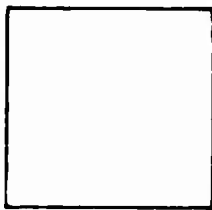


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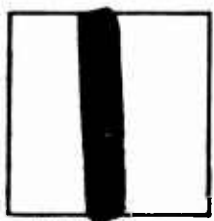
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THE APPLICABILITY OF PHOTOGRAPHY AND SPECTROSCOPY
TO SOME PROBLEMS IN INTERIOR BALLISTICS

BY

D. J. CRAWFORD, JR.

CAPT. ORD. DEPT.

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April 14, 1937.

D. J. Crawford, Jr. Capt. ORD. Dept.

**THE APPLICABILITY OF PHOTOGRAPHY AND SPECTROSCOPY
TO SOME PROBLEMS IN INTERIOR BALLISTICS**

The purpose of this report is to point out the applicability of photographic and spectrographic studies of powder gas flames in the approach of some problems of interior ballistics, and to present a program of experimental work based upon these methods.

SYNOPSIS

The report contains a survey of some recent research work on internal combustion engines, photosensitized reactions and high temperature high pressure catalytic reactions. The theoretical aspect of the lag of the total radiation field behind the thermodynamic temperature of the products of combustion and its relationship to the initiation of shock waves is discussed briefly. A program is presented suggesting photographic and spectrographic methods for obtaining information concerning linear burning rate, total radiation, transient and unstable products of combustion, temperature, accelerated erosion tests and the photo-sensitization and control of accelerated burning. Bibliography appended.

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The success of photographic and spectrographic methods in the study of transient phenomena in the burning, explosion and detonation of fuels in internal combustion engines leads to the conviction that these methods have a wide applicability, in the study of problems of interior ballistics and the development and improvement of propellant explosives. In the light of theories of modern physics and with the rich background of developments in other fields tremendous advances should be possible at this time.

A survey of recent experimental work paralleled in nature to that outlined herein leads through a vast store of literature on photosensitized reactions, catalytic reactions and explosive fuels. Particular attention has been given to treatises dealing with the transition from explosion burning to detonation conditions as evidenced by "knock" in internal combustion engines and the so-called shock waves developed in guns fired with high loading density and fast burning powders. By high speed photography the detonation zones have been located¹ and by spectroscopy

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- (a) Withrow and Boyd. Ind. Eng. Chem. May 1931
(b) Rassweiler and Withrow, Ind. Eng. Chem. June 1936

differences between explosion and detonation burning conditions have been isolated¹ in, (1) radiation distribution over the period of burning; (2) concentration of C_2 , CN , CH , OH and other radicals and unstable compounds in flame fronts and combustion products; (3) pre-flame formation of aldehydes and unstable peroxides under detonation conditions; (4) effect of the addition of detonation inducers and inhibitors on the above mentioned observations. A spectrographic method for transient flame pyrometry has been developed, for which extreme accuracy is claimed².

These studies have included the numerous theories of detonation and detonation inhibitors and have lead to a thorough investigation of the recently developed thermo-physical theory³. By bringing together the experimental evidence collected through means indicated

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1. (a) Clark and Thee, Ind. Eng. Chem, 18 (1926) P528
(b) Clark and Henne, Journal S.A.E. 20 (1927) P264
(c) Malschert - Thesis, Mass. Inst. Tech. 1928
(d) Rassweiler and Withrow, Ind. Eng. Chem. May 1932
(e) Rassweiler and Withrow, Ind. Eng. Chem. Aug. 1933
(f) Rassweiler and Withrow, Ind. Eng. Chem. Dec. 1933
(g) Rassweiler and Withrow, Ind. Eng. Chem. Dec. 1934
 2. (a) Ruark and Urey, Atoms, Molecules and Quanta P514
(b) Loomis and Perrott, Ind. Eng. Chem. 20 (1928) F1004
 3. (a) Colliard, Ord. Dept. Reference Library B-9496
(b) Semenov, Z Physik 46 (1927) P109.

above and some recent developments in theoretical and applied physics it is possible to extend and amplify the hypotheses of Colliard and at the same time remove some of the inconsistencies existing between the several theories concerning detonation inhibitors.

It is suggested that the gas kinetic energy of the products of combustion lags the radiant energy field of the explosion system and that under very rapid burning conditions the lag is sufficient to cause a material deviation from the Van der Waal equation;¹

$$\left(P + \frac{a}{v^2}\right) (V - b) = RT.$$

The Van der Waal equation assumes equilibrium between the thermodynamic temperature of the system and its radiant field. This equilibrium can never be attained in the transient reactions which transpire in an internal combustion engine or gun. The more rapid the rate of burning the further from equilibrium will conditions be. To account for this lag the equation must be written:-

$$\left(P + \frac{a}{v^2}\right) (V - b) = RT - \left[\sum E_r - \int P(\nu, T') d\nu \right]$$

where E_r is the radiant energy distribution of the total

1. Born - Atomic Physics - P20.

radiation field determined experimentally by methods suggested herein and $F(\nu, T')$ is the computed radiant energy distribution according to Planck's radiation law¹, T' being the gas kinetic temperature arrived at through Van der Waal's equation assuming equilibrium between the radiant energy field and the gas kinetic energy. If the rate of burning can be accurately determined experimentally the expression

$$\sum E_r - \int F(\nu, T') d\nu$$

becomes a parametric function of the specific heat of the gas aggregate. In the case of very rapid burning this lag manifests itself in excessive heating of the combustion vessel and in a shift of the total radiation spectrum farther into the ultra violet. When the shift into the ultra violet goes below wave length 2600 Å it may be expected to establish threshold conditions for photosensitized dissociation² and chain reactions through both direct irradiation³ and through inelastic collision

1. $U_r = \frac{8\pi h \nu^3}{c^3} \times \frac{1}{e^{\frac{h\nu}{kT}} - 1}$ Born - Atomic Physics p190

2. Ruark & Urey, Atoms molecules and quanta -p489 to 515

3. (a) Photochemical decomposition of nitrous oxide and nitric oxide. MacDonal. Jour.Chem.Soc.1928-pl.
 (b) Morikawa, Benedict and Lazer. Jour. Chem.Physics, 5 (1937).

with highly excited molecular products of combustion¹, resulting in linear burning rates or dissociation approaching or reaching detonation velocity. Such phenomena, rather than delayed ignition, account for shock waves under quick burning conditions². The oscillatory shock waves in internal combustion engines are known to be accompanied by an oscillatory radiation emission--absorption phenomenon of comparable frequency³. Hence it is not unreasonable to conclude that the true nature of the oscillatory wave is physio-chemical between a state of high dissociation--radiation emission and a state of recombination-radiation absorption. A periodic reversal of the water gas reaction following the original detonation shock would account for observed pressure and radiation changes and considerable spectrographic and photographic evidence can be brought to prove that this is precisely what occurs in the case of internal combustion engine detonation⁴.

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1. (a) Wm. Manning - Decomposition of Nitrous Oxide sensitized by mercury vapor - Thesis, Dr. Philosophy Brown Univ. 1933.
(b) Semenov, Zeits fur Physik 46 (1927) 109.
 2. Fourth Partial Report - Interior Ballistic Firings - Aberdeen P. O. 1929 - p46.
 3. The Physical Effects of Detonation in a Closed Cylindrical Chamber, C.S. Draper - N.A.C.A. Report No. 493
 4. (a) Withrow and Boyd, Ind. & Eng. Chem. May 1931
(b) Rassweiler and Withrow, Ind. Eng. Chem. May 1932

By studying the action of metallic photosensitizing agents and metallo-organic detonation inhibitors and comparing the nature of their emission spectra and resonant energy states¹, a theory is developed which explains the action of each and establishes a definite interrelation. This theory, if it can be experimentally proven, provides an efficient method for transforming high frequency radiation to lower frequency radiation or gas kinetic energy and thus inhibiting threshold conditions for photo-sensitized reactions².

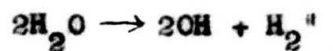
Although beyond the scope of this paper the hypotheses set forth above are subject to rigorous statistical treatment. In literature cited, Clark and Henne,³ Rassweiler and Withrow⁴, and Bell⁵ have proved the applicability of the spectrograph in studying the water gas reaction in transient high pressure flames and afterglows. Rassweiler and Withrow conclude that the usual formulation of the water gas equilibrium $H_2 + CO_2 \rightleftharpoons H_2O + CO$ is incomplete at the temperature dealt with in internal combustion chambers. They quote Berl in suggesting "that this

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1. Int. Crit. Tables. Vol. 5.
 2. Duchene - Contribution to the Study of Normal Burning in Gaseous Carbureted Mixtures: Part II. N.A.C.A. Tech Memo No. 548 (1930)
 3. Journal S.A.E. 20 (1927) p 264
 4. Ind. and Eng. Chem. May 1932.
 5. Proc. Royal Soc. B324 (1937).

equilibrium can be formulated equally well in the following way, which shows the chain mechanism for the combustion of hydrogen and carbon monoxide:-



Besides these reactions, the thermal decomposition of water probably occurs at high temperatures existing in the gases which emit the afterglow. Bouhoeffler and Reichardt formulate this in the following way:



These types of chain and dissociation reactions are known to be extremely sensitive to photo-activation by inelastic collision with excited gases¹.

Based upon these studies the following outline of a research program is proposed:-

1. Investigate the sensitivity of standard powders to direct irradiation by an ultra violet field of high energy density of all wave lengths down to 1800Å. It is very probable that certain frequency bands will cause spontaneous ignition and possible detonation at temperatures below the

1. Ruark and Urey. Atoms, Molecules and Quanta. p508-509

normal ignition point. Both nitrous oxide and nitric oxide are subject to photo-chemical dissociation by ultra violet irradiation in the region between wave lengths 1800 \AA and 2000 \AA ¹.

2. Investigate the possible photosensitization of ignition or detonation of standard powders by "collisions of the second kind"² with cool mercury vapor thrown into the wave length 2537 \AA resonant state by irradiation with a mercury arc. The electron voltage of this mercury resonant state corresponds almost exactly to the heat of dissociation of hydrogen and also to an oxygen vibrational state of unusual actinic power³. It is used in many organic dissociation processes, for the initiation and propagation of chain reactions and for sensitizing combustion under otherwise prohibitive conditions⁴. It is a more powerful sensitizer than direct irradiation. Mercury vapor in this case becomes an indispensable tool of research in that it can be excited to establish an inelastic collision and radiation system of high energy density in the ultra violet photo-chemical zone without raising its

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1. MacDonald - Jour. Chem. Soc. (1928) pl.
 2. Ruark and Urey - Atoms, Molecules and Quanta P489-515
 3. Ruark and Urey - Atoms, " " " p 498.
 4. Emeleus and Stewart, Effect of Light on the Ignition of Monosilane - Oxygen Mixtures - Trans. Faraday Soc., No. 187 (1938).

thermodynamic temperature to the ignition point of the subject material.

3. If positive results are obtained in either of the above experiments investigate the tendency of unstable metallo-organic compounds to inhibit or induce the phenomenon. Compounds of metals in the same period as mercury such as thallium, lead and bismuth should be used. The action of combustible organics of high quadrupole moment and of heavy halogens with considerable molecular absorption bands between wave lengths 1800 \AA and 2700 \AA should be investigated. The action of these three types of operators in inhibiting detonation is not identical¹. In going to high pressure conditions consideration must be given to pressure broadening² of resonant states. Bismuth with two strong resonant states corresponding to wave lengths 2897 \AA and 2937 \AA may be an efficient operator. Ipatieff³ points out that the catalytic activity of metallic elements is related to their position in the periodic table and it seems that consideration must also be given to their atomic resonant energy states, which is in agreement with the McLewis

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1. Clark and Henne, Journal S.A.E. 20 (1927) p 264
 2. White. Introduction to Atomic Spectra p 451.
 3. Ipatieff - Catalytic Reactions at High Pressure and Temperature p503-530.

theory of catalysis of which he ascribes considerable credit.

4. By means of absorption and Raman spectra determine the vibrational energy states and decomposition products of standard powders as they are brought slowly to the temperature of spontaneous ignition. Experiments of this nature should be the source of considerable information concerning the chemistry and mechanics of burning.

5. Develop eprouvettes with optical windows to withstand pressures up to limiting practical values for glass, fluorite, quartz and quartz-ice combinations. Bell¹ reports the use of direct quartz windows with explosion pressures as high as 500 atmospheres but was troubled with surface cracks. The use of 45° reflecting prisms well supported and with only a small portion of their surface under pressure is recommended. Ice has about the same transparency to ultra violet radiation as quartz and the combination of the two frozen into stainless steel mirror adapters appears promising. "Lucite", a recently developed optical plastic from the DuPont Laboratories should be thoroughly investigated.

1. Spectrographic Studies of the Explosive Combustion of Methane. Proc. Royal Soc. Jan. 1937.

6. By high speed photography determine the linear rate of burning of smokeless powder in a closed chamber, and locate zone of origin of shock waves. In the power law $\frac{dr}{dt} = KP^n$ the value for the exponential constant n is taken all the way from less than .5 to 1.5¹. To limiting pressures of optical windows this value can be determined photographically. By using a quartz system and irradiating the combustion chamber with the ultra violet source of a hot quartz mercury arc, then filtering the light before photographing to shut off all frequencies above the lowest frequency radiated by the burning powder it should be possible to determine by high speed motion pictures the process of burning of all types of granulation. Using a solid quartz cylinder head, moving pictures of internal combustion engine flames have been taken at the rate of 5000 per second². Satisfactory filters can be prepared³.

7. Using quartz optical system to limiting pressures determine by spectroscopy the radiation distribution, ultra violet shut off frequency, intermediate products of combustion which can be isolated in flame front spectra,

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1. (a) Tschappat - Ordnance & Gunnery
(b) Project for a new table for Interior Ballistics.
Dederick - Aberdeen Proving Ground 1930.
 2. Rassweiler and Withrow - High Speed Motion Pictures of Engine Flames - Ind. Eng. Chem. 28 (1926) p 672.
 3. Wood - Physical Optics. p 14-15

afterglow radiation distribution and all final products of combustion which can be isolated in spectra. If possible isolate differences between explosion burning with smooth pressure curve and explosion-detonation burning with shock wave indications in pressure curve. Correlate absorption frequencies of powder as it approaches its ignition point with the distribution of its radiation field under the above-mentioned conditions. Study the effect of metallo-organic additions on the depression of the ultra violet field¹, over the range which might be expected to exercise photo-sensitizing influence on powder molecules approaching their ignition point. Correlate time-pressure curve with radiation distribution in all studies. In connection with this work an ultra sensitive $\frac{dp}{dt}$ indicator should be developed for the study of shock waves. An instrument of this kind was developed at Mass. Inst. of Tech., for use on internal combustion engines².

8. Extend the spectrographic method for transient flame pyrometry³ employed on internal combustion engines

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1. Duchene. Part II N.A.C.A. Tech. Memo No. 548 (1930)
 2. Draper. N.A.C.A. Report No. 493 (1934)
 3. Loomis and Perrott. Ind. & Eng. Chem. 20 (1928) p 1004.

to the measuring of closed chamber powder gas temperatures. Correlate temperatures thus obtained with pressure curves and radiation distribution.

9. Investigate by spectroscopy the temperature, the molecular composition and the potential energy states of muzzle flashes from standard powders. Information obtained should throw light on remaining energy of gases. Ultra violet, high speed motion pictures of muzzle flashes should also give information on unburned portion of charge and effects of different types of granulation.

10. Investigate to determine the relationship, if any, existing between the heat of adsorption of oxygen and the resistance of a metal to powder gas erosion. There is good reason to believe that up to the time that the heat of adsorption of oxygen is supplied to the surface of a metal it is protected by a film of one or more molecular layers of oxygen, which, when the surface reaches a critical temperature, is released and forms an oxide with the surface layer of the metal. This oxide upon being removed by the abrasion of the high velocity gases leaves the surface unprotected and in a state favoring the diffusion of, and combination with contiguous gases, until it drops below the critical

temperature and again forms the adsorbed oxygen layer. Tungsten is known to exhibit this phenomenon and has a critical temperature of 1200°C¹. Positive results will go far to explain why, with a given metal, erosion appears to vary directly with powder temperature, but with a given powder temperature, follows no definite law with respect to melting point or any other physical property investigated. Positive information will be of great value in the investigation of erosion resisting material and coatings and will furnish a basis for setting up an accelerated erosion test. Tests of this nature should be carried out in closed chambers with the test plug serving as a gas part between the burning charge on one side and a small expansion chamber on the other. In this way the temperature of the plug at the time of firing can be controlled by pre-heating devices and the effect of pressure observed. By providing the expansion chamber with an optical window the build up of iron in the flame at the plug orifice can be observed with the spectrograph giving the resistance to erosion of the subject metal with a relatively few firings. The same test is applicable to the erosive tendencies of different powders.

1. Ipatieff. Catalytic Reactions at High Temperatures and Pressures . p519.

The extensive scope of this program is well recognized, as are the prerequisites of equipment and trained personnel for its execution. The logical place for the initiation of most of the items is Aberdeen Proving Ground. However, the Metallurgical Laboratory at Watertown Arsenal is probably best prepared to undertake the work on erosion. It would be necessary to add a quartz spectrograph of large classification to the present equipment which is a grating type and lacks the light conservation required for transient flame spectroscopy. This additional instrument will fill a need that is already being felt with the rapid development of spectrographic methods.

The work on flame and afterglow emission, absorption and Raman spectra will require a large quartz spectrograph with auxiliary light source equipment and a density comparator provided for reading plate densities. Bausch and Lomb have such a density comparator on the market.

For the high speed photography the high speed camera at the Proving Ground is probably on a par with the one described by Hassweiler and Withrow although of different design, but would have to be equipped with a quartz optical system for ultra violet work. Selective timing devices are

also probably available. The development of high pressure optical windows must become a part of the program but shop personnel at the Proving Ground are competent to handle the work. The irradiation experiments will require a quartz mercury lamp with a laboratory source of direct current, reaction chambers with quartz windows, and a certain amount of glassware and small laboratory equipment. It will also be desirable to use low vacuum equipment requiring a mechanical pump. Crow and Grimshaw¹ evacuated their closed chambers after loading and employed gaseous igniters. It is assumed that required pressure measuring devices and oscillograph equipment is available. Crow and Grimshaw describe a spring manometer with a "million to one" mirror multiplier which appears to be adaptable to a combination with the $\frac{dp}{dt}$ indicator previously mentioned.


Should this program be taken up actively it is suggested that full advantage be taken of such consultary service as can be obtained from Dr. R. W. Wood,

1. On the Equation of State of Propellant Gases.
Phil. Trans. Royal Soc. A 682 (1951) 1

Physics Dept., John Hopkins University, who is an authority on resonance phenomena of metallic gases and has originated some work in spectroscopy of explosives; Drs. Rassweiler and Withrow, General Motor Laboratory, who for several years have been actively engaged in an extensive research program on photography and spectroscopy of engine flames; and Dr. V. N. Ipatieff, Chemistry Dept., Northwestern University, who since 1900 has been engaged in experimental work on catalytic reactions at high pressure and high temperature and is one of the worlds best recognized authorities on the subject.

Due appreciation is expressed to Col. R.H.Sommers, Ord., Dept. U.S.Army for the loan of notes and literature on interior ballistics, to Miss F. M. O'Halloran, Librarian at Watertown Arsenal, for assistance in securing sources of information and to Mr. J. Sterner, Junior Physicist, Watertown Arsenal for critical comments.

Respectfully submitted,


D. J. Crawford, Jr.,
Capt. Ord. Dept.

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