IMPACT IGNITION OF HYDROGEN-AIR MIXTURES

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Due to the flammable nature of hydrogen in air mixtures, their presence requires formal assessment of safety issues. Such an assessment can take the form of probabilistic or deterministic analysis including a consideration of the consequences of events and the means of mitigation available. Data for use in such analyses are not always present in the literature. The case of an accidental impact ignition of a hydrogen in air mixture has been considered and experimental data obtained.

KEYWORDS: impact, ignition, hydrogen air explosion

INTRODUCTION

Hydrogen is used as a reagent in the chemical process industries and increasingly as a fuel for vehicular transport, e.g. buses. It can also be generated in waste storage plant within the nuclear industry. However, the flammable range of hydrogen in air is substantial and consequently generation and uses of hydrogen will always require various forms of safety/risk assessments. It is common to assume that formation of a flammable hydrogen in air mixture will inevitably lead to an explosion. This is usually a sensible approach, to ensure the safety case is robust. However, there are significant pessimisms in many areas in such safety cases, such as generation and release rates, ignition probabilities, and overpressures generated by an explosion. When multiplied together, these pessimisms are many orders of magnitude away from the expected behaviour. This may lead to considerable over-engineering, operational and maintenance costs and restrictions on throughput, which can significantly delay the decommissioning of plant, and prolong radioactive risk. In certain cases, it may be necessary to prepare a multi-legged safety case, in which a number of factors including their routes of mitigation are considered, since it may not be possible to prepare a deterministic argument. This allows a more realistic appreciation of the likely events, their consequences and their prevention/mitigation and so more clearly define engineering, operational and maintenance requirements.

In this paper, the decommissioning of a nuclear waste storage plant is considered. In nuclear waste storage, hydrogen is mainly produced by either corrosion (e.g. Magnox fuel cladding, which is a 99% Mg alloy) or by radiolysis of aqueous solutions. In most scenarios, measures such as ventilation or inerting can be relied upon to prevent flammable atmospheres forming. The safety case usually assumes the probability of ignition of a flammable mixture as 1, if a fault condition occurs. One potential fault scenario concerns retrieval of waste from silos where significant hold-up of hydrogen within a sludge bed has occurred. In such cases small but significant transient releases of hydrogen (and hence transient flammable atmospheres) could occur. It is possible to envisage a number of different mechanical impacts being caused by retrieval. Kinetic energies involved in such impacts could range up to several kJ. Projectiles could have un-constrained movement before or during impact. Impacts could involve either metal on concrete or metal on metal. The angle of impact could vary considerably giving rise to different components of tangential or normal impulses. Finally, there could be pyrophoric material and/or rust at the impact interface.

Under these conditions the essential prerequisites for an explosion to occur may be established: i.e. an ignition source caused by the mechanical impact and the presence of a flammable atmosphere. A series of experiments on impact ignition of hydrogen-air mixtures is described below, and an ignition road map has been developed.

Other ignition sources such as electrical discharges, are the subject of many other papers, and are not considered further here.

EXPERIMENTAL IMPACT IGNITION STUDIES

Much of the experimental work programme has centred on the use of drop weight type apparatus. This was chosen primarily as many of the envisaged impact scenarios involved the impact of freely moving bodies. Although clean metal on clean metal impacts have been studied, much of the work has been concerned with impacts involving either pyrophoric or thermite reactions with Magnox. In such cases the initial impact initiates a highly exothermic reaction, as indicated by the shower of white hot sparks produced, which is an effective ignition source for any surrounding flammable hydrogen/air atmosphere. Ignition involving thermite or pyrophoric reactions occur with projectile kinetic energies before impact often over an order of magnitude lower than for ignition between clean steels.

DESIGN AND OPERATION OF DROP WEIGHT IMPACT RIG

Much of the work carried out to date has been conducted using the drop weight impact apparatus, as shown in Figure 1. A projectile with an attached test piece is fired downwards towards an inclined steel plate bolted to a large heavy anvil. The angle of the 15 mm thick plate attached to the anvil could be varied between 25 and 65° to the horizontal. Inclined plates made from various materials were used.

The projectile used in the majority of tests was constructed from a 60 mm dia. steel cylinder filled with lead. The test pieces consisted of discs which were attached to the projectile body to give an overall length of 260 mm and mass of 7.3 kg. These were machined to give the impact edge a radius of 5 mm. To obtain high velocities, in a contained environment, a modified air cylinder was used to apply a large impulse to the projectile. The steel enclosure was vented through a cut out window so as to allow explosion of hydrogen gas with an overpressure of less than 0.1 bar. Before each experiment was carried out the vent opening was sealed with a sheet of plasticised film (cling film) that could be easily blown open when an explosion occurred. The gas concentration was monitored by drawing off

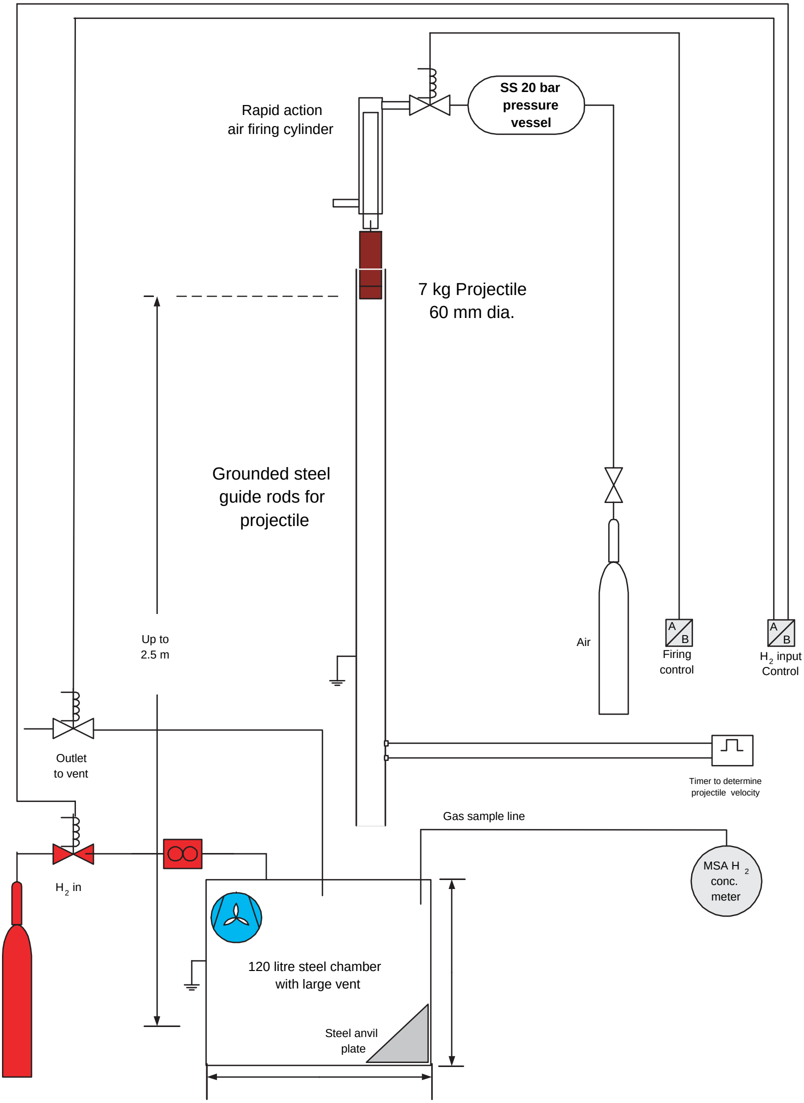


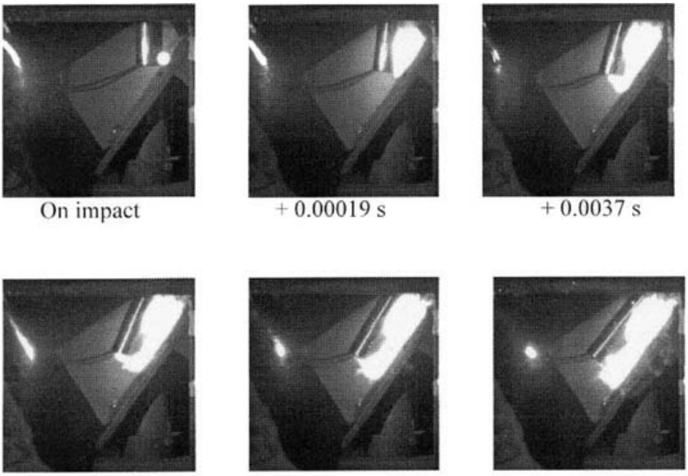
Figure 1. Schematic diagram of the drop weight impact apparatus

samples through a small bore brass tube which terminated in the near vicinity of the impact zone. Flammable gas concentration was measured using an AUER EX-METER 11 (P) analyser calibrated for hydrogen. The projectile was fired by operating the modified air cylinder or dropped under gravity depending on the impact velocity required to achieve final impact velocities of 5 m/s to 12 m/s. In some cases the impact event was also filmed using high speed video, enabling the work done by tangential and normal impulses to be determined. Frames from one such video are given in Figure 2.

PARAMETRIC SENSITIVITY

Because the ignition of hydrogen air atmospheres is dependent on a large number of parameters, LSBU has conducted specific experiments to identify and investigate the effect of the most important parameters. In some cases factorial experimental design was used (enabling the effect and interactions of a number of parameters to be studied simultaneously). The main parameters investigated were:

- hardness of the projectile and target
- available kinetic energy prior to impact



On impact + 0.00019 s + 0.0037 s

+ 0.0056 s + 0.0075 s + 0.0093 s

Impact with 4 kg projectile. Magnox/rust surface. 1071 PPS

55 degree plate. Impact velocity 11.2 m/s. 22% H/air

Figure 2. Images taken from high speed video of an impact ignition involving a thermite reaction

- angle of the inclined plate (target)
- wetting of the plate (target)
- hydrogen concentration
- the presence of rust on the plate (target)

Within the limitations of the test apparatus, these parameters have been investigated over ranges likely to be encountered in practice. Table 1 gives some indicative results obtained for a range of impacts, using the 7.3 kg projectile impacting a target inclined at 45 degrees. Based on these results the effects of the various parameters are discussed below.

Table 1. Ignition probability for 22% H_2/air mixture with 45 degree inclined plate. 15 tests carried out for each set of conditions (7.3 kg projectile)

	Head	Plate	Surface	m/s	Ignitions	KE (J)	Ig Pr.
1	ss	ss	clean	12	0	526	0.00
2	Inc	Inc	clean	12	0	526	0.00
3	ss	ss	Mag particles	8	15	234	1
4	ss	ss	Mag particles	6	13	131	0.87
5	ss	ss	Mag particles	5	5	91	0.33
6	ss	ss	Mag particles	4.7	4	81	0.27
7	ss	ss	Al particles	10	3	365	0.20
8	Al	ms	with added rust	10	6	365	0.40
9	Mag	ms	clean	10	0	365	0.00
10	Mag	ms	rusty surface	10	3	365	0.20
11	Mag	ms	rusty + Mag par	10	7	365	0.47
12	Mag	ms	Mag particles	10	0	365	0.00
13	ss	ss	dry sludge + Mag	10	6	365	0.40
14	ss	ss	dry sludge	10	2	365	0.13
15	ss	ss	dry sludge + Mag + rust	10	6	365	0.40
16	ss	ss	wet sludge + Mag	10	9	365	0.56
17	ss	ss	wet sludge	10	3	365	0.20
18	Mag	ms	with added rust	10	7	365	0.47
19	Al	ms	rusty surface	10	2	365	0.13
20	Inc	conc	clean	8	0	234	0.00
21	ss	conc	with wet Mag particles	10	7	365	0.70
22	ss	conc	with sludge	10	0	365	0.00
23	ss	conc	with dry Mag particles	10	13	365	0.87

Inc = Inconel, Mag = Magnox, ss = 18/8 stainless steel, ms = mild steel, Al = aluminium alloy (Duralumin), sludge = corroded Magnox, Conc = concrete, KE = kinetic energy.

HARDNESS OF PROJECTILE AND TARGET

Hardness has consistently been found to be the most significant factor affecting ignition probability in experiments performed at LSBU. For example, in a factorial experiment, investigating ignition from the impact of Magnox particles between surfaces, increasing hardness from a HV of 118 to 501 had a far more significant effect than increasing the kinetic energy of the projectile from 130J to 375J. Other factors included in the study (i.e. inclination of the target between 35 and 45 degrees, hydrogen concentration between 10 and 30% (v/v) and water content of Magnox particle layer (between 0 and 66% by weight) were found to be far less significant.

The role of hardness is also apparent by examination of Table 1 (runs 4 and 12), where an impact of Magnox particles between a stainless steel (HV 340) projectile and target gave a much higher ignition probability than for Magnox particles between a Magnox projectile (HV 65) and a mild steel target (HV 180). The difference between hard and soft materials is considered to relate mainly to the impact area. With softer projectiles considerable deformation of the projectile is found to take place on impact. The amount of energy dissipated (for a given KE (kinetic energy) prior to impact) in deformation of the material or as frictional heating is however (estimated from the volume of material deformed) not that different to that with harder materials. The smaller deformation on impact however results in much smaller contact areas and so although the total amount of frictional heat generated might be similar, the frictional heat flux is higher, producing higher surface temperatures.

KINETIC ENERGY AVAILABLE PRIOR TO IMPACT

The KE (kinetic energy) of the projectile has been found to be the second most significant factor. In terms of magnitude it is interesting to observe that the available KE required for ignition is not that different to results for methane/air [1, 2] (e.g. for High Mg content alloy projectiles striking a rusty surface). This points to the initiation of the thermite or pyrophoric reaction being the controlling factor. The subsequent ignition of hydrogen/air is a second order effect.

ANGLE OF THE INCLINED PLATE

Altering the angle will affect the amount of work done by normal (i.e. deformation) and tangential (friction) impulses. Frictional heating is largely responsible for temperatures generated on impact. Depending on additional factors (i.e. projectile geometry, coefficients of friction and restitution) frictional work peaks at an impact angle of around 45 degrees. At extremes (i.e. 90 or 0 degrees) frictional heating during impact and ignition probabilities tend to zero. However, as indicated by e.g. the factorial experiment mentioned above, deviations away from the optimum by 10 degrees or so are less significant in determining the overall ignition probability than hardness or the kinetic energy available.

WETTING OF THE PLATE

For most impacts envisaged within a waste silo, the interface would be wet. This has therefore been included as a parameter in some experiments. As can be seen from Table 1, the presence of water does not prevent ignition. A significant interaction was noted between the angle of impact and wetting of the target.

However, if a plate is wet for a significant period then additional factors come into play such as corrosion, which in the case of Magnox will produce a sludge containing Magnox metal and its products of corrosion. This will have the effect of reducing the probability of ignition. This phenomenon requires further study.

HYDROGEN CONCENTRATION

Hydrogen concentration (between 10 & 30% (v/v)) was not found to be significant in factorial experiments. This is not unexpected as it appears to be the initiation of the thermite/pyrophoric reaction that largely determines whether or not ignition will occur.

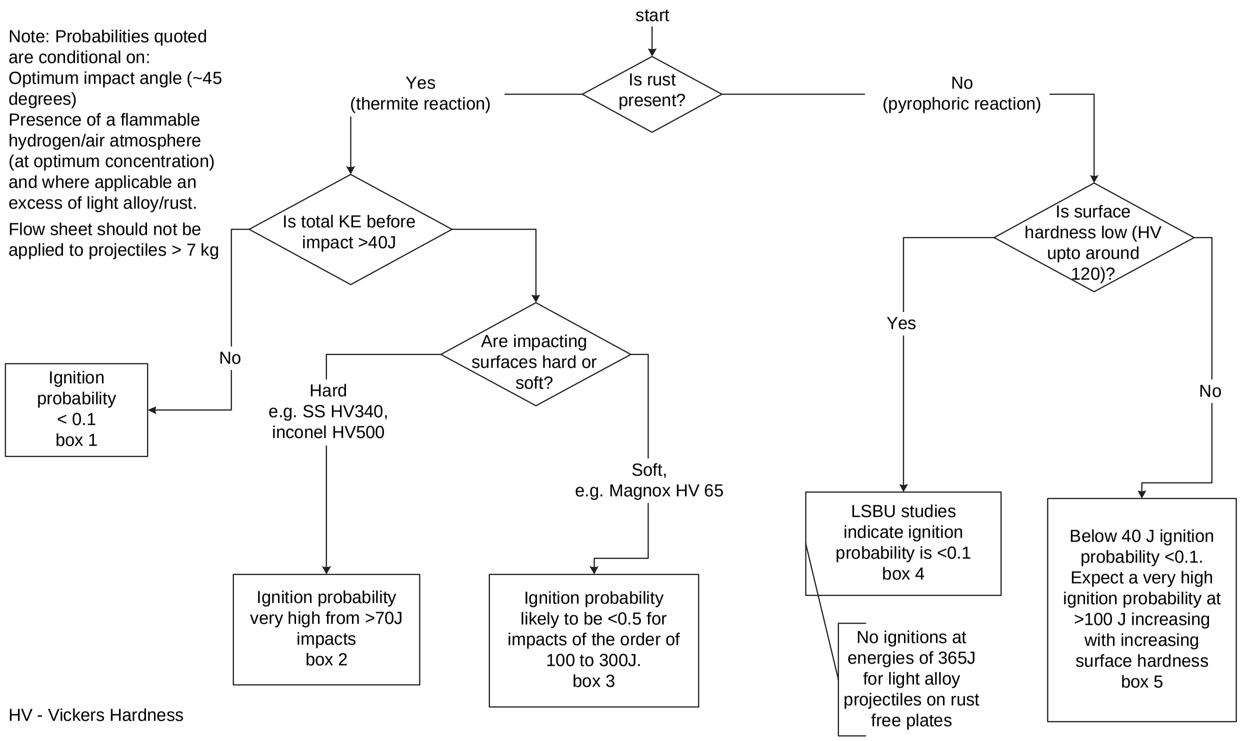
PRESENCE OF RUST

In common with studies reported in the literature [1] (primarily for methane/air mixture) it is observed (cf. runs 9&10 on Table 1) that for ignition to occur in impacts between light alloy projectiles and a steel inclined plate, rust needs to be present at the point of impact. This suggests that thermite reactions are more easily initiated during an impact of solid light alloy on steel than are pyrophoric reactions in air. Clearly it can be seen from Table 1 that, for impacts involving light alloy particulate between two hard surfaces, ignition in the absence of rust is possible and may occur at relatively low kinetic energies prior to impact.

PROBABILITY OF IGNITION

The results of the various experimental studies have been utilised to produce a simple flow sheet for the estimation of ignition probability. The flow sheet, given in Figure 3, essentially captures the results of the experimental work. The factors considered on the flow sheet are those that have been identified (i.e. available kinetic energy, hardness of materials, presence of Magnox and rust) as being most significant in determining ignition probability, other factors are assumed to be ‘worst case’. Given that many impacts will occur normal to a surface, it is obviously pessimistic to assume an impact angle of around 45 degrees. It is currently thought that this can be considered, in effect, as a reduction in the available kinetic energy (since frictional heating on impact will be reduced and the contribution to heating from deformation is small) and an adjusted value used in conjunction with the flow sheet. Further work is planned to underpin this approach.

In addition to the above approach, work is also continuing to further understand the mechanics of impact events leading to ignition. It is anticipated that such data will enable more accurate specification of conditions required for ignition in terms of, e.g. a critical frictional heat flux, which could then be directly related to a specific impact event.



All energies refer to total K.E immediately prior to impact

For pyrophoric materials other than Al or Mg alloys (e.g. zirconium) or reducible oxides other than rust threshold energies may be lower

Figure 3. Flow sheet for estimation of ignition probability for impacts involving freely falling bodies and light alloys with thermite or pyrophoric reactions

CONCLUSIONS

The relative importance of various factors affecting the probability of ignition from impacts of freely falling bodies, involving thermite or pyrophoric reaction has been assessed and utilising the probability data obtained a flow sheet for the estimation of ignition probability has been produced. Obviously there are many types of impact outside of the flowsheets applicability. Work is currently planned in a number of areas. A new high mass impact apparatus (up to 50 kg projectiles) has been designed and commissioned. Tests with the new apparatus are being undertaken to extend criteria for ignition to higher mass projectiles. In addition this apparatus is being used to investigate ignition probability from impacts between clean steel surfaces.

Ignition from impacts in which the motion of the projectile is constrained (such as a hammer blow) has also been studied. In such experiments ignition can be achieved with much lower available energy. However unlike drop weight impacts where the motion of the projectile on impact depends essentially on projectile geometry, inclination of target and coefficients of friction and restitution, the mechanics of a constrained impact are more complex. As such, laboratory experiments cannot so readily be related to a range of real impacts. Work to develop usable criteria for estimation of ignition probability from constrained impacts is ongoing.

The work reported in this paper has demonstrated that there is significant pessimism in always assuming an ignition probability of 1 resulting from mechanical impact. This offers the potential to reduce pessimism and avoidance of over complex design whilst still maintaining a robust safety case.

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