



# Stability of acetylene–propane–butane and acetylene–hydrogen gas mixtures subjected to shock wave action



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## HIGHLIGHTS

- Experimental setup with a shock wave loading of acetylene was described.
- The concentrations of inhibitors are given under which the decomposition occurs.
- Maps of acetylene decomposition on the pressure and concentration are presented.
- Estimation of the temperature behind the reflected shock wave was made.

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## ABSTRACT

Inhibition of exothermal spontaneous decomposition of acetylene by dilution with a gaseous propane–butane mixture or with hydrogen, which themselves serve as fuels, was carried out experimentally. Mixtures of acetylene with the inhibitors, located in a cylindrical shock tube, were heated by a reflected shock wave. The source of the shock wave was provided by a detonation wave initiated in a stoichiometric acetylene–oxygen mixture and then converted to a shock wave in the acetylene–inhibitor mixture. Minimal limits of bulk concentrations of inhibitors were obtained over a range of initial pressure 0.1–0.25 MPa at which no spontaneous decomposition of acetylene occurred.

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## 1. Introduction

One of the promising gas fuels in devices that utilize both deflagration combustion (welding, cutting of metal) and detonation combustion (sputtering, disintegration) is a mixture of acetylene with air (or with oxygen), which is characterized by high caloric power, high parameters of detonation products, etc. However, the disadvantage of pure acetylene is its ability to decompose to form carbon and methane or hydrogen with energy release and possible detonation. This concern limits the use of acetylene from the safety standpoint [1].

The process of pyrolysis is directly related to the process of explosive self-decomposition. In [2] the process of pyrolysis was investigated in a jet device, in [3] it was investigated in a shock tube. A great number of works have been devoted to hydrocarbon pyrolysis associated with soot formation [4–6]. A mechanism for

acetylene pyrolysis with soot particle formation from polyynes was presented in [7,8]. The role of the polyynes and *H*-abstraction– $C_2H_2$ -addition (HACA) pathways was shown in [9]. In [7,10] the role of benzol molecules and aromatic molecules in the process of soot particle growth was shown.

There are a great number of rather complicated kinetic schemes that describe acetylene decomposition [3,11,12]. In [13] the induction period of acetylene pyrolysis was studied. Colket et al. suggested that the reaction was initiated by acetone, an impurity in acetylene [14].

The process of explosive acetylene decomposition can be greatly affected by certain diluents. The addition to acetylene of a component that interferes with the process of decomposition enables one to solve this problem [15].

The pyrolysis of binary acetylene–benzol mixtures was investigated in [10]. In [16] soot formation from a mixture of acetylene and benzol was investigated in a jet device, the period of soot formation induction having been reduced from 3–9 ms down to 0.6–1.0 ms. The pyrolysis of acetylene, ethanol and acetylene–ethanol mixtures in the 975–1475 K temperature range has been studied in [17]. The cited works were carried out under flow

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## Nomenclature

4(mol%)	molar concentration	$T_{est}$	estimated temperature
33 wt%	weight fraction	$C$	limit concentration of inhibitor (molar)
$P_0$	initial pressure of mixture	$M$	mach number
$P_{exp}$	measured shock wave pressure	$\gamma$	gas constant
$P_{est}$	estimated pressure		

reactor conditions but not shock tube conditions. The characteristic times are different for a shock tube and for a flow reactor.

Stability studies of MAPP<sup>®</sup> gas, containing methylacetylene stabilized with alkane and alkene hydrocarbons [18], have shown it to be stable up to 600 °C and 7.5 MPa, so long as there is no depletion of the added stabilizers [19]. The US Bureau of Mines [20] examined the explosive properties of acetylene mixtures containing  $C_3$  and  $C_4$  alkanes, natural gas, carbon dioxide, nitrogen, helium, and hydrogen at pressures up to 0.78 MPa. The study showed that, up to 0.78 MPa, explosive hazards could be avoided by the addition of 20–40(mol%) hydrocarbons, 41(mol%)  $CO_2$ , 55(mol%)  $N_2$ , 58(mol%)  $He$  or 61(mol%)  $H_2$ . It was shown in [21] that additives of 0.5–10(mol%) nitric oxide, hydrogen halides, or vinyl bromide can be used to stabilize acetylene. Holtappels and Schonbucher [22] tested the explosive limits of mixtures of  $C_2H_2/N_2$ ,  $C_2H_2/CO_2$ ,  $C_2H_2/H_2$ ,  $C_2H_2/NH_3$  and  $C_2H_2/C_2H_4$  with an ignition source at various pressures and temperatures up to 20 MPa and 150 °C, respectively. In [23] acetylene was injected into the intake port as a secondary fuel and diesel was injected directly into the cylinder of a DI diesel. It was concluded that without loss in thermal efficiency, safe operation of acetylene is possible in the timed port injection technique.

The stability of binary gas mixtures of acetylene and methane containing up to 10(mol%) acetylene, and a ternary gas mixture of 4(mol%) acetylene, 20(mol%) hydrogen, and 76(mol%) methane at pressures up to 3600 psig and temperatures up to 200 °C was investigated in [24].

However, the nature of inhibition may not only be thermodynamic, but also kinetic. It is generally agreed that the role of chain inhibition at higher pressures can be neglected in comparison with heat inhibition with inert freons. Thereby, there have been few articles concerned with chain inhibition of chemical reactions at higher pressures [25]. However, it is the range of pressures above 0.1 MPa that is of the greatest interest and importance, not only from the scientific point of view (chemicals processes in flow and behind shock waves) but from the practical point of view (storing and utilization of fuel gases). The results of thermodynamic analysis showed that the inhibition of explosive decomposition of acetylene at a pressure of 1 atm is not only of a thermal nature, but also kinetic in nature [26]. In the latter work it was experimentally found that the minimal concentrations of inhibitors preventing the spontaneous decomposition of acetylene behind shock waves were 7% for propane–butane and 12% for hydrogen.

The main products of acetylene thermal decomposition are soot particles and methane (and/or hydrogen). A key role of acetylene in the soot formation process was revealed. The most widely used HACA model (*H*-abstraction– $C_2H_2$ -addition) of soot formation is based on the assumption of a key role of acetylene molecules in the whole soot formation process. Usually, to study details of the kinetic mechanism of soot formation and acetylene pyrolysis, the experiments are conducted with the use of highly diluted mixtures of acetylene with a buffer gas. In this case, the temperature rise due to acetylene pyrolysis is the most important factor, which influences the kinetics of the whole process.

This paper is devoted to experimental determinations of the lower concentration limit of an inhibitor such as a liquefied propane–butane mixture (household gas) or hydrogen, above which

no spontaneous decomposition of acetylene occurs. Mixtures of pure acetylene with additives (hydrogen or propane/butane) without any inert dilution were studied.

## 2. Experimental set and experimental technique

Inhibition of acetylene decomposition has been investigated behind a shock wave. For this purpose a shock tube was used, comprised of two sections: a detonation combustion section and a measuring section with an internal diameter of 20 mm (Fig. 1). A ball cock was used as the connecting element between the two chambers of the tube. The originality of using the ball cock is that it avoids laborious substitution of the ruptured diaphragm between experiments. This gives us the possibility to increase the number of experiments several fold.

The detonation section was filled with a stoichiometric acetylene–oxygen mixture up to a necessary initial pressure, and the measuring section with an acetylene–inhibitor mixture up to the same pressure as in the closed ball cock. Once the sections had been filled, the ball cock was opened and the acetylene–oxygen mixture was ignited by a spark. The electrical energy of discharge was 2 Joules. Fig. 1 shows the trajectory of a detonation wave in the detonable acetylene–oxygen mixture, which passed through the interface between the sections from left to right and then changed to a shock wave in the test acetylene–inhibitor mixture.

The decomposition and the inhibition of acetylene were investigated behind the reflected shock wave. The mixture behind the reflected shock wave was immobile; therefore the observation and diagnostics of the combustion/decomposition processes were essentially simplified. The reflection of the shock wave and the trajectory of a decomposition wave are shown in Fig. 1. The decomposition of acetylene started 30–250  $\mu$ s after the reflection.

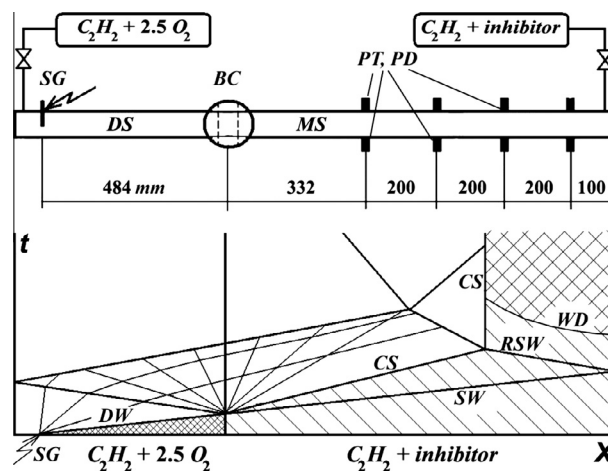


Fig. 1. A schematic diagram of an experimental setup and X–t diagram of compression of mixtures. DS – detonation section; MS – measuring section; BC – ball cock; SG – spark gap; PT – piezoelectric pressure transducers; PD – photodiodes. DW – detonation wave, SW – shock wave; CS – contact surface; RSW – reflected shock wave; WD – wave of decomposition.  $\tau$  – delay of decomposition.

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