SELF IGNITION OF HYDROGEN BY VARIOUS MECHANISMS[†]

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INTRODUCTION

With the almost inevitable transition to some form of hydrogen based economy, the potential for spontaneous or self ignition of hydrogen is clearly important. A number of mechanisms have been suggested which may account for this phenomenon and there have been studies previously by others to a greater or lesser degree. This paper reports the results of studies to investigate selected potential mechanisms for this behaviour:

- Sudden adiabatic compression in shock wave formation.
- Charging of a hydrogen jet leading to electrostatic ignition by corona discharge.
- Charging of particles within the hydrogen stream leading to electrostatic ignition by corona discharge.

Health and Safety Laboratory (HSL) have performed experiments in order to attempt to define conditions under which hydrogen can apparently self ignite and to confirm which mechanisms may account for this behaviour.

Experiments were performed to investigate:

- Ignition by adiabatic compression due to boundary layer failure.
- The current and polarity necessary to ignite pre-mixed clouds of hydrogen/air by corona discharge.
- The charge produced by a hydrogen jet emerging into free air both with and without entrained particles within the jet.

LITERATURE REVIEW

The first part of this project was a literature review, published separately (Gummer and Hawksworth 2007). The design of these experiments was based on Astbury and Hawksworth (2005), Golub et al. (2006) and Dryer et al. (2007).

In particular the work of Dryer gave a starting point for the experiments to investigate the ignition of sudden releases of compressed hydrogen to atmosphere. In addition to being significant as probably the first piece of experimental work to demonstrate ignition of releases into 'normal' everyday type environments, it was important as it also identified the influence of downstream obstructions on the propensity to ignite hydrogen.

Corona discharges are a continuous electrostatic discharges occurring when sharp points are raised to a high potential with respect to the surroundings, due to locally intensified electric fields near to the point. The potential at which a corona discharge will commence from a discharge point is given by the following equation:

$$V_o = 18\sqrt{r} \tag{1}$$

where: V_o = corona onset potential (kV)
r = radius of curvature of the discharge point (cm)

It is generally accepted that corona discharges are incapable of igniting typical hydrocarbon/air mixtures, though they have been implicated in the ignition of more sensitive flammable atmospheres such as hydrogen/air mixtures, such as anecdotal evidence of ignition of hydrogen vented from tall vent stacks.

EXPERIMENTAL PROGRAMME

PHASE 1 – DIFFUSION IGNITION TESTS

EXPERIMENTAL ARRANGEMENT

The work in this part of the programme involved tests using a bursting disc assembly and down-stream geometry similar to that used by Dryer; a photograph of the assembly is shown at Figure 1. The fitting marked A in the photograph was specially manufactured to take a Kistler pressure transducer so that the pressure profile in the cavity immediately downstream of the bursting disc could be recorded as the disc burst. A reducer B was used to connect various further downstream fittings, labelled C to K in Table 1. Some experiments were carried out with no fittings (i.e. hydrogen released directly from source). All the experiments were performed outside with the hydrogen released to open atmosphere. The hydrogen for the tests was supplied from HSL's 1000 bar experimental hydrogen facility; a schematic of the set up is shown at Figure 2.

RESULTS

A total of 85 tests were carried out using variations on downstream geometries and pressures. A list of the geometries tested is given in Table 1. With no fittings to restrict or reflect the flow, there was no ignition up to 831 barg. The lowest disc burst pressure at which an ignition was obtained was 35.5 bar; this corresponded to a transient cavity pressure (downstream of the bursting disc) of 28.6 bar. Examples of releases that resulted in ignition are given in Table 2, and examples of those that did not are given in Table 3. A plot of cavity pressure against burst pressure for ignitions and non-ignitions is shown at Figure 3.

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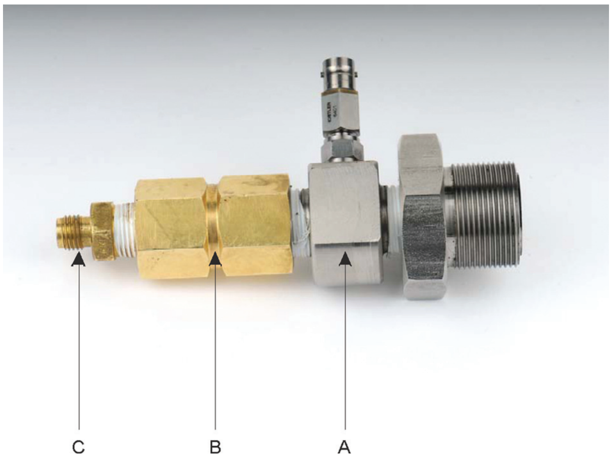

Figure 1. Geometry downstream of bursting disk

EXPERIMENTAL PROGRAMME PHASE 2, PART 1– IGNITION OF HYDROGEN/AIR MIXTURES BY GENERATED CORONA DISCHARGE

In these experiments, corona discharges were generated deliberately using a high voltage electrode in order to assess under what type of corona conditions hydrogen/air mixtures may be ignited.

EXPERIMENTAL ARRANGEMENT

A cylindrical vessel of dimensions (1.22 m dia × 1.70 m long) was used to investigate the conditions under which H_2-air mixtures may be ignited by corona discharges.

It was considered unlikely that in a well-earthed H_2 handling system a situation would arise where two conducting items were close to each other but at significantly different potentials. Therefore, initially, a wire electrode (0.38 mm diameter nichrome) was positioned centrally in the vessel, using the vessel walls (approximately half a metre away) as the "earthed" electrode. When no ignitions occurred with this arrangement, an additional electrode (a metal plate electrically bonded to the vessel) was placed approximately 30 mm away from the corona point to generate higher corona currents.

Table 1. Details of downstream flow geometry components used for burst disc experiments

Fitting ID	Fitting type
A	$\frac{1}{2}''$ NPT pipe nipple 2″ long with piezo transducer
B	$\frac{1}{2}''$ to 3/8″ NPT female brass reducing union
C	3/8″ NPT male to $\frac{1}{4}''$ male Swagelok tube reducer
D	3/8″ BSP plug (flat face) with 5.7 mm bore
E	3/8″ BSP plug (flat face) with 6.7 mm bore
F	3/8″ BSP plug (flat face) with 7.7 mm bore
G	3/8″ BSP plug (flat face) with 8.7 mm bore
H	3/8″ BSP plug (flat face) with 9.7 mm bore
I	3/8″ BSP plug (flat face) with 10.7 mm bore
J	3/8″ BSP plug (flat face) with 2.7 mm bore
K	3/8″ BSP plug (flat face) with 9.2 mm bore

The relationship between the applied potential and the magnitude of the resulting corona current has been studied by previous workers and a relationship of the following type has been proposed (Cross).

$$I = KV(V - V_o) \tag{2}$$

where: I = corona current (A)
V = applied potential (V)
V_o = corona onset potential (V)
K = a constant

The vessel was fitted with explosion relief consisting of physically weak electrically conducting plastic film that was physically and electrically bonded to the vessel to block out external static electric fields. The vessel was enclosed by a Faraday cage formed by an iron wire mesh supported on a frame. The current passing from the vessel to earth was measured using a Keithley Electrometer type 6514 in order to determine the corona current directly as a current measurement. The analogue voltage output of the Keithley was fed into a National Instruments (NI) A/D converter, type USB 6229, with a 16 bit resolution. The data was logged and processed using the LabView Signal Express software package. Very small background currents (about 100 pA) were detected from the test vessel, despite the presence of the Faraday cage, due to dust impingement. However, these were several orders of magnitude lower than the corona currents being studied and so did not introduce a significant error.

Attempts were made to ignite known concentrations of H_2-air mixtures by inducing a corona discharge within the gas mixture. The H_2 concentrations used in the tests were within a range around the concentration for which the lowest spark ignition energy is observed (i.e. 28% v/v) with a concentration of H_2 between 26% v/v and 33% v/v in air being employed. The concentrations of the H2-air mixture at the inlet and exhaust ports of the vessel were determined in the following way. The oxygen concentration was sampled using a calibrated Servomex 570 O_2 analyser and the H_2 concentration was calculated by difference. Figure 4 shows a schematic of the experimental arrangement used for these tests.

RESULTS

Data obtained in these tests show that corona currents appeared to vary with the square of the applied potential as proposed by others. This is demonstrated in Figure 5. It is interesting to note that the corona current was higher in the hydrogen/air mixtures than it was in air, for both positive and negative potentials applied to the wire.

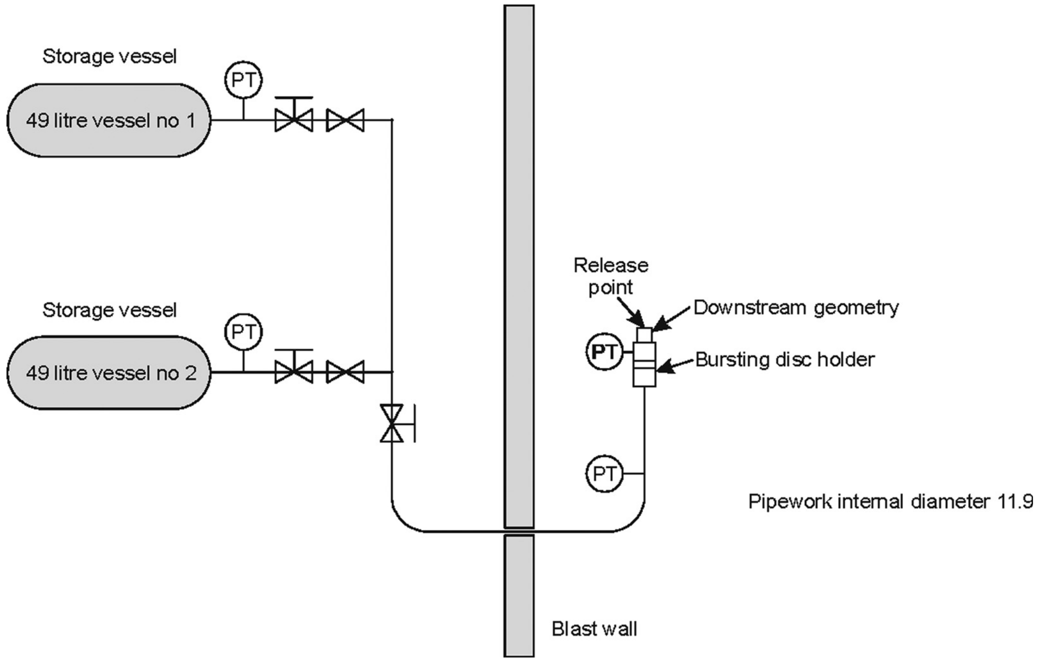

Figure 2. Simplified schematic of the spontaneous ignition high-pressure H_2 test facility

Table 2. Conditions for diffusion ignition from high-pressure H_2 releases

Test no	Disc material	Disc thickness (mm)	Downstream geometry	Cavity pressure (bar)	Burst pressure (bar)
Burst 48	Steel	0.203	A	24.17	241.3
Burst 47	Steel	0.254	A	28.2	390.3
Burst 45	Steel	0.305	A	28.81	419.0
Burst 39	Steel	0.178	A + B	17.09	154.1
Burst 41	Steel	0.178	A + B	19.78	192.5
Burst 60	Steel	0.178	A + B	30.27	159.0
Burst 58	Steel	0.178	A + B	16.24	151.0
Burst 19	Steel	0.305	A + B	39.06	411.0
Burst 69	Steel	0.381	A + B	28.56	439.5
Burst 20	Steel	0.051	A + B + C	28.56	36.9
Burst 04	Steel	0.127	A + B + C	83.86	112.0
Burst 05	Steel	0.127	A + B + C	86.10	115.2
Burst 10	Al (hard)	0.25	A + B + C	67.50	91.0
Burst 13	Steel	0.127	A + B + D	58.23	132.7
Burst 12	Steel	0.127	A + B + D	58.47	135.1
Burst 14	Steel	0.127	A + B + E	56.64	132.7
Burst 15	Steel	0.127	A + B + F	51.64	139.4
Burst 29	Steel	0.051	A + B + G	12.70	39.3
Burst 26	Steel	0.051	A + B + G	14.04	46.6
Burst 16	Steel	0.127	A + B + G	33.81	121.1
Burst 37	Steel	0.127	A + B + G	31.25	116.8
Burst 17	Steel	0.127	A + B + H	28.56	132.1
Burst 38	Steel	0.178	A + B + I	28.08	165.7
Burst 21	Steel	0.127	A + B + I	23.32	137.6
Burst 31	Steel	0.051	A + B + K	12.70	39.3

Table 3. Conditions for non-ignition from high-pressure H_2 releases

Test no	Disc material	Disc thickness (mm)	Downstream geometry	Cavity pressure (bar)	Burst pressure (bar)
Burst 44	Steel	0.178	A	18.31	161.99
Burst 42	Steel	0.178	A	20.63	187.62
Burst 43	Steel	0.178	A	20.02	182.13
Burst 51	Steel	0.203	A	22.22	221.19
Burst 52	Steel	0.178	A	17.21	179.08
Burst 55	Steel	0.038	A	24.17	259.03
Burst 50	Steel	0.203	A	19.41	218.14
Burst 49	Steel	0.203	A	19.29	216.92
Burst 59	–	–	A + B	37.96	105
Burst 40	Steel	0.127	A + B	15.99	122.92
Burst 08	Steel	2 × 0.127	A + B	27.47	279
Burst 68	Steel	0.381	A + B	25.27	573
Burst 09	Al (soft)	0.12	A + B + C	16.6	22
Burst 11	Al (soft)	2 × 0.12	A + B + C	33.57	45
Burst 24	Steel	0.038	A + B + C	16.97	24.66
Burst 22	Steel	0.038	A + B + C	18.19	26.49
Burst 36	Steel	0.051	A + B + G	11.35	36.25
Burst 25	Steel	0.051	A + B + G	10.25	34.42
Burst 33	Steel	0.051	A + B + H	13.18	28.32
Burst 34	Steel	0.051	A + B + H	9.4	39.31
Burst 32	Steel	0.051	A + B + H	8.91	38.7
Burst 28	Steel	0.051	A + B + I	7.08	37.48
Burst 27	Steel	0.038	A + B + J	23.32	25.88
Burst 30	Steel	0.051	A + B + K	12.94	42.97
Burst 35	Steel	0.051	A + B + K	9.4	36.25

The results from all of the trials are given in Table 4. The only condition that resulted in ignition of the H_2-air mixtures was with a positive potential of >20 kV applied to the wire and with a 30 mm gap between the wire and the earthed electrode, the discharge having a current of approximately 150 μA. It is possible that the corona had transitioned into an arc by this stage. This ignition was repeated with H_2-air gas mixtures of 28% v/v and 30% v/v of H_2. No ignitions were obtained with a negative potential applied to the wire (corona currents of up to 290 μA).

EXPERIMENTAL PROGRAMME PHASE 2, PART 2 – INVESTIGATION OF CORONA DISCHARGES DURING HIGH-PRESSURE H_2 RELEASES

Experiments were performed to investigate whether the release of pressurised H_2 could result in incendive corona discharges and, ultimately, ignition of the released H_2.

EXPERIMENTAL ARRANGEMENT

An electrostatic field mill was positioned just below a stainless steel pipe, which had a 2″ nominal bore and length of

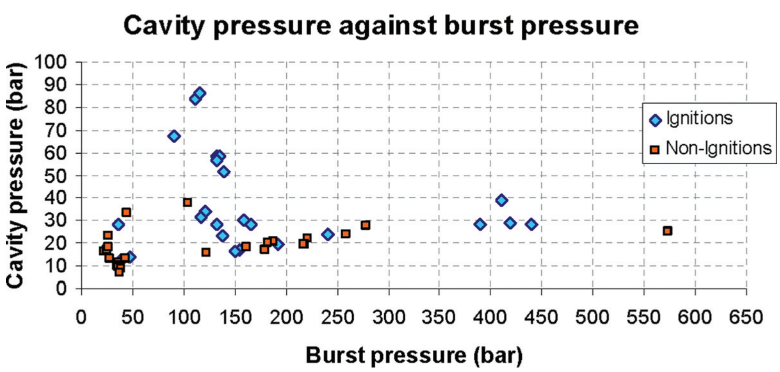

Figure 3. Plot of cavity pressure against burst pressure for cases of ignition and non-ignition

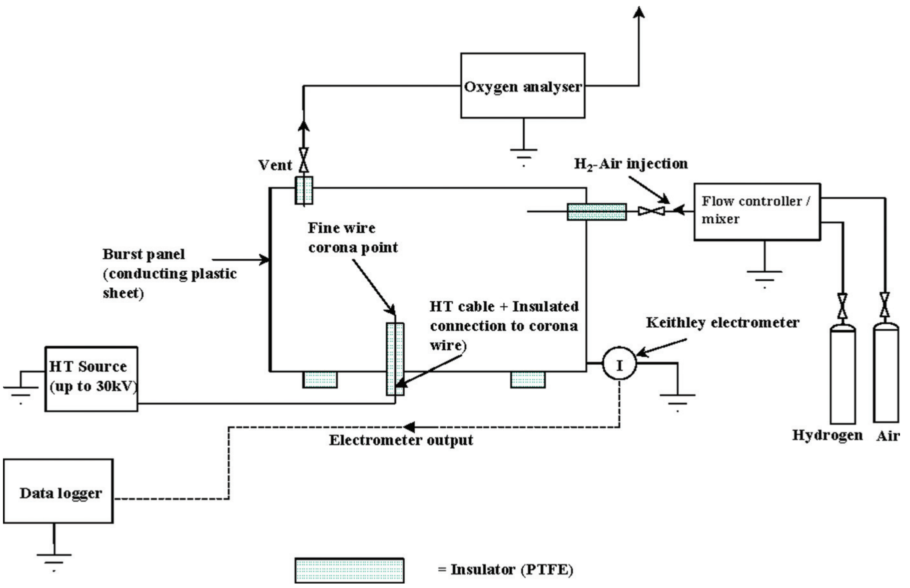

Figure 4. Schematic of the experimental arrangement used for ignition of H_2-Air mixtures from generated corona discharges

2.5 m, to measure the electric field in the direction of the H_2 release. Fine wires (0.38 mm diameter nichrome) were placed at various positions in an attempt to promote corona discharges.

H_2 was released from the high-pressure H_2 facility, for a duration of four seconds from a starting nominal pressure of 200 barg (i.e. typical pressure for commercially available cylinders). The released gas was directed into a stainless steel pipe with 2-inch nominal bore and length of 2.5 m. In some cases, powder was placed inside the pipe to be dispersed by the H_2 flow in order to increase the electrostatic effects. Two different dusts were used: a polypropylene plastic powder consisting of coarse (1–2 mm) and fine particles of a few hundred microns, and a fine (ca. 10 μm) iron (III) oxide powder (rust).

An Industrial Developments Bangor (IDB) Model 422 electric field meter was used to measure the electric field. The amount of charge on the dispersed dust was inferred by measuring the charge (of opposite polarity) transferred to the pipe. A Keithley Electrometer type 6514 was used to measure the voltage accumulated on a 1 μF capacitor connected to a 2.5 m pipe – to assess the charge transfer between the pipe and the powder. In some experiments, an alternative system was used to measure the charge on the powder; a John Chubb Instruments JCI178 charge-measuring device was used to measure the charge on the pipe directly. The analogue voltage outputs of these instruments were fed into an NI A/D converter, type USB 6229, with a 16-bit resolution and coupled to a PC driven by NI software (Signal Express and LabView). For corona detection, the corona wires were connected to earth via a 500-ohm resistor and the voltage generated across the resistor was used as the input to the high frequency Techtronic TDS784D oscilloscope; an

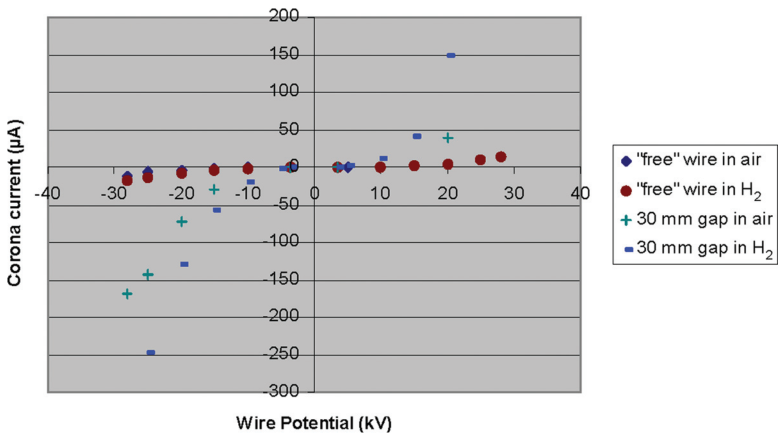

Figure 5. Corona current versus potential applied to the wire electrode

Table 4. Summary of ignition tests on hydrogen/air mixtures by corona discharges

Test run	Hydrogen conc'n (% v/v)	Electrode configuration	Max. potential of corona wire (kV)	Max. corona current (μA)	Ignition (Y/N)	Comments
1	28.4	0.38 mm nichrome wire in centre of vessel	−28	−16.7	N	
2	29.8	0.38 mm nichrome wire in centre of vessel	−28	−18.2	N	
3	30.3	0.38 mm nichrome wire in centre of vessel	+28	+14.0	N	
4	32.2	0.38 mm nichrome wire in centre of vessel	−28	−18.5	N	
5	32.2	0.38 mm nichrome wire in centre of vessel	+28	≈ +13	N	
6	25.5	0.38 mm nichrome wire in centre of vessel	−28	−16.2	N	
7	28.4	0.38 mm nichrome wire in centre of vessel	+28	+13.4	N	
8	30.0	0.38 mm nichrome wire, earthed plate 30 mm away	−28	≈ −290	N	
9	30.0	0.38 mm nichrome wire, earthed plate 30 mm away	+20	+150	Y	Ignition occurred as potential increased just above +20 kV; electrometer damaged by current surge
10	28.0	0.38 mm nichrome wire, earthed plate 30 mm away	−28	≈ −290	N	
11	28.0	0.38 mm nichrome wire, earthed plate 30 mm away	+20	+150	Y	Ignition occurred as potential increased just above +20 kV

image of the scope trace was saved to disc as required. The response of the oscilloscope to laboratory generated corona discharges was assessed before the trials to ensure that the system was capable in principle of detecting such discharges and to establish a typical trace for such an event.

A schematic of the experimental arrangement is shown at Figure 6.

RESULTS

No charges generated were sufficient to ignite hydrogen air mixtures in the experiments with dust dispersions of up to 160 g of iron oxide or polypropylene in hydrogen releases from 200 bar storage. However, there was some evidence that corona discharges may have been generated at the wire electrodes placed close to the dispersed hydrogen, although this was only evident when powder was also dispersed.

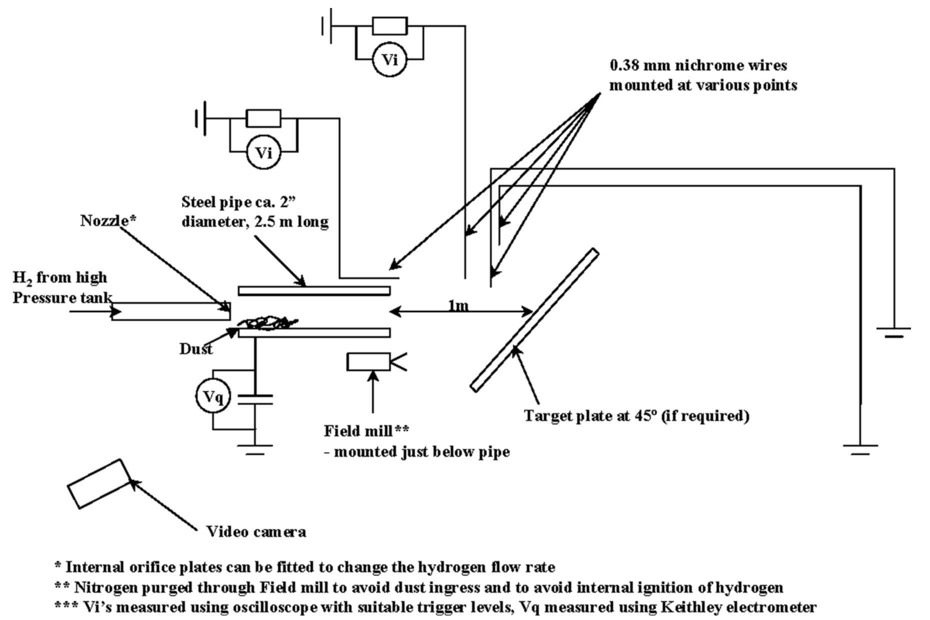

Figure 6. Schematic of the experimental arrangement used for investigating corona discharges during high-pressure H_2 releases

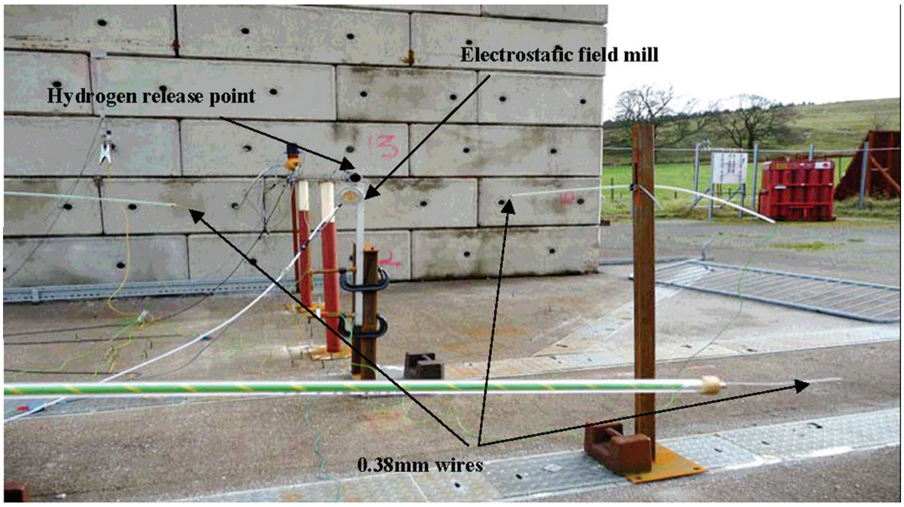

Figure 7. Arrangement to investigate electrostatic field/discharges during hydrogen release

CONCLUSIONS

For the first phase of experimental programme, the following conclusions were made for diffusion ignition tests involving bursting discs and pipe work downstream of a boundary failure:

- No ignitions were observed below a cavity pressure of 8.8 barg.
- Ignitions were always observed above a cavity pressure of 27.5 barg.
- Releases of H_2 to atmosphere with no restrictive and reflective downstream geometry present resulted in no ignitions up to a burst pressure of 831 bar.
- Tests which included reflective downstream geometry always produced ignitions at disc burst pressures above 260 bar.
- The lowest burst pressure for ignition was 35.5 bar with a reflective geometry configuration and vent area of 17.3 mm^2 and soft ductile bursting discs were less likely to produce an ignition than a non-ductile disc.
- Whether ignition occurs appears to be related to the rise time of the pressure pulse produced by the disc failing. The rise time is related to the burst pressure and also the extent of downstream constriction after the bursting disc.
- When H_2 leaked into cavity before a burst, no ignitions occurred.
- No ignitions were observed when weakened pipes were used for open geometry tests at pressures of up to 417 bar.

In the second phase of the experimental programme the following conclusions for tests involving corona discharges were attained:

- H_2-air mixtures were ignited by corona discharges generated by raising a fine wire to a high potential.

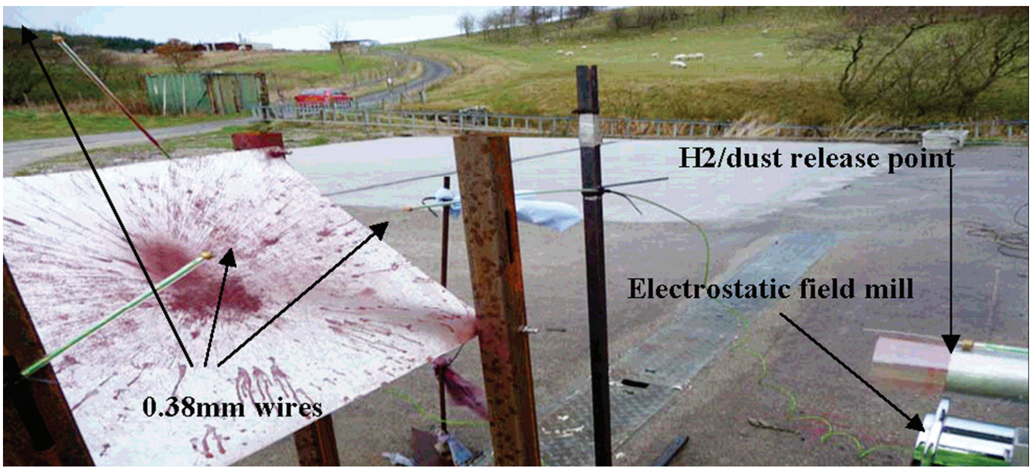

Figure 8. Arrangement to investigate electrostatic field/discharges during hydrogen release and dust dispersion

- Ignitions occurred with positive corona discharges at a current of approximately $+150\ \mu A$ and potential of $+20$ kV for a wire point and plate electrode system with a 30 mm separation.
- No ignition was observed with negative currents of up to approximately $-290\ \mu A$ and potential of -28 kV.
- Dispersion of dusts up to 160 g with H_2 released from 200 barg did not appear to generate hazardous electric fields, in terms of incendive corona discharges.
- Ignition can be produced by corona discharges of the type that might be produced where fine points may be at a potential of several tens of kV above the surrounding atmosphere. Such situations could be expected at the top of tall vent stacks, tens of metres above ground, in the presence of large atmospheric electric fields (e.g. during snow fall). Such incendive corona discharges appear to be unlikely in horizontal releases of H_2 close to ground level.

NOMENCLATURE

V_o = corona onset potential (kV)
r = radius of curvature of the discharge point (cm)
I = corona current (A)
V = applied potential (V)

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