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TECHNICAL MEMORANDUMS

NATIONAL ADVISORY COMMITTEE FOR AERONAUTICS

No. 547

CONTRIBUTION TO THE STUDY OF NORMAL BURNING
IN GASEOUS CARBURETED MIXTURES

By M. R. Duchêne

PART I

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By M. R. Duchêne.

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PART I.

Chapter I.

Historical.

1. Normal Burning.— The study of the rapid propagation of a region of combustion in explosive mixtures has been made the object of numerous investigations having in view a greater perfection in technique of the explosion engine.

If a mixture of a combustible gas and air contained in a long tube be ignited, the flame at first proceeds along the tube slowly and at a regular rate. After being propagated in this manner, however, for a certain distance, it loses its uniform motion and proceeds with accelerated speed and thus may reach a high velocity.

If, in place of the gas confined in the tube, the explosive mixture is made to flow through the tube and is ignited at the orifice, the flame may progress within the tube and against the

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current more slowly than in the case first mentioned. For a certain flow velocity the flame will remain stationary. The rate of propagation of the flame will then be equal and opposite to the rate of gas flow in the tube.

It is convenient to designate the slow propagation of the combustion region by the term "normal burning" (deflagration). Further on its characteristics will be more precisely described.

2. Historical.— In 1816 Davy (Royal Soc. London, Nov. 9, 1815. Ann. de Ch. et de Phys. (1816) 1, p.136, (1817)) observed that the most explosive mixture of fire-damp and air, the flame took less than a second to traverse a tube a foot long. Noticing that the combustion was not propagated if the tube was of small diameter, the idea occurred to him that resulted in the invention of the miners' safety lamp where the small openings of the metallic mesh function the same as short tubes of small diameter.

In 1866 Bunsen (Pogg. Ann. 131, 161 (1866)) measured the rate of propagation of the flame in a mixture of hydrogen and oxygen and found 30 meters per second as a result. He made this determination by balancing the flame area against the flow of the explosive gases whose rate was known.

Schoesing and Mondesir made marks at stated distances along a glass tube filled with a mixture of carbon monoxide and air. The rate of propagation was so slow in the combination of these

gases used that it could be directly measured. These investigators called attention to the importance of agitating the mixture during propagation.

Mallard (Ann. des Mines 7 series, VII, 135) repeated the method of Bunsen. To a quantity of air he added increasing quantities of the combustible gas and found that the velocity of flame propagation in the resulting mixtures increased with increasing amounts of the combustible added till a maximum was reached; after this a decrease in the rate resulted when more of the combustible gas was added. He also found that the maximum velocity did not correspond, in the case of air and fuel to the chemical proportions for complete combustion.

By plotting his results as indicated in Figure 1, and prolonging the two sides of his figure until they meet the horizontal axis representing no velocity, this author thought thereby to determine the inferior and superior limits of inflammability of the explosive mixture considered. Le Chatelier has shown that this conception is incorrect. (A.M. 8th series XIX, 338, (1891).)

More recently Richard Wheeler (Chem. Soc. Trans. (1914) 105, (2505)) has shown that the diagram does not consist of two intersecting right lines, but is a continuous curve on which the two limits of inflammability lie parallel to the horizontal axis of the figure. See Figure 1 (Michelson, Ann. der Phys. u. Chem. 37, 1, (1889), first showed that the relation between mix-

ture ratio and flame velocity was expressed by a continuous curve.)

Mallard and Le Chatelier much improved the experimental technique employed in investigations of gaseous explosions. In order to indicate the time at which the flame passed a given point in the tube a number of methods were tried out - electrical, pneumatic, and photographic. Of these the photographic has proved the most efficient and is in general the most used.

The Photographic Method

The propagation of flame in the explosive mixture takes place in a tube placed horizontally. The light emitted by the moving flame reacts on a photographic plate or film moving at known rate perpendicular to the tube. In this way the movement of the flame front impresses itself on the photographic plate as a curve whose tangent at any point reveals the velocity of the flame movement in space at the instant corresponding to the point on the flame trace. The time function is recorded on the photographic plate synchronously with the explosive reaction.

Mallard and Le Chatelier secured these impressions on a photographic plate that was allowed to fall vertically between guides. They also used strips of sensitized paper attached to a revolving drum.

In the first case the velocity was calculated from the height through which the plate fell. In the second case it was

obtained from the record of a tuning fork of period 1/100 second provided with shutters that intercepted the light from a burning magnesium ribbon as it fell on the photographic plate.

These investigators found the light from the explosive reaction $\text{CS}_2 + \text{O}_2 \longrightarrow$ and $\text{CS}_2 + \text{NO}_2$ of excellent photographic quality. Many of their experiments were carried out with these gases.

Since the results obtained by these investigators are so fundamental, we give below a brief resumé of them.

The flame trace these investigators secured by this method is seen to consist of three rather well-defined parts:

- 1st. A propagation at constant velocity following the ignition. Region marked OA, Figure 2.
- 2d. A propagation at very variable velocity corresponding to a vibratory state of the explosive gases. Region AB.
- 3d. A propagation at a very rapid and constant rate. Region BE.

This third mode of propagation was named by its discoverers, Berthelot and Vieille, the "explosive wave." It was discovered nearly simultaneously by the investigators Mallard and Le Chatelier.

The velocity of the explosive wave is found to be a physical magnitude characteristic of a given explosive mixture.

The form of container, its material, the nature of its walls,

seem to be without influence on this characteristic magnitude; but initial temperature and pressure seem to have some slight influence on its value.

The velocity of propagation of the explosive wave is great. It is of an order of many thousand meters a second.

The mode of propagation other than that by the explosive wave will be termed normal burning (deflagration). In general, if the flame is started near the opening of a tube open at both ends, for example, it propagates itself at first at uniform velocity; but the vibratory motion that establishes itself in the gases somewhat later give to the flame a vibratory motion also. This initial uniform motion itself depends upon various factors other than the composition of the gases, as for example, pressure and temperature as may be seen from the following table taken from the paper by Mallard and Le Chatelier (Ann. des Mines IV, 296).

Gaseous mixture	Tube dimensions		Distance at uniform speed	Velocity
	length	diam.		
	m	mm	m	m
CS ₂ + 3NO ₂	3	30		1.25
	2	20		1.34
	1	20		1.30
		30	1	
		20	0.80	
		10	0.50	
		4		0.00
		10		0.75
		20		1.28
		30		1.35

The position in the tube of the point of ignition has considerable influence on the form taken by the flame trace. While ignition at the open extremity of a tube gives a trace like that shown at Figure 2, starting with a right line, the trace produced when the ignition point is placed at the closed end of the tube is not straight but is parabolic in form. This modification in the initial form of the flame trace is explained by the fact that to the velocity of propagation is added the velocity with which the gases are expelled from the mouth of the tube. The heated gases displace the gases ahead of them and the flame trace partakes of the resultant motion.

Dixon (J. Chem. Soc. XCVII, pt. 1, 661 (1910); also CXXIII 1025 (1923)) has repeated and extended the studies of the earlier investigators. He has employed photographic methods. He made use of a photographic film placed on a cylinder one meter in circumference. This cylinder can be rotated from 30 to 100 revolutions per second.

This author has studied in particular the propagation of flame in explosive gaseous mixtures where ignition was induced by adiabatic compression. For these experiments he enclosed the mixture in a glass tube sealed at one end, the other end closed by a movable piston (See Fig. 3).

This device is placed horizontally before the rotating drum that gives the film a motion perpendicular to the length of the tube. The piston is forced rapidly into the tube containing

the gases by means of a ram falling against the end of the piston rod. The heat generated by the adiabatic compression ignites the gases and the subsequent movement of the flame is recorded photographically on the film.

By this device Dixon secured interesting records as to the origin of the flame in the mass of the mixture; as to the ratio of volumetric compression necessary to induce ignition; as to the velocity of flame propagation and as to the inception of the explosive wave.

In the case of ignition by adiabatic compression one pictures a great number of points within the explosive mixture that are brought simultaneously to ignition temperature; as a consequence one would think ignition should occur throughout an extended mass of the explosive gases. On the contrary, ignition is observed to take place at a point - owing possibly to some local differences in composition. On some of his photographic records there appear rectilinear flame traces, and nearly perpendicular to the edges of the film there is the flame trace of the explosive wave.

Dixon has made observations on a large number of explosive gases and even on explosive mixtures of vapors maintained at temperatures of from 50°C to 100°C . He has also made use of the ignition spark at the end of compression.

Dixon concludes that the form and dimensions of the explosion chamber, as well as the position of the ignition have marked

influence on the course of flame propagation.

Recently Laffitte (Ann. de Physique, Xth series IV, Nov. Dec. 1925) in the course of a new study of the explosive wave has obtained some excellent photographs of its propagation in gaseous mixtures. The tube in which the experiments were carried out was closed at both ends. There is exhibited very plainly in these photographs the inception of the explosive wave following a course of normal burning at variable speed. These figures do not show the preliminary oscillatory stage as seen in the figures secured by Le Chatelier, who used an open tube.

The mathematical theory of the propagation of the explosive wave has been treated by Chapman, (Phil. Mag. 47, 90 (1899)) Jouget, (J. Math. 1, 347 (1905), 2, 5, (1906)). Crussard, (Bull. Soc. Ind. Min. VI, (1907)) (and by Becker, Z. Physik. 8 321 (1923)).

It will not be out of place to refer here to the principal results obtained from these theoretical studies.

An initial adiabatic compression is propagated in an inert gas by successive waves much the same as a shock is propagated along a train of cars. Each compressional wave formed by the expansion of the wave preceding suffers an elevation of temperature; then it quickly expands. This is the mechanism of impact waves that has been studied by Riemann (Gott. Ges. d. Wiss. 8 (1860)) and by Hugoniot (J. math. pure et appl. 4^e series, III, 477 (1887)).

3. Detonation.-- If, instead of occurring in an inert gas, the compressional impact wave is set up in a gaseous explosive mixture, it may easily be conceived that the temperature within an intense impact wave may rise above the temperature of ignition of the explosive gaseous mixture. Combustion takes place within the wave and the additional heat energy thus quickly liberated produces a like effect continuously in the unburned gases ahead of the expanding wave.

It is admitted that in the case of the explosive wave the propagation of the impact wave and the region of chemical reaction are synchronous. The unburned gases ahead of "the impact and combustion wave" (the expression is due to Jouguet) do not suffer any change or disturbance as a result of the approaching wave (this is because the velocity of propagation of the wave is greater than that of sound in the gases). It is this fact that accounts for the constant velocity of the explosive wave.

This hypothesis has been verified, a posteriori, by the remarkable agreement found to exist between calculated and observed values (See N.A.C.A. Technical Memorandum No. 506: Impact Waves and Detonation, Part II, p.17, by R. Becker.)

4. Normal Burning.-- If the transmission of a compressional wave from the preceding one is insufficient to produce ignition, the propagation of the area of reaction takes place by heat conduction. The resulting process is of an entirely different order from that of detonation.

The impact wave of compression in the case of the explosive wave propagates itself by heating the unburned gases by adiabatic compression. In the case of normal burning the gases are brought much more slowly to ignition temperature by heat conduction.

In this case the flame traverses gases that have already been disturbed by the passage of sound and of weak compressional waves. It is conceivable that this disturbance may have an important influence on the rate of flame propagation and further, since the rate of propagation is much slower than in detonation the loss of heat from the walls of the container and the nature of their surface may also have an important bearing on the reaction process.

It follows from these considerations that a study of the processes of normal burning is much more complicated than the subject of detonation. When the reaction by normal burning runs its course within a rigid container the problem indeed becomes very complicated.

For these reasons a tentative theory of the process of normal burning has been applied only to the case of the propagation of the reaction in long tubes. Jouguet has offered the following formula for the rate of propagation in this case:

$$u_0 = \frac{T_0}{\sqrt{p_0}} \sqrt{\frac{nRkA}{C}} \sqrt{\frac{\theta - \tau}{\tau - T_0}}$$

where T_0 = initial temperature of the mixture
 p_0 = initial pressure
 R = gas constant given by $p\sigma = nRT$
 k = coefficient of heat conductivity
 A = coefficient of reaction velocity
 C = sp. ht. at constant pressure
 θ = temperature of combustion
 τ = temperature of ignition

This formula is in fact only an approximation and is useful only as showing the magnitudes upon which u_0 may depend.

Its application is impossible for the reason that it is not known how A (the velocity coefficient) varies. Among other things it offers an explanation why the maximum velocity of propagation in a mixture of hydrogen and oxygen requires an excess of hydrogen above that indicated by the chemical formula (50% instead of 29%). It is known that the conductivity of hydrogen is six times that of air so that it should be possible with mixtures containing more than 1/3 their volume of hydrogen, that the gain in conductivity will have the effect of exceeding the diminution of temperature of combustion.

For the case of methane and air the case should be even more marked than that for hydrogen, for

$$k \text{ (methane)} = 6.47 \times 10^{-5}$$

while $k \text{ (air)} = 5.22 \times 10^{-5}$

We have seen in fact that maximum velocity is obtained for a 12% mixture; the chemical formula corresponds to 9.4%.

Chapter II.

Object of the Study and Experimental Equipment

5. Object of Study.-- In the present study it is proposed to provide an equipment permitting the study of the propagation of the region of reaction in mixtures of air and carbureted gases enclosed within a cylinder. Ignition is produced at the end of compression by an electric spark. Such experimental conditions approach those imposed on the reaction in the cylinders of gas engines. With this apparatus it is proposed to determine:

- 1st. The influence of the richness of the explosive mixture on the rate of flame propagation, the hydrocarbons employed being benzene C_6H_6 , cyclohexane C_6H_{12} , cyclohexene C_6H_{10} , and hexane C_6H_{14} .
- 2d. The influence of the degree of volumetric compression on one of the hydrocarbons, C_6H_{12} .
- 3d. The influence of the variation of initial temperature of the mixture before compression.
- 4th. The influence of tetraethyl-lead on the propagation -- notably on the formation of the explosive wave.

6. The Apparatus.-- The apparatus made use of in these studies was suggested by that developed by Dixon. It consists of

the following parts (Fig. 4):

- 1st. A compression cylinder and explosion chamber which can be maintained at a given initial temperature.
- 2d. A carburetor.
- 3d. A device for producing quick compression.
- 4th. An ignition system.
- 5th. Photographic apparatus.

7. The Compression Cylinder and Explosion Chamber.- We have sought to avoid the frailty of Dixon's apparatus by producing the compression in a steel cylinder of 30 mm bore; the explosion chamber only being of glass (See Figs. 5, 6).

This arrangement permits the use of a steel piston provided with cast segments. Its freedom from leakage is very satisfactory. A washer placed between the segments may be slightly oiled from time to time, thus insuring ease of motion.

The range of the piston is 262 mm; the piston rod is guided in its course by a sleeve K, to be seen at the extremity of the cylinder opposite the glass tube. At the other extremity is a ring B' against which also rests the open end of the glass tube. This tube is made of pyrex glass 3 mm thick. Two different tubes were employed in the course of these studies; they are shown at Figure 9. The tube is clamped between the steel cylinder and the block B.

Since the pressure securing the tube should be as light as

possible to avoid fracturing, packing is placed about the tube end to insure against leakage (Fig. 7).

The closed end of the tube carries tungsten electrode for igniting the gases and butts against the support B provided with a leather cushion.

The ring B' carries also a stopcock r, communicating with the explosion chamber by an opening bored in the ring (shown as entering the compression tube in Figure 6). Through this opening the gases are led into the cylinder or removed from it.

The cylinder is placed in a container and surrounded by a nonconducting packing material. An electrical heating wire permits its temperature to be raised 80 degrees in 50 minutes. The temperature is controlled by two thermometers placed one at each end of the container. The container thus has but two openings: one to expose the glass tube in which the explosion takes place; the other to permit the manipulation of the stopcock r.

8. The Carburetor.— The gaseous mixtures are prepared in an iron vessel of 2 liters capacity (Fig. 8). There are two orifices, the one M, for introducing the liquid to be studied; the other R, for evacuation, for introducing compressed air, for establishing communication with the cylinder and for expelling the gaseous mixture after a run has been made.

The carburetor may be heated electrically, also the leads connecting it with the cylinder. A thermometer placed in a

sheath at the top gives the temperature of the mixture.

A vacuum and a pressure indicator are also accessories to the outfit. These insure against leakage before each trial.

By means of a pipette graduated to 1/100 cubic centimeters a given quantity of the hydrocarbon may be introduced into the carburetor through the stopcock M. There is then added the desired quantity of air at a given pressure. This operation may be performed rapidly since the vapor pressure of the hydrocarbon may be neglected in establishing the final pressure. The ratio $c = \frac{\text{hydrocarbon}}{\text{air}}$ may thus be known in terms of mass.

These operations completed, the mixture is then heated to a temperature insuring the vaporization of the hydrocarbon. In the meantime the cylinder is brought to the required temperature and filled with the carbureted mixture. To this end the cylinder was first evacuated. The filling is then a simple matter of manipulating stopcocks. Before compression and firing the charge in the cylinder has been brought to atmospheric pressure. The stopcock R is then closed.

9. The Compression.-- A steel ram weighing about 10 kg is suspended from the ceiling by an aluminum grid 2.5 meters long (Fig. 9').

The rigidity of the system in its vertical plane of displacement is secured by means of steel wires fastening the ram from three points to the axis of rotation.

This pendulum, when it is released after being displaced from its position of equilibrium, strikes against the piston rod with a velocity depending on the height of displacement. This height was kept the same for all observations made.

If we denote by V the initial volume of the gases and by v the volume of the glass tube, then the ratio of compression will be $r = \frac{V}{v}$.

10. The Ignition.- A little before the piston is driven home in the tube the ram engages a spring which breaks the circuit in the primary of an induction coil. The secondary then induces a spark across the gap in the tungsten wires sealed in the end of the glass tube.

The characteristics of the ignition sparks used are given for the corresponding experiments.

11. The Photographic Equipment.- This apparatus consists of a wide-angle lens of short focus affixed to a large dark chamber on wheels within which is a cylinder 65 cm in diameter on which is wound a photographic film 2 meters in length.

The cylinder, turning freely on its axis, is operated by a small motor. The velocity of rotation may be varied between 4 and 10 revolutions per second.

The film is held on the surface of the drum by means of pins joining the two ends, and by clips along the edges.

The dark chamber may be opened from the front bearing the lens. The focus is adjusted through an opening near the lens.

The speed of the rotating drum is determined by means of a contact which, at each revolution closes a circuit operating a relay. This signal records these contacts on a revolving drum along side the trace made by a tuning fork.

In the course of our experimental work we found it preferable to register the time on the photographic film itself. This was accomplished by installing on the opposite side of the camera from that just mentioned, another lens system shown in Figure 10 that projects on the film the periodic image of the luminous slit in the shutters of a vibrating tuning fork of period $1/140$ second.

(To be followed by Technical Memorandum No. 548, containing Part II of this article.)

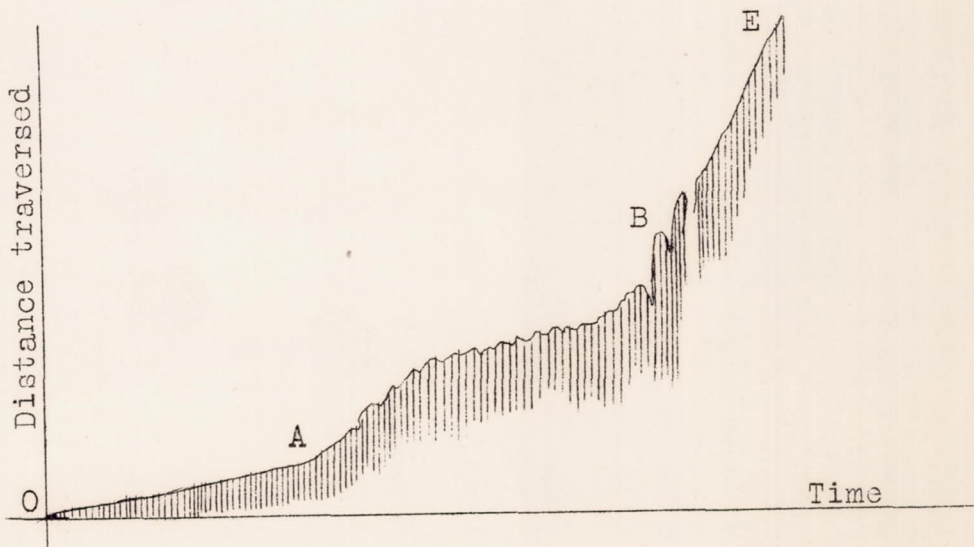
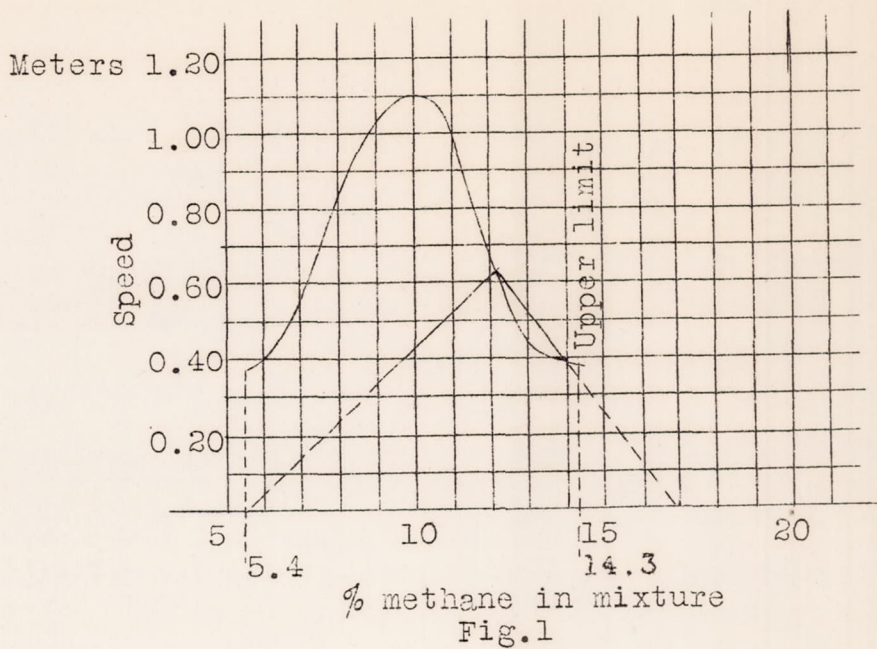


Fig.2

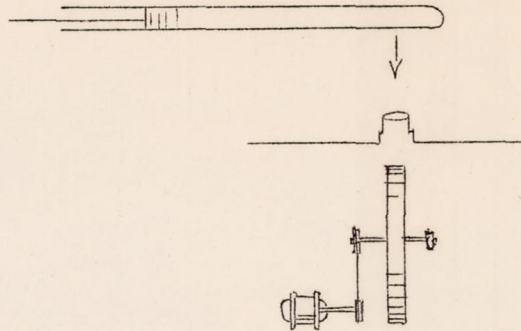


Fig. 3

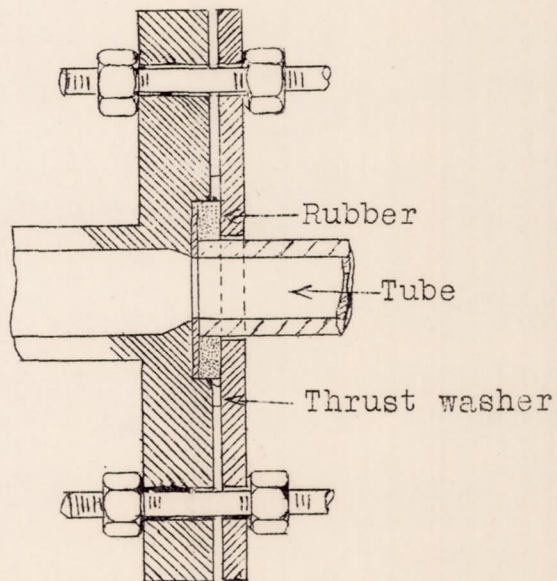


Fig. 7 Joint between tube and cylinder

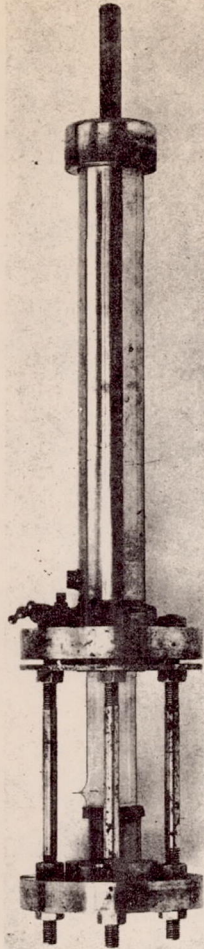


Fig.5

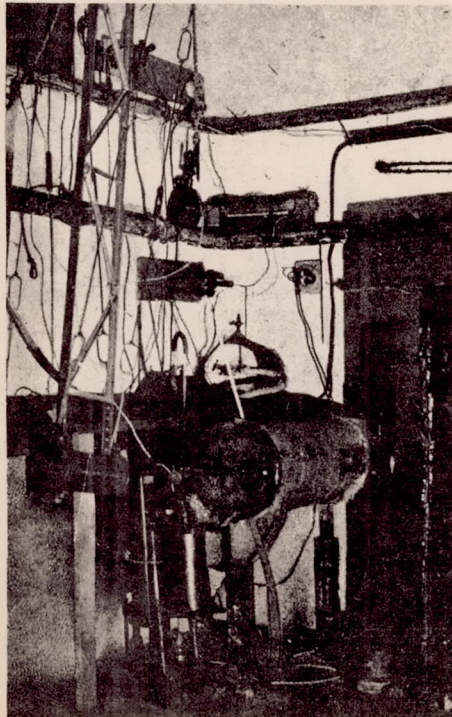
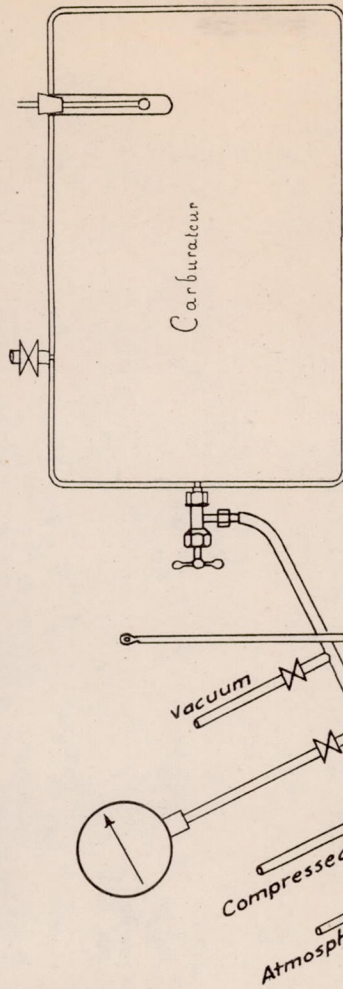


Fig.4

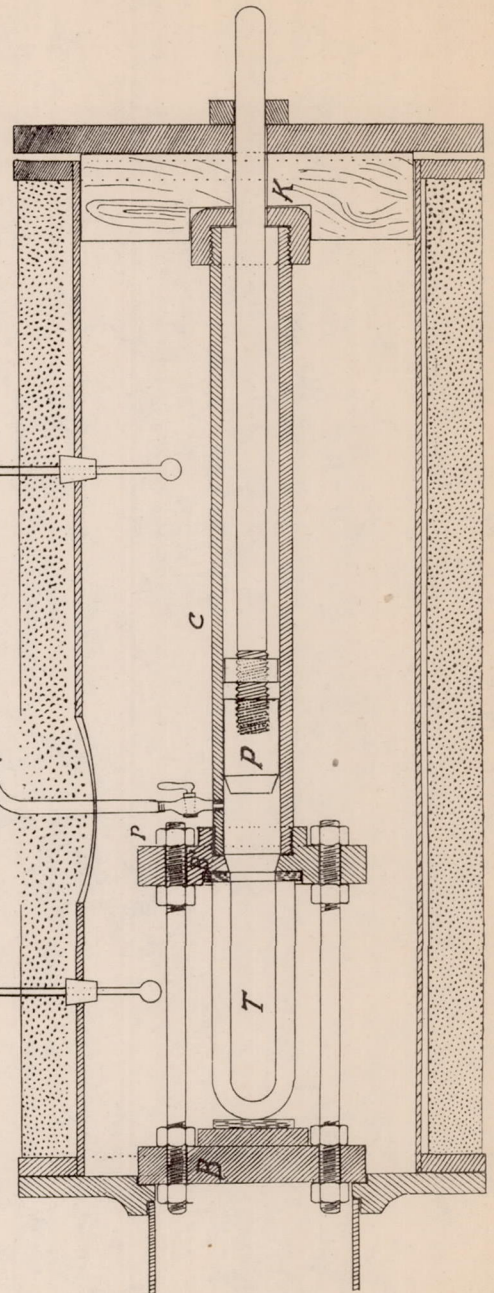


Fig.6 Compression apparatus.

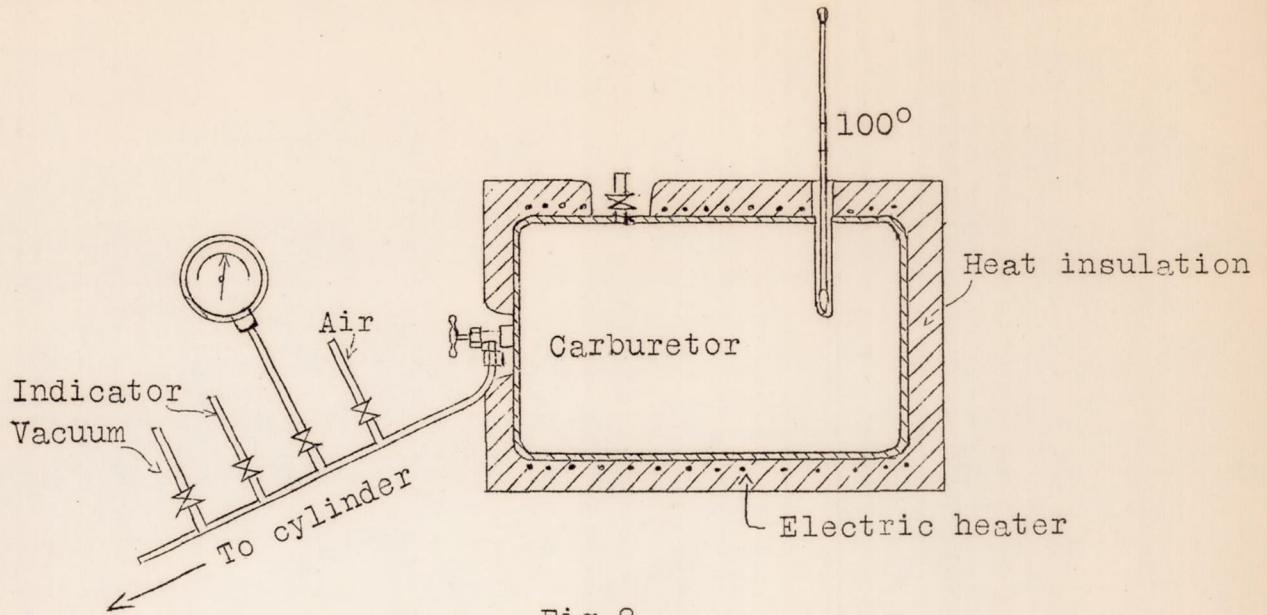


Fig.8

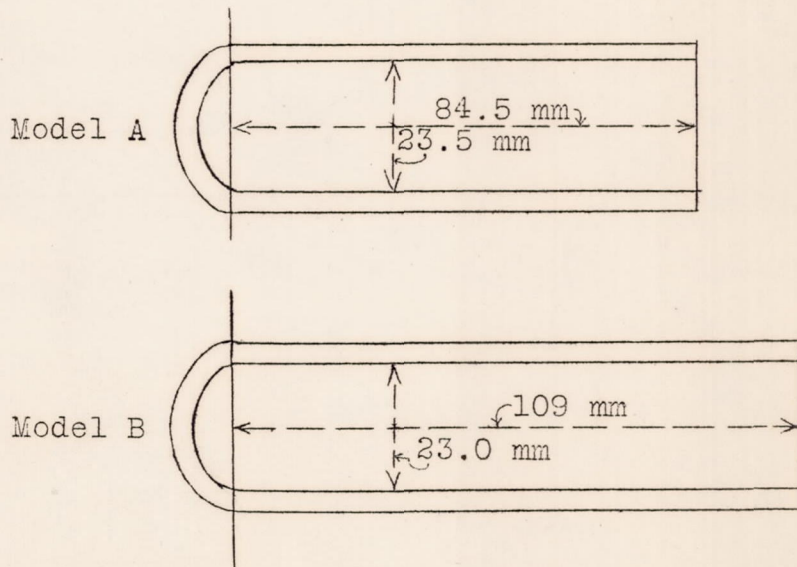


Fig.9 Tubes used in the tests.

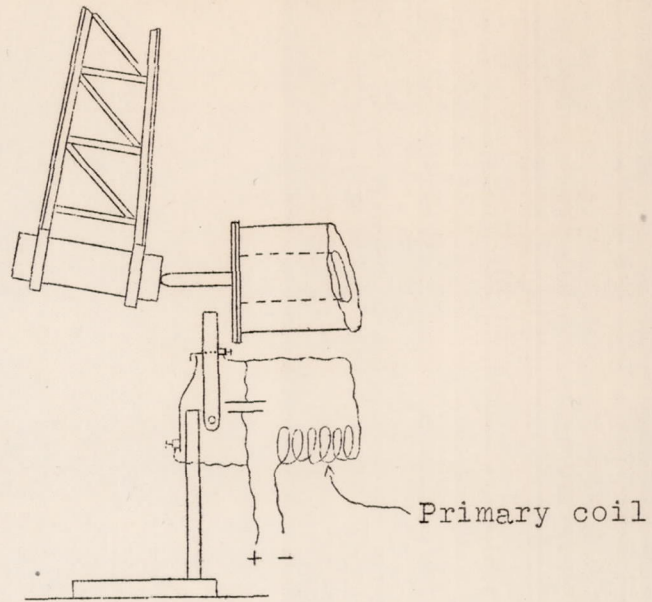


Fig.9b Interrupter (or make-and-break).

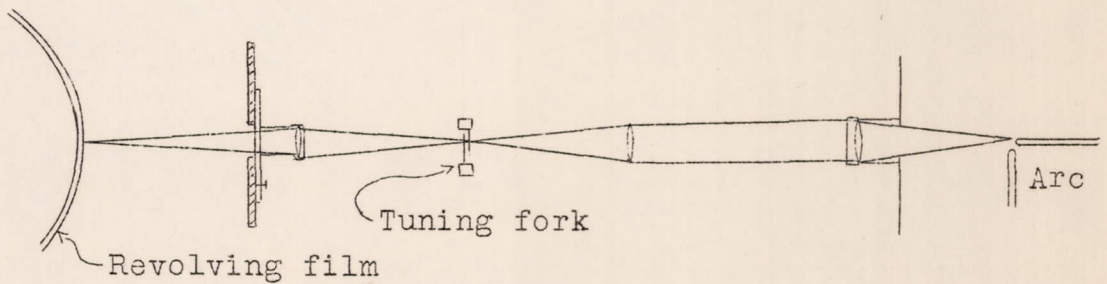


Fig.10 Time recorder.