9	2.25 (Figure 14)	0	$T_C = 6$	$T_1 = -1.1$ $T_2 = 16$ $T_3 = 22$	Ramp 42 to 57 with derivative
10	2.25 (Figure 15)	15	$T_{TH} = 6$	$T_1 = -1.1$ T_2, $T_3 = 6$ $T_4 = 21$	Ramp 42 to 57 with derivative

a. Percent $CoCl_2$ is relative to the mass of $NaBH_4$. Figure numbers associated with each run are indicated.

b. The hydrogen generation reactor, cooling water, and gas hose were immersed in water at the temperatures indicated.

c. Height of the control thermocouple (T_c) or thermistor (T_{TH}) is relative to the perforated bottom plate, Figure 1b.

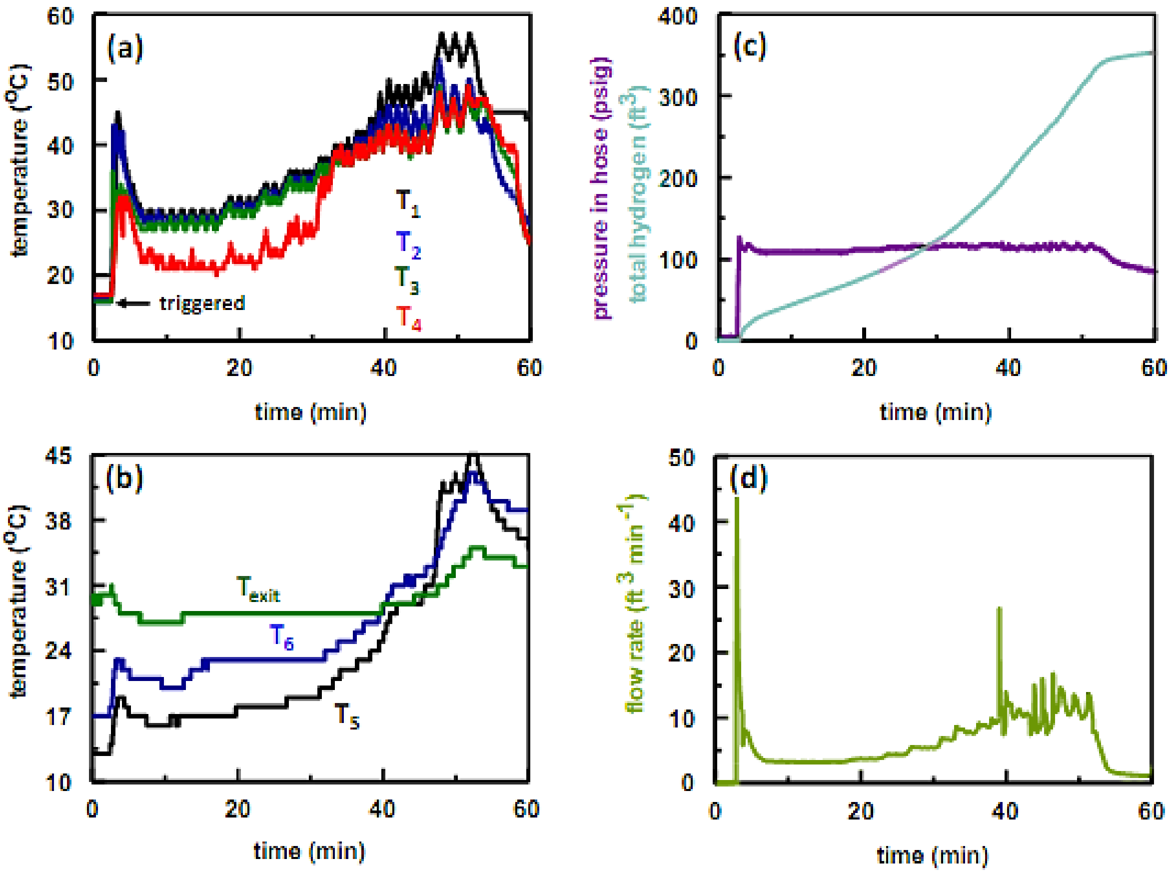

Figure 8. Results measured as a function of time for the 330 SCF H_2 gas generation, 4.6:1 H_2O:$NaBH_4$ ratio run using the three chamber, two sets of cooling coils configuration, Figure 1b. The test was carried out at a temperature of 15 °C. The catalyst was 2.75% $CoCl_2$ relative to $NaBH_4$. (a) Temperatures measured in the lower chamber. See Table 2 for placement of the thermocouples. (b) Temperatures measured in the upper chamber at the trigger assembly, T_5; 6 inches above T_5; T_6; and the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure in the hose and total hydrogen gas. (d) Measured flow rate.

Table 2 and Figure 9 summarize the results obtained for Run 2. In this run, the initial set point was 45 °C, but stepped up manually to 55 °C at the 16-minutes mark. The temperature reached the initial 45 °C set point in 4 minutes. No overshoots were observed for any of the thermocouples T_1–T_4, in Figure 9a, showing that the water flowing though both sets of cooling coils was efficiently carrying away the heat. No spikes in the pressure or flow rate were observed after the reaction was triggered, Figures 9c and 9d, respectively. For the first 17.6 minutes of reaction, thermocouples T_1–T_4 tracked one another. During this time, all four thermocouples were in contact with the reagents. At the 19.8-minutes mark, the temperatures recorded by thermocouples T_2–T_4 were lower than that of T_1, Figure 9a, indicating that these thermocouples were no longer in contact with the reagents. Thermocouples T_5, T_6, and T_{exit} stayed at or below 40 °C, Figure 9b. The flow rate increased when the set temperature was increased, Figure 9d. The total time of reaction was 28.7 minutes and the plot of total hydrogen gas volume as a function of time was nearly linear.

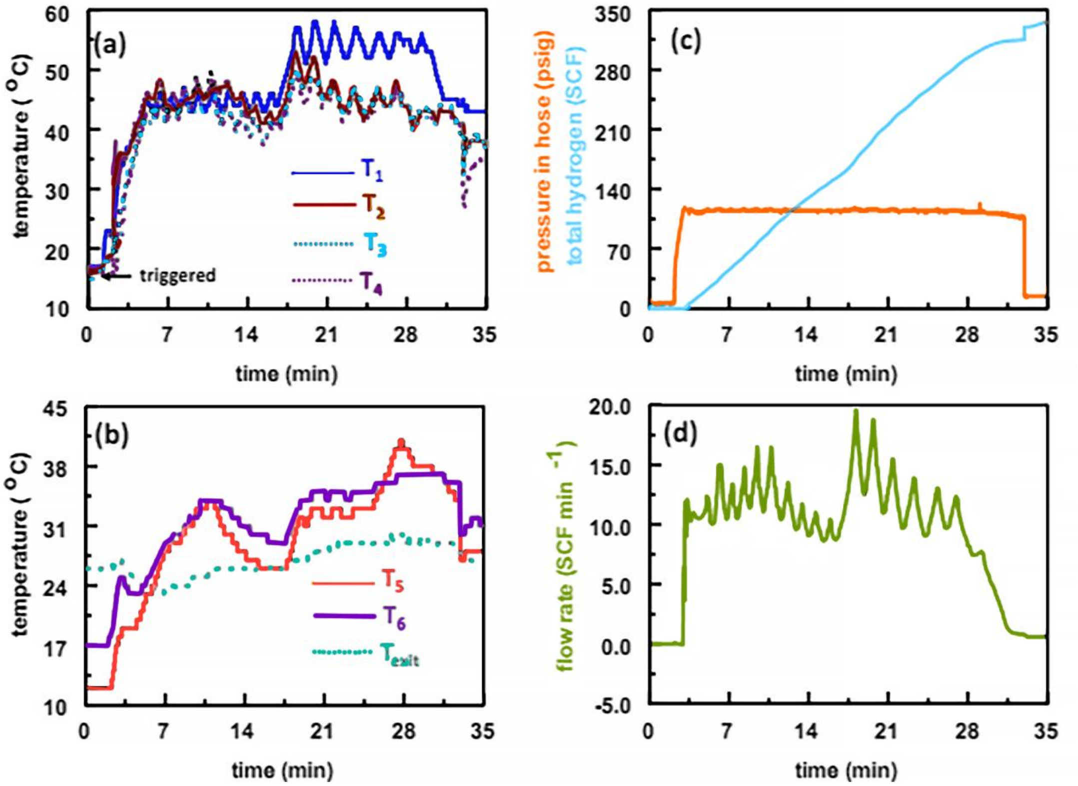

Figure 9. Results measured as a function of time for the 330 SCF H_2 gas generation, 4.6:1 H_2O:$NaBH_4$ ratio run using the configuration shown in Figure 1b. Table 2 lists experimental conditions. (a) Temperatures measured in the lower chambers. (b) Temperatures measured in the upper chamber at the trigger assembly, T_5; 6 inches above T_5, T_6; and the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure into the hose and total hydrogen gas. (d) Measured flow rate.

Results of Run 3 are summarized in Table 2 and Figure 10. Run 3 was conducted using an initial set temperature of 50 °C, followed by a manual step to 55 °C 10.3 minutes into the reaction. Run 3 flow rate and total hydrogen profiles are similar to those of Run 2. Except for the time interval 6.3–9.1 minutes into the reaction, the temperature profiles were also similar to those observed for Run 2. In the 6.3–9.1 minutes interval, T_1–T_3 registered higher temperatures than T_C and peaked at a temperature of 65 °C. Because the Run 3 set temperature was initially higher and occurred earlier than for Run 2, the total reaction time (21.5 minutes) was shorter than that of Run 2.

Control of the reaction rate was automated for runs 4, 5, 6, 8, 9 and 10. To automate control of the reaction, a predetermined temperature profile was enforced by a control algorithm. This profile ramped the set temperature from 42 to 57 °C in 1 °C increments every 2 minutes. This temperature ramp began when the reaction was activated. Previous runs imposing a single set temperature to control the reaction exhibited overshoots and undershoots of up to ±3 °C that produced large excursions in the hydrogen gas flow rate. To dampen these excursions and produce a smoother gas flow rate, a derivative component was added to the temperature control algorithm. That is, a proportional and derivative control (PD) system [3] was employed in addition to ramping the temperature. The results of the automated temperature control with and without derivative components are summarized in Table 2 for Run 5 (Figure 12) and Run 4 (Figure 11) respectively. For these runs, the placement of thermocouples is indicated in Table 2.

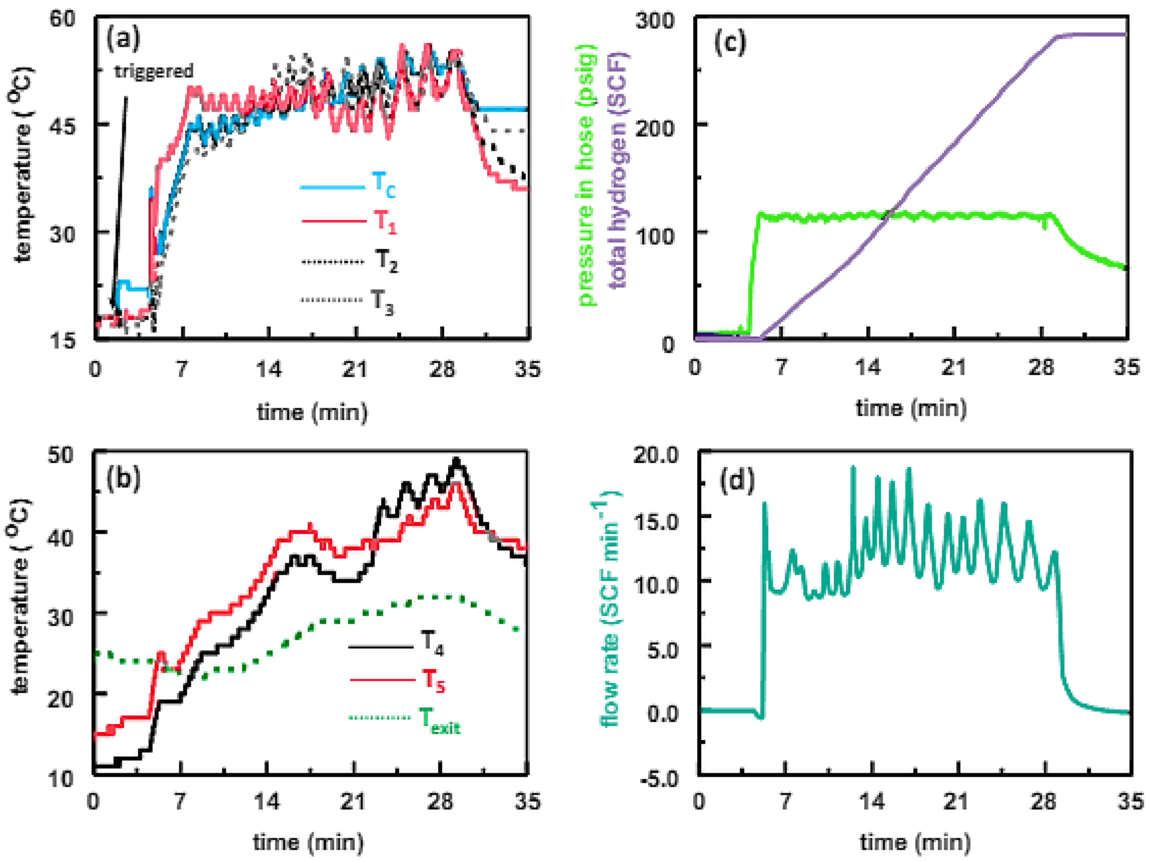

Figure 10. Results measured as a function of time for the 330 SCF H_2 gas generation, 4.6:1 H_2O:$NaBH_4$ ratio run using the configuration shown in Figure 1b. Table 2 lists experimental conditions. (a) Temperatures measured in the lower chambers. (b) Temperatures measured in the upper chamber at the trigger assembly, T_4; 6 inches above T_4, T_5; and the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure into the hose and total hydrogen gas. (d) Measured flow rate.

With no derivative component, Run 4 took 5.7 minutes for the reaction to reach the set temperature (44 °C) before cooling started, Figure 11a. Unlike previous runs where the temperatures T_C and T_1–T_3 instantly increased when the water from the upper chamber entered the lower ones, only a slight increase in temperature was recorded during Run 4 by thermocouple T_C, Figure 11a. This was probably due to the fact that the unit had been assembled 32 days prior to conducting the run. During that time, the sodium borohydride had become compacted. Consequently, extra time was required to get the sodium borohydride into solution. Initial temperature overshoots and undershoots were ±2 °C. By the 14-minutes mark, these undershoots and overshoots increased to as much as ±6.5 °C. The resultant flow rate, Figure 11d, showed excursions between 0.3 and 6.9 SCF per minute. The temperature of the gas exiting the unit varied between 23 and 32 °C, Figure 11b. Total hydrogen as a function of time was nearly linear, Figure 11c. When the derivative component was added to the control algorithm, the temperature overshoots and undershoots were dampened, as shown in Figure12a, and excursions in the flow rate diminished, Figure 12d. As shown in Figure 12c, the total hydrogen gas as a function of time was linear. Run 6 was a repeat of Run 5 and produced similar results.

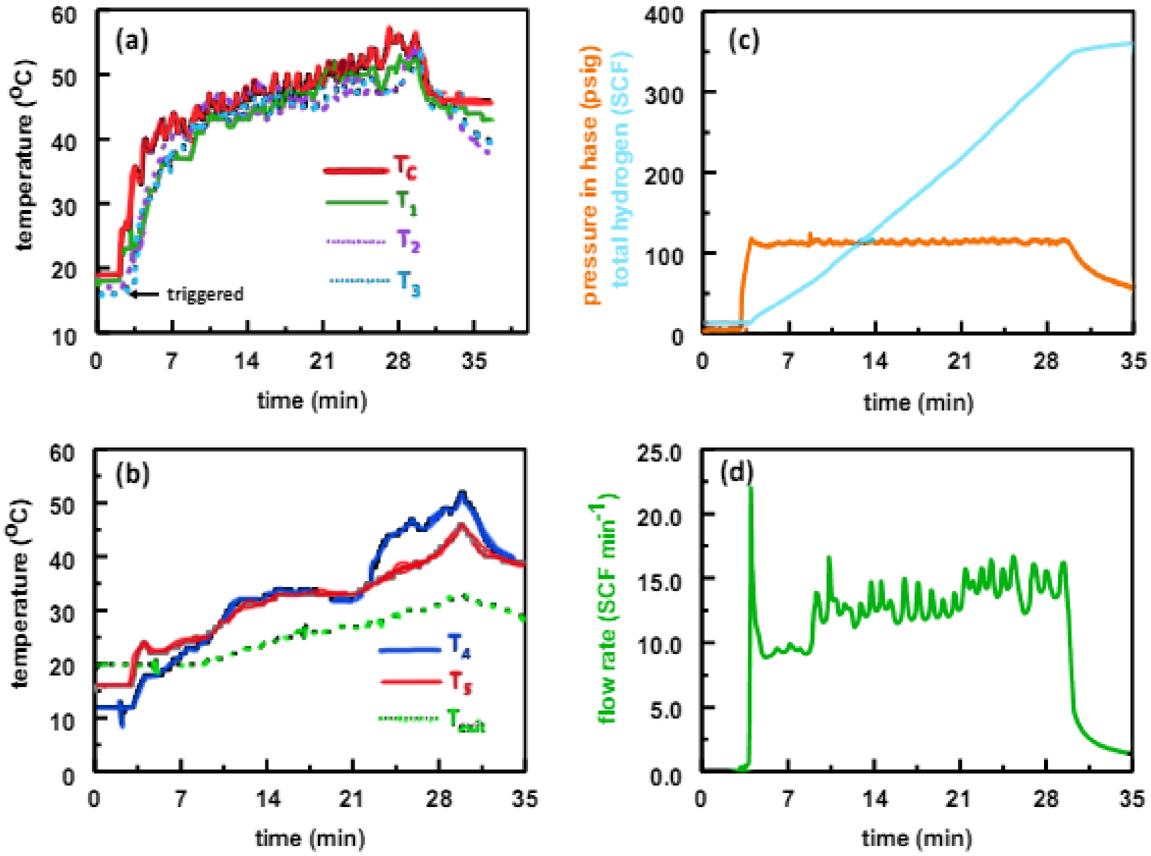

Figure 11. Results measured as a function of time for the 330 SCF H_2 gas generation, 4.6:1 H_2O:$NaBH_4$ ratio run using the configuration shown in Figure 1b. Table 2 lists experimental conditions. (a) Temperatures measured in the lower chambers. (b) Temperatures measured in the upper chamber at the trigger assembly, T_4; 6 inches above T_4, T_5; and the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure into the hose and total hydrogen gas. (d) Measured flow rate.

Up until this point, all experiments were conducted in a water bath temperature of 15 °C. Runs 7 and 8 of Table 2 were conducted in bath temperatures of 25 °C. Run 9 of Table 2 was conducted in a bath temperature of 0 °C. The results of Run 7, a manually controlled Run at 25 °C, are summarized in Table 2. For this Run, very few saw teeth were observed in the temperature and flow rate profiles. Cooling was active for approximately 80 % of the reaction period. The flow rate varied between 8.5 and 23 SCF per minute. This was not a well-controlled reaction. Better control was achieved during Run 8 using the automated ramp with a derivative component, Figure 13. Run 9 was operated in a 0 °C bath and used the automated ramp with a derivative component for temperature control. Run 9 results are summarized in Figure 14. As shown in Figure 14a, it took 13.8 minutes for this 0 °C “cold start” reaction to reach the set temperature (46 °C) at which time cooling was automatically activated by the control algorithm. By comparison, it only took 1.5 minutes to reach the set temperature (46 °C) for a “warm start” bath temperature of 25 °C as seen in Figure 13a for Run 8. Flow rate undershoots and overshoots during Run 9 were larger than those observed during the 25 °C Run 8.

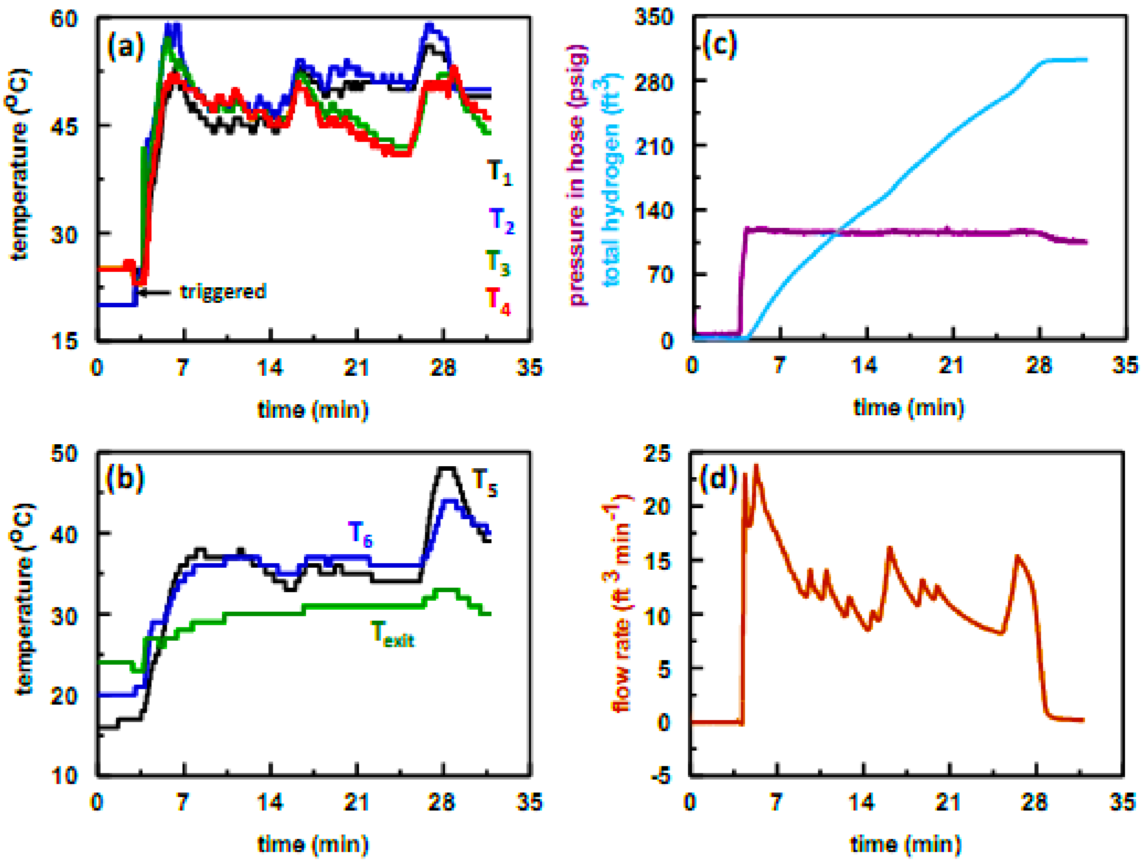

Figure 12. Results measured as a function of time for the 330 SCF H_2 gas generation, 4.6:1 H_2O:$NaBH_4$ ratio run using the three chamber, two sets of cooling coils configuration, Figure 1b. Table 2 lists experimental conditions. (a) Temperatures measured in the lower chambers. (b) Temperatures measured in the upper chamber at the trigger assembly, T_5; 6 inches above T_5 T_6; and the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure into the hose and total hydrogen gas. (d) Measured flow rate.

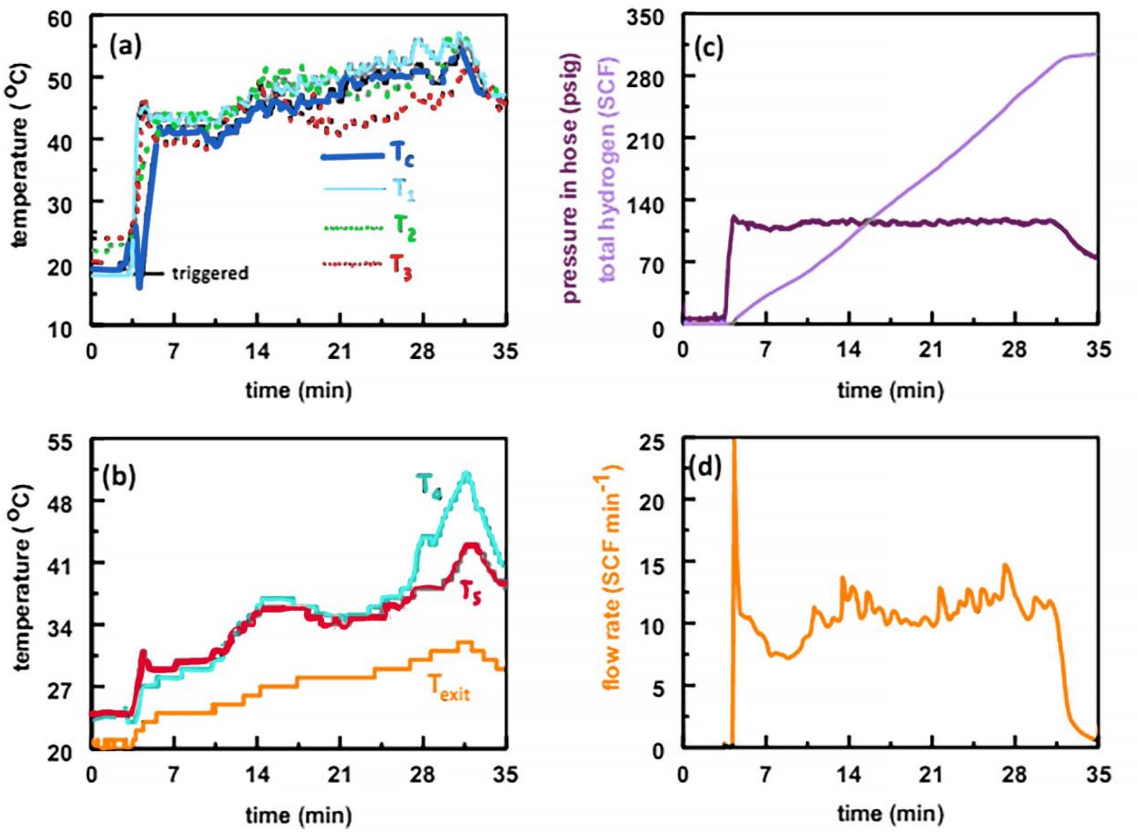

Figure 13. Results measured as a function of time for the 330 SCF H_2 gas generation, 4.6:1 H_2O:$NaBH_4$ ratio run using the configuration shown in Figure 1b. Table 2 lists experimental conditions. (a) Temperatures measured in the lower chambers. (b) Temperatures measured in the upper chamber at the trigger assembly, T_4; 6 inches above T_4, T_5; and the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure into the hose and total hydrogen gas. (d) Measured flow rate.

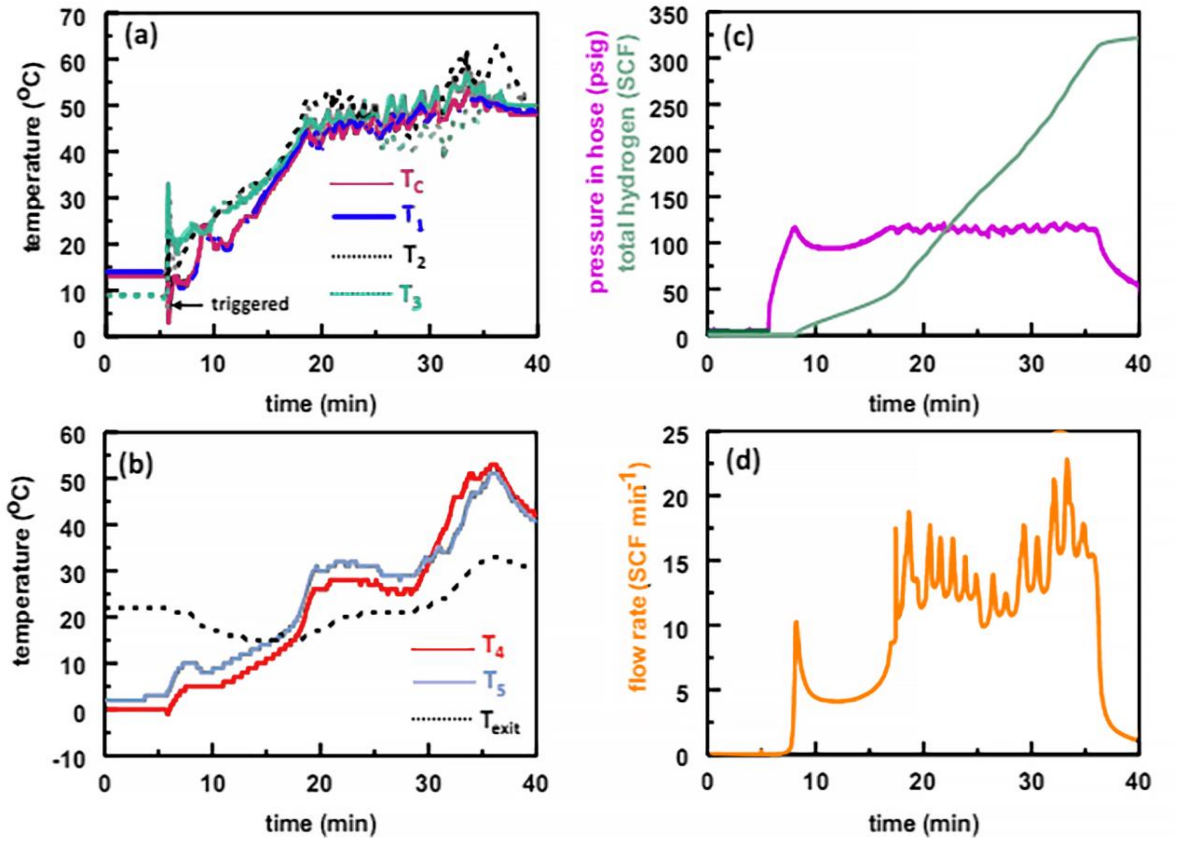

Figure 14. Results measured as a function of time for the 330 SCF H_2 gas generation, 4.6:1 H_2O:$NaBH_4$ ratio run using the configuration shown in Figure 1b. Table 2 lists experimental conditions. (a) Temperatures measured in the lower chambers. (b) Temperatures measured in the upper chamber at the trigger assembly, T_4; 6 inches above T_4, T_5; and the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure into the hose and total hydrogen gas. (d) Measured flow rate.

In Run 10, the control thermocouple was replaced with a thermistor. Thermocouples operate over a wider range of temperatures than thermistors; however, thermistors are better at sensing small changes in temperature. Consequently, a thermistor should theoretically improve the performance of the control algorithm's derivative component. In fact, this is what we see from the results of Run 10 as presented in Figure 15. The control algorithm with thermistor input substantially dampened both the temperature and flow rate undershoots and overshoots.

In all the runs summarized in Table 2, the % relative humidity of the gas exiting the hose was between 10 and 50 % lower than that of the outside air. Consequently, the gas exiting the hose was dry.

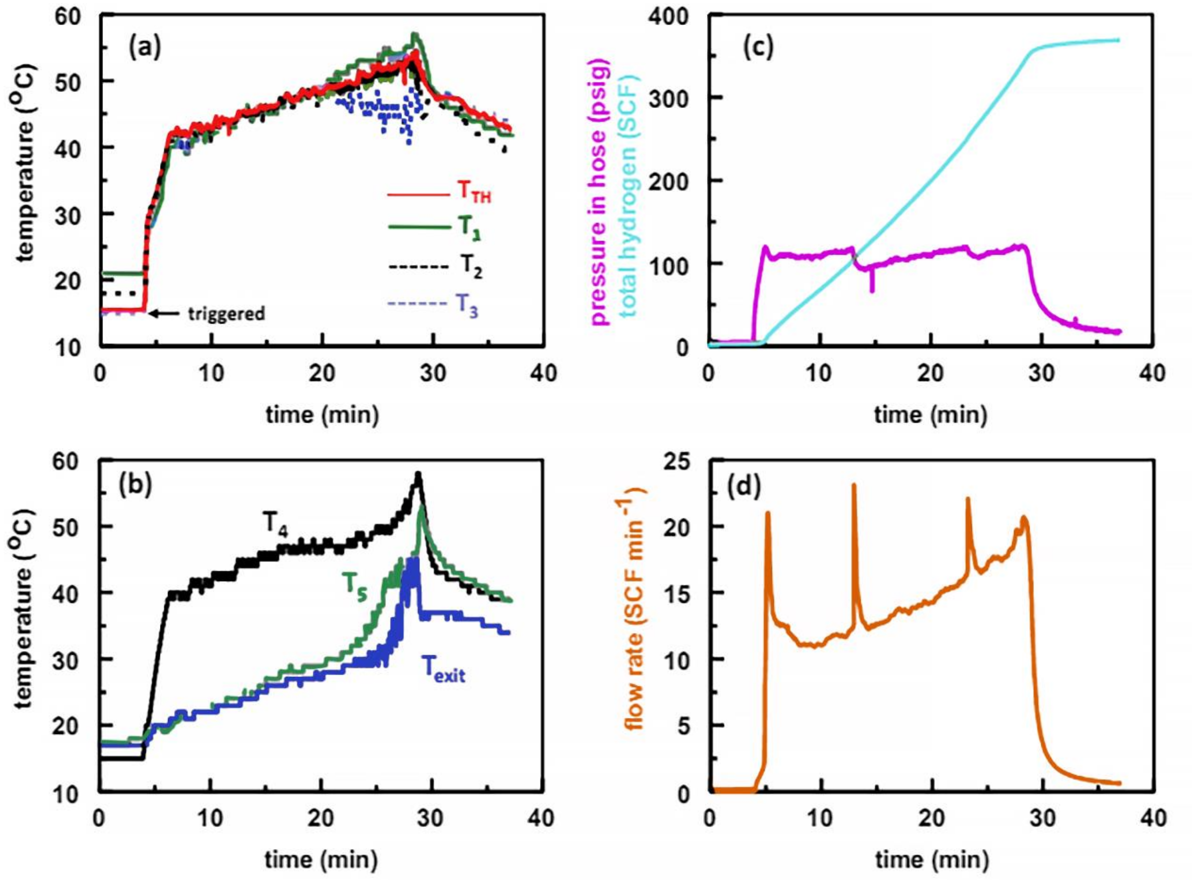

Figure 15. Results measured as a function of time for the 330 SCF H_2 gas generation, 4.6:1 H_2O:$NaBH_4$ ratio run using the configuration shown in Figure 1b. Table 2 lists experimental conditions. (a) Temperatures measured in the lower chambers. (b) Temperatures measured in the upper chamber at the trigger assembly, T_5, and the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure into the hose and total hydrogen gas. (d) Measured flow rate. Spikes in the flow rate are caused by sticking of the pressure release valve.

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5. RESULTS OBTAINED USING THE TWO CHAMBER, TWO SETS OF COOLING COILS CONFIGURATION

The purpose of the configuration, shown in Figure 1c was to minimize the separation between the two sets of cooling coils. Unlike the other two configurations, no basket was used to contain the reagents. Only a single test run was conducted with this configuration. The amount of $CoCl_2$ used in this reaction was 2.25%. The $CoCl_2$ was placed at the bottom of the lower chamber and the $NaBH_4$ was added on top of it. For this test the external bath temperature was 15 °C. Thermocouples T_1–T_4 were placed 3, 11, 16, and 22 inches from the bottom of the lower module, respectively. Thermocouples T_5 and T_6 were placed 17 and 11 inches from the top of the upper module, respectively. Thermocouple T_2 was used for control. The initial set temperature was 45 °C, which was then manually stepped up to 50 and 55 °C as the reaction progressed. Results are summarized in Figure 16.

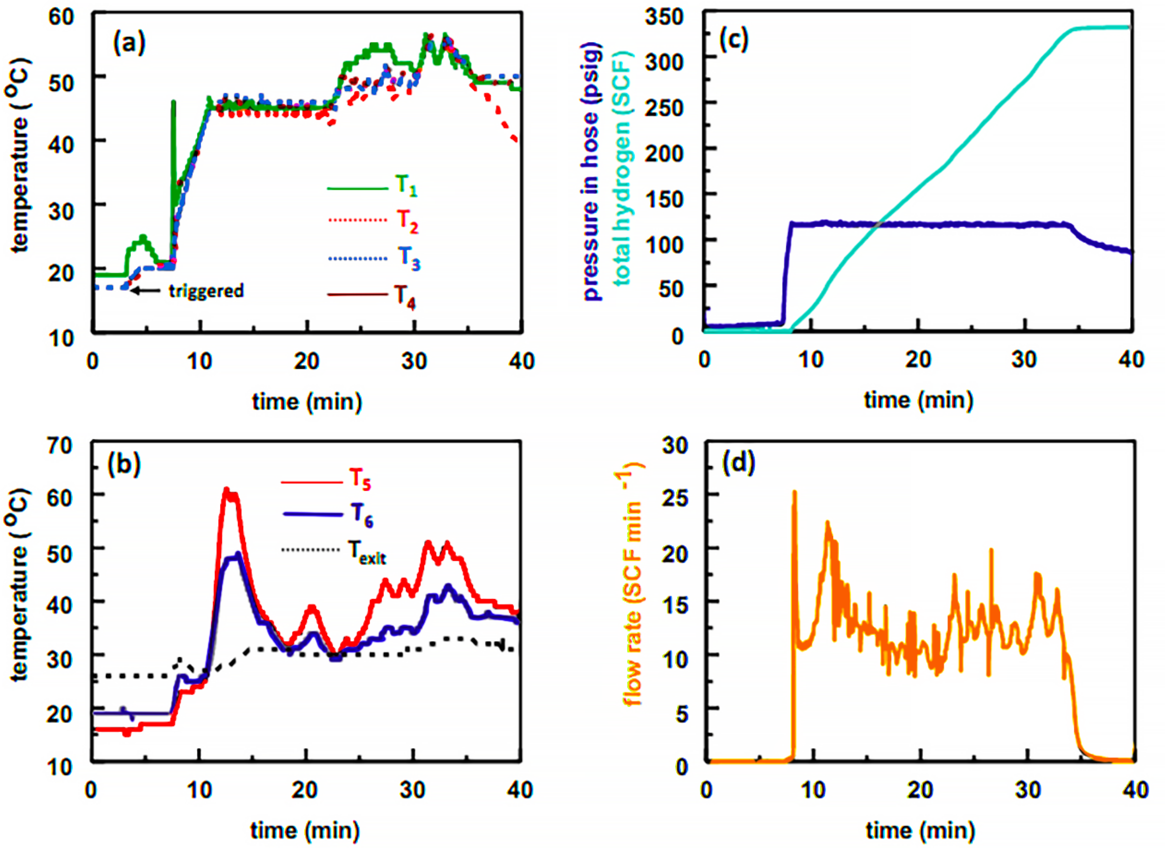

Figure 16. Results measured as a function of time for the 330 SCF H_2 gas generation, 4.6:1 H_2O:$NaBH_4$ ratio run using the configuration shown in Figure 1c. (a) Temperatures measured in the lower chamber. (b) Temperatures measured in the upper chamber at the trigger assembly, T_5 and T_6; plus the gas exiting the hydrogen generation unit, T_{exit}. (c) Measured pressure into the hose and total hydrogen gas. (d) Measured flow rate.

Figure 16a exhibits a 4 °C temperature bump registered by thermocouple T1 upon triggering. It took 7.7 minutes for the reaction to reach the initial set temperature of 45 °C. This delay in reaching the set temperature, relative to the two other configurations, is attributed to the absence of the basket. In the other two configurations, the sodium borohydride is confined to a cylindrical basket within the cylindrical space formed by the cooling coils. For the two configurations employing a cylindrical basket, water from the upper chamber flows unobstructed outside the cooling coils to the bottom of the reactor allowing the $CoCl_2$ and $NaBH_4$ to quickly react forming the catalyst that ignites the reaction. Absent a basket, the water has to percolate through 3.6 kg of $NaBH_4$ before reaching the $CoCl_2$. Despite the delay, the reaction was well controlled. The total reaction time was 33 minutes. The temperature of the gas exiting the hydrogen generation unit was 26–33 °C, Figure 16b. The flow rate as a function of time, Figure 16c, varied between 10 and 20 SCF per minute. The total hydrogen volume as a function of time was nearly linear, Figure 16d.

6. CONCLUSIONS

In this report, the results of testing hydrogen generation reactors capable of producing up to 330 SCF of hydrogen gas were discussed. The hydrogen generation reactor was modular and consisted of either two or three chambers. In these tests, the upper chamber housed the water while the lower chamber contained $NaBH_4$ and $CoCl_2$. A thermocouple or thermistor in the reagent bath was used to monitor and control the reaction. When producing 330 SCF of H_2, the reagent bath level from the lower chamber did reach the chamber above, despite the fact that the reaction occurred under pressure. For the configurations absent upper chamber cooling coils, thermal runaways occurred, as the hottest zone of the reaction was at the air/reagent interface. Consequently, a controlled reaction required configurations where cooling coils were always in contact with the air/reagent interface. A predetermined temperature profile was enforced as part of the reaction control algorithm. This temperature profile ramped the control temperature from 42 to 57 °C in 1 °C increments every 2 minutes. A derivative control component was used in addition to this linear step-wise temperature ramp. By using a control thermistor, capable of sensing small temperature changes, the derivative control algorithm produced a more even gas flow than runs employing a thermocouple for control. The enforced temperature profile functioned successfully for external bath operating temperatures of 0, 15, and 25 °C. For those reactions that were successfully controlled, the temperature of the gas exiting the hydrogen generation unit was below 40 °C. It was also shown that the relative humidity of the exiting gas was significantly below that of ambient indicating that the gas was dry.

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14. ABSTRACT

As manufacturing processes using helium have increased worldwide, so has the demand for helium. Since helium is a non-renewable resource, this increased use in manufacturing has led to dwindling supplies of helium worldwide and a corresponding increase in cost. At the same time the use of lighter than air (LTA) vehicles, such as aerostats for military and commercial applications, has increased. Traditionally these vehicles have been inflated using helium. However, with the rising cost and scarcity of helium, the use of hydrogen gas to inflate LTA vehicles has been gaining greater acceptance. This report focuses on the generation of hydrogen gas as an alternative to helium as a lighter than air gas source.

15. SUBJECT TERMS

Hydrogen benefits; hydrogen metal hydride; hydrogen gas generator; Thermocouple

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