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Short Communication

REACTIONS BETWEEN HALOGENATED HYDROCARBONS AND METALS – A LITERATURE REVIEW

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It has long been recognised that under certain conditions violent reactions can occur between light metals, particularly aluminium, and halogenated hydrocarbons. As far back as 1920, Berger [1] reported that zinc can react readily with carbon tetrachloride and it is suspected that the potential hazards of such reactions were recognised much earlier than this. The increased use of both light metals and halogenated solvents has led to an increased awareness of possibly dangerous reactions, and HSE Safety Inspectors have given warnings of potential problems when chlorinated hydrocarbons come into contact with aluminium plant, particularly under pressure. Increasingly, HSE Inspectors are being asked for advice by, for example, spray equipment manufacturers and users without the benefit of an up-to-date review of potential hazards. For this reason, this review of the literature available on reactions between metals and chlorinated hydrocarbons was commissioned, in order to bring the topic into perspective. The paper mainly describes reactions between aluminium and chlorinated hydrocarbons but also considers reactions of other metals, particularly magnesium, lithium, sodium and potassium, and other halogenated hydrocarbons, including those in polymeric form such as PTFE.

Reaction mechanism

The mechanism of reaction between metals and halogenated hydrocarbons is not exactly clear but one mechanism is clearly favoured by many research workers. Here, the hydrocarbon attacks the metal to form, for example, aluminium chloride in the case of aluminium. This then acts as a catalyst for an exothermic polymerisation reaction of the hydrocarbon. Thus for aluminium and chloroform:

$$Al + CHCl_3 \rightarrow Al\text{–alkyl chlorides}$$

$$CHCl_3 \xrightarrow{AlCl_3} \text{polymer}$$

Some authors assert that water must first be present for this chain of events to occur, but reactions are cited later where water had been deliberately excluded in order to increase safety, yet a violent reaction still occurred. The influence of pressure and temperature is unclear, but since both increasing pressure and temperature will increase the reaction rate, it is reasonable to assume that a hazard at ambient conditions will be heightened at increased pressure and temperature, apart from the fact that confinement will always be present in a pressurised system.

It is not the intention here to investigate the mechanism of reactions between metals and halogenated hydrocarbons, but merely to report a fairly limited literature review which should indicate where possible hazard areas exist.

Results of review

The results of the review are given in a grid form, reading off the metal against the halocarbon. This will yield a reference number, and a very brief description of the reported reaction whence the full reference can then be found.

The following should be noted with respect to the table and references:

1. The metals are listed in the vertical axis of the grid in alphabetical order of their chemical symbols.

2. The halocarbons are listed in the horizontal axis of the grid, first in order of increasing carbon number, then increasing hydrogen number, then in alphabetical order of attached halogen(s). Thus CBr_4 precedes $CHBr_3$ which in turn precedes C_2HCl_3 and $C_3H_6Cl_2$.

3. It is not claimed that the list of reactions is exhaustive, and doubtless, other reactions have been reported which were not traced. Similarly, it must not be assumed that a blank square in the grid represents zero hazard. Such a square *may* represent zero hazard, but it may also mean that no reference has been traced or that no work has been reported on that particular combination. It would be reasonable to assume that where a reaction between, for example, aluminium and carbon tetrachloride has been reported, that a similar reaction (maybe of a different degree of intensity) would occur between aluminium and carbon tetrabromide or tetraiodide.

4. Much of the data has been obtained from Bretherick [2], where excellent resumés of the reactions are given, and also from the NFPA Manual of Hazardous Chemical Reactions [3]. Without the enormous amount of information given in these two publications, the work in collecting the information on these reactions would have been much more time consuming.

Table 1

Metal	Halogenated hydrocarbons							
	CBr_4	CCl_4	CF_4	CI_4	$CBrF_3$	CCl_2F_2	CCl_3F	$CHBr_3$	$CHClF_2$
Al		19, 32, 45, 48, 73, 74	6, 32, 49		6, 49	6, 17, 32, 49, 62	32,49		6, 32, 49
Ba		20, 22, 52					22, 52		
Be		23							
K		24, 56						24	
K/Na alloy	61	56						63	
Li	29, 53	29, 30, 31, 53, 54		29			29	29, 53	
Mg		32, 33				34			
Na		36, 37							
Pu		42							
Sm									
Ti									
U		66							
Zn		1							
Zr		47, 67							

Metal	Halogenated hydrocarbons									
	$CHCl_3$	CHI_3	CH_2Br_2	CH_2Cl_2	CH_2I_2	CH_3Br	CH_3Cl	$C_2Cl_2F_4$	$C_2Cl_3F_3$	C_2Cl_4
Al	32			11, 79		50, 81	8, 13, 18, 32, 45	6, 32, 49	4, 6, 32, 49	12, 14
Ba									21, 22, 52	22, 52
Be										
K	57		24	57	24		57			25
K/Na alloy	28, 61			61, 63			61		65	
Li	29, 53	53		29, 53	29, 53				29	29
Mg	32					81	32			
Na	36, 58			36, 41, 58			36, 58			
Pu										
Sm									43	
Ti									44	
U										
Zn				79		81				
Zr										

Results and discussion

From a purely practical point of view, the main metals of interest in this review are aluminium, magnesium and zinc. Many of the other metals cited as

Table 1 (continued)

Metal	Halogenated hydrocarbons									
	C_2Cl_6	C_2HCl_3	C_2HCl_5	$C_2H_2Cl_2$	$C_2H_2Cl_3F$	$C_2H_2Cl_4$	$C_2H_3Cl_3$	$C_2H_4Br_2$	$C_2H_4Cl_2$	C_2H_5Cl
Al	15	9, 10, 12, 51, 68, 72, 77		68	4		9, 78, 79	35		
Ba		22, 52								
Be		23								
K			24			24, 60	24	35	24	24
K/Na alloy			63				26, 63			
Li		29								
Mg		33					9	35		
Na		59		59		36, 38, 59, 60		35		
Pu										
Sm										
Ti		44								
U										
Zn	46						79			
Zr										

Metal	Halogenated hydrocarbons									
	$C_3H_6Cl_2$	C_4H_9Br	C_6H_5Br	C_6H_5Cl	$C_7H_4BrF_3$	CCl_4 + $CHCl_3$	$(C_2ClF_3)_n$ grease	$(C_2F_4)_n$ (PTFE)	"Fluoro-lube"	$C_6H_4Cl_2$ + $C_3H_6Cl_2$ + $C_2H_4Cl_2$
Al	16, 75					32, 45	7	71	70	5
Ba										
Be										
K										
K/Na alloy								27, 64		
Li										
Mg					55	32		69, 71, 80		
Na		39	39	40						
Pu										
Sm										
Ti										
U										
Zn										
Zr										

being reactive are of a rare nature, and their reactivity is not surprising. The conclusions of this review will therefore concentrate on aluminium, magnesium and zinc, the latter being included because of the implications for galvanised plant.

Reactions involving aluminium are by far the most numerous of those reported, and many of these reactions involve the metal in the form of powder, dust, flitters and turnings. It is evident that, along with temperature and pressure, the physical division of the metal increases its reactivity and any contact of finely divided aluminium (or magnesium) with a halogenated hydrocarbon should be viewed with caution. Of the cited reactions involving aluminium, half involve the metal in this finely divided form, and the same is true of all but two of the reactions involving both magnesium and zinc. There is clearly sufficient evidence to suggest that mixtures of any halogenated hydrocarbon with powdered aluminium, magnesium or zinc should be regarded as potentially dangerous. Indeed, in the intentional Grignard reaction between magnesium turnings and an alkyl halide for organic syntheses, caution has to be exercised because of its self-accelerating nature [2].

As far as the hazard associated with "bulk" metal (as opposed to powder, dust, turnings etc.) in combination with halocarbons are concerned, the references tabulated below are relevant. Other less relevant references include Ref. [10] (catalysis by aluminium chloride) and Ref. [62] (involving molten aluminium).

Ref.	Metal	Component
5	Al	Tank
6	Al	Pump impellor
7	Al	Bearing surfaces
8	Al	Autoclave
13	Al	Regulator parts
16	Al	Transfer pipe
18	Al	Baffle plates
35	Mg	General hazard data
49	Al	Fresh surfaces
50	Al	Tube
70	Al	Bolt being screwed into block
72	Al	Mould
75	Al	Tanker
78	Al	Gearwheel
79	Al and Zn	High pressure pump
81	Al, Zn, Mg	General hazard warning

Examining these references a little more closely shows that those which give rise to most concern are Refs. [5, 13, 16, 18, 75, 78, 81]. In all of the other references there is a factor which would increase reactivity due to pressure, heat or fine division of the metal. For example, in Ref. [6] a pump impellor is quoted as the reacting metal component. It is reasonable to suspect that in this case the impellor could have fouled the pump casing, giving rise to frictional heating and possible abrasion of fine particles of aluminium. Little information is available on the physical form of the aluminium in Ref. [8], although it is suspected that it may be a powder, and the general hazard warning in Ref. [35] does not specify the form of the metal.

The remaining seven references listed above do, however, quote reported incidents or give warnings of the reaction between (mostly) aluminium and certain halocarbons at apparently ambient conditions, although the common factor in all of these references would appear to be the long time involved before a reaction occurred. The shortest quoted time is "a few hours" in a transfer pipe in warm weather [16]. Work carried out in Japan following an explosion in a cargo storage area has, however, shown that temperatures as high as 85 °C can be achieved in metal drums stored in metal containers in an air temperature of about 35 °C, so it is reasonable to expect that the explosion in the pipe could have been triggered by temperature elevation if there was no flow in the pipe.

There would appear to be little evidence, therefore, that there is any hazard associated with short term exposure of bulk metals to halogenated hydrocarbons at ambient temperatures and pressures; the problem areas seem to be (i) prolonged storage (ii) pressurised systems and (iii) systems where fresh metal surfaces or metal powder may be produced.

Résumés of cited references

1 E. Berger, C.R. Acad. Sci., 170 (1920) 29. Zinc powder and carbon tetrachloride burns readily.

2 L. Bretherick, Handbook of Reactive Chemical Hazards, Butterworths, 3rd edn., 1985. Grignard reagents; self accelerating reactions plus individual entries.

3 National Fire Protection Association, Manual of Hazardous Chemical Reactions, 491M, Boston, 1975.

4 Pot. Incid. Rep., ASESB, 39 (1968) 4–11. Mixtures of aluminium powder with trichlorofluoroethane and trichlorotrifluoroethane will flash or spark on heavy impact.

5 Anon., Chem. Eng. News, 33 (1955) 942. An aluminium tank containing a mixture of *o*-dichlorobenzene, 1,2-dichloroethane and 1,2-dichloropropane exploded violently after seven days.

6 B.J. Eiseman, J. Amer. Soc. Heat. Refr. Air Cond. Eng., 5 (1963) 63. An aluminium impellor reacted exothermally with dichlorodifluoromethane. Later tests showed similar results decreasing in order of intensity with tetrafluoromethane, chlorodifluoromethane, bromotrifluoromethane, dichlorodifluoromethane, 1,2-dichlorotetrafluoroethane and 1,1,2-trichlorotrifluoroethane.

7 J.R. Laccaube, Fluorolube–aluminium detonation point, General Dynamics Report 7E.1500, 1958. Aluminium bearing surfaces under load react explosively with polychlorotrifluoroethylene greases or oils.

8 V.J. Atwell, Chem. Eng. News, 32 (1954) 1824. During the methylation of methylpropane with methylchloride in the presence of aluminium and aluminium chloride, a detonation occurred.

9 L. Bretherick, Handbook of Reactive Chemical Hazards, Butterworths, 3rd edn., 1985, p. 26. (i) Violent decomposition may occur when trichloroethane comes into contact with aluminium or magnesium; (ii) Aluminium-dusty overalls were cleaned with trichloroethylene. A violent ignition occurred during drying.

10 Anon., R. Soc. Prev. Accid. Ind. Accid. Prev. Bull., 21 (1953) 60. Polymerisation of trichloroethylene is catalysed by aluminium chloride and the reaction is self-accelerating.

11 R.D. Coffee, Loss Prev., 5 (1971) 113. Aluminium powder undergoes an uncontrollable exothermic reaction with dichloromethane above 95 °C and under pressure.

12 R.F. Schwab, Loss Prev., 5 (1971) 113. Several violent reactions have occurred between aluminium and trichloroethylene or tetrachloroethylene in vapour degreasers.

13 R. Corley, Loss Prev., 5 (1971) 114. Methyl chloride diffused in a regulator and reacted with aluminium components to form alkylaluminium compounds. These ignited when the regulator was dismantled.

14 H.J. Heinrich, Arbeitsschutz, 6 (1966) 156–157. The presence of water and aluminium chloride in an aluminium powder–tetrachloroethylene mixture lowered the initiation temperatue to such an extent that an explosion occurred due to overheating of residues on heating coils in an aluminium degreasing plant.

15 A. Lamoroux and J. Meyer, Mem. Poudres, 39 (1957) 435-445. The reaction of aluminium powder with hexachloroethane in alcohol is not initially violent, but may become so.

16 A.C. Hamstead, G.B. Elder and J.C. Canterbury, Corrosion, 14 (1958) 189t–190t. An aluminium transfer pipe carrying 1,2-dichloropropane failed due to corrosion in warm weather after only a few hours.

17 I. Hartmann, Ind. Eng. Chem., 40 (1948) 756. In dichlorodifluoromethane vapour, aluminium dust ignited at 580 °C, and suspensions of the dust in the vapour on sparking gave strong explosions.

18 Anon., MCA Case History, No. 2160, Manufacturing Chemists Association, Washington. The interaction of methyl chloride and aluminium baffle plates in a tanker produced trimethylaluminium which caused a fire.

19 G.W. Wendon, Ind. Eng. Chem., Prod. Res. Dev., 16 (1977) 112. The production of aluminium chloride by ball-milling aluminium in carbon tetrachloride is potentially hazardous.

20 Anon., Serious Accident Series, Vol. 23 and Suppl., Washington, USAEC, 1952. A violent reaction occurred when cleaning lump barium under carbon tetrachloride.

21 Anon., Ind. Res., 9 (1968) 15. Finely divided barium, slurried with trichlorotrifluoroethane, exploded due to frictional initiation.

22 Pot. Incid. Rep., ASESB, 39 (1968) 4–11. Granular barium is susceptible to detonation when in contact with trichlorofluoromethane, carbon tetrachloride, 1,1,2-trichlorotrifluoroethane, tetrachloroethylene or trichloroethylene.

23 Pot. Incid. Rep., ASESB, 39 (1968) 4–11. Powdered beryllium mixed with carbon tetrachloride or trichloroethylene will flash on heavy impact.

24 H. Staudinger, Z. Angew. Chem., 35 (1922) 657; H. Staudinger, Z. Elektrochem., 31 (1925) 549; F. Lenze and L. Metz, Chem.-Ztg., 56 (1932) 921. Mixtures of potassium with a range of halocarbons are shock sensitive. Mono-, di-, tri- and pentachloroethane, bromoform, dibromo- and diiodomethane have been studied. Mixtures with carbon tetrachloride are also shock sensitive and mixtures with tetra- or pentachloroethane may explode spontaneously.

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26 Lawrence Radiation Lab. Inform. Exch. Bull., 1 (1961) 2. An explosion occurred in a valve following contact between potassium/sodium alloy and 1,1,1-trichloroethane.
27 H. Staudinger, Z. Elektrochem., 31 (1925) 550. A Teflon-coated stirrer used to stir potassium/sodium alloy under propane ignited when the speed was increased.
28 E. Schmidt et al., Chem. Abs., 83 (1975) 163034s. Potassium/sodium alloy may explode on contact with chloroform.
29 H. Staudinger, Z. Elektrochem., 31 (1925) 549; E. Mueller, Pot. Incid. Rep., ASESB, 39 (1968) 4-11. Lithium shavings will explode, sometimes violently, with bromoform, carbon tetrabromide, carbon tetrachloride, carbon tetraiodide, chloroform, dichloromethane, diiodomethane, trichlorofluoromethane, tetrachloroethylene, trichloroethylene or 1,1,2-trichlorotrifluoroethane.
30 L.R. Pittwell, J.R. Inst. Chem., 80 (1959) 552. Shearing samples of lithium immersed in carbon tetrachloride caused an explosion.
31 BDH Catalogue Safety Note 969DD/14.0/0773, British Drug Houses, 1973. Lithium which had been washed in carbon tetrachloride exploded when cut with a knife.
32 C.C. Clogston, Bull. Res. Underwriters Lab., 34 (1945) 5. Powdered magnesium or aluminium may explode on contact with methyl chloride, chloroform or carbon tetrachloride, or mixtures of these.
33 Pot. Incid. Rep., ASESB, 39 (1968) 4-11. Powdered magnesium mixed with carbon tetrachloride or trichloroethylene will flash on heavy impact.
34 I. Hartmann, Ind. Eng. Chem., 40 (1948) 756. Magnesium dust ignited at 400°C in dichlorodifluoromethane and suspensions exploded violently on sparking.
35 Hazardous Chemical Data, NFPA, Boston, 1973, 140. Interaction of magnesium, aluminium, sodium or potassium with 1,2-dibromoethane may be violent.
36 H. Staudinger, Z. Elektrochem., 31 (1925) 549; F. Lenze et al., Chem.-Ztg., 56 (1932) 921. Explosions initiated by shock or impact may occur with mixtures of sodium and carbon tetrachloride, chloroform, dichloromethane, methyl chloride or tetrachloroethane.
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38 Manufacturing Chemists' Association Safety Data Sheet, MCA SD-34, Washington, DC, 1949. Similar hazard to (37) above with sodium and tetrachloroethane.
39 R.R. Read, L.S. Foster, A. Russell and V.L. Simril, Org. Synth., Coll. Vol. 3 (1955) 158. The reaction of bromobenzene, 1-bromobutane and sodium in ether becomes violent above 30°C.
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44 Pot. Incid. Rep., ASESB, 39 (1968) 4-11. Mixtures of powdered titanium and trichloroethylene or 1,1,2-trichlorotrifluoroethane flash or spark under heavy impact.
45 Anon., Angew. Chem., 62 (1950) 584. Heating aluminium powder with carbon tetrachloride, chloromethane or carbon tetrachloride/chloroform mixtures in closed systems to 152°C may cause an explosion, particularly if traces of aluminium chloride are present.

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49 B.J. Eiseman, J. Amer. Soc. Htg. Refr. Air Cond. Eng., 5 (1963) 63; C.C. Clogston, Bull. Res. Underwriters Lab., 34 (1945) 5; Anon., Chem. Eng. News, 39 (27) (1961) 44, Molten metal; Anon., Chem. Eng. News, 39(32) (1961) 4. Vigorous reactions have been observed between fresh aluminium surfaces and dichlorodifluoromethane, chlorodifluoromethane, trichlorotrifluoroethane, trichlorofluoromethane, dichlorotetrafluoroethane, bromotrifluoromethane and tetrafluoromethane.
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57 H. Staudinger, Z. Elektrochem., 31 (1925) 549. Potassium reacts explosively with chloroform, dichloromethane or methyl chloride.
58 H. Staudinger, Z. Elektrochem., 31 (1925) 549. Sodium plus chloroform, dichloromethane or methyl chloride, forms a shock-sensitive system.
59 NAVWEPS OP 3237 (1964) 28, Bureau of Naval Weapons, Navy Dept., Washington, DC; Fire and Accident Prevention, 42, 1956, USAEC Information Bulletin Series, Washington, DC. The addition of sodium to 1,2-dichloroethylene, trichloroethylene or tetrachloroethane may form products which are spontaneously flammable in air.
60 Chemical Safety Data Sheet SD-34, Manufacturing Chemists Association, 1949. Tetrachloroethane may explode with potassium or sodium.
61 H. Staudinger, Z. Elektrochem., 31 (1925) 549. Sodium/potassium alloy is impact sensitive with carbon tetrachloride, chloroform, dichloromethane or methyl chloride.
62 Anon., Chem. Eng. News, 39(27) (1961) 44. Molten aluminium dropped into liquid dichlorodifluoromethane burned incandescently below the surface. The reaction with trichlorofluoromethane is much slower and burning stops after a few seconds.

63 J.W. Mellor, A Comprehensive Treatise on Inorganic and Theoretical Chemistry, Vol. II, Suppl. 2, Longmans, 1961, p. 83. Sodium/potassium alloy can explode with bromoform, dichloromethane, tetrachloroethane or pentachloroethane, and such mixtures are impact sensitive.

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65 F.O. Pancner, 1966 (Communication; listed in NFPA 491M, 1975). When two drops of trichlorotrifluoroethane were added to sodium/potassium alloy, a violent explosion took place.

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67 W.W. Allison, 1969 (Communication; listed in NFPA 491 M, 1975). An explosion occurred when zirconium sponge was placed in a beaker of carbon tetrachloride.

68 R.C. Smart, Ind. Fire Expl. Hazards, 2 (1947) 26. Finely divided aluminium reacts violently in contact with hot dichloroethylene or hot trichloroethylene.

69 H. Ellern, Military and Civil Pyrotechnics, Chemical Publishing Company, New York, 1968, p. 298. Burning of a mixture of magnesium and PTFE is initiated at 493 °C.

70 ASESB, Report 112, Washington, 1967. An explosion occurred when an aluminium bolt lubricated with Fluorolube was being screwed into an aluminium block.

71 M.J. Day and B.C. Turner, HSE Unpublished Report No. IR/L/IN/HM/81/1, 1981. In the course of an incident investigation, mixtures of finely divided aluminium or magnesium with PTFE were made to react violently on heating in a test tube.

72 G.T. Harding, HSE Unpublished Report, Ref. M/FCG/896/84(12), 1984. An exothermic reaction occurred between a trichloroethylene-based mould release agent and an aluminium liner. Choking acrid fumes of hydrogen chloride were produced.

73 Anon., Chem. Age, 63 (1950) 155. A mixture of carbon tetrachloride and aluminium powder exploded during ball-milling.

74 Anon., Chem. Eng. News, 32 (1954) 258. A mixture of carbon tetrachloride and aluminium powder is impact sensitive.

75 I. Chem. E. Loss Prev. Bull. 054, 1983. Two thousand gallons of propylene dichloride (US designation of dichloropropane) spilled from a parked aluminium truck after reacting with the tank and corroding several square feet.

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77 H. Heiss and F. Heck, Arbeitsschutz, 13 (1963) 266a–266f. The use of trichloroethylene for degreasing aluminium strip presents no hazards provided a well stabilised brand of solvent is used and that it is renewed as soon as it is neutralised (pH 7).

78 Anon., Univ. Safety Assoc. Newsletter, 16 (1982) 5. An attempt to clean a motor assembly containing an aluminium alloy gearwheel by soaking overnight in 1,1,1-trichloroethane led to a gross degradation of the assembly.

79 Extracts from DeVilbiss Company Safety Warning Release Letter, No. 4632, October 1981. Two explosions were caused by the reaction between aluminium and halogenated hydrocarbons in high pressure pumps ... Two widely used solvents are 1,1,1-trichloroethane and methylene chloride (US designation of dichloromethane) ... Similar reactions can occur with zinc, which makes galvanized steel suspect.

80 T.T. Griffiths, J. Robertson, P.G. Hall and R.T. Williams, Int. Jahrestag Fraunhofer Institut fur Treib und Explosivestoffe, 1985. Magnesium powder reacts exothermically with PTFE powder even in an inert atmosphere.

81 MCA Case History, No. 746, 1966. Washington. Metallic components of zinc, aluminium and magnesium are unsuitable for service with bromomethane due to the formation of pyrophoric Grignard-type compounds. An explosion occurred of a bromomethane–air mixture by pyrophoric methylaluminium bromides caused by corrosion of an aluminium component.