30. *On an Irregular Mode of Spherical Propagation of Flame.*¹⁾

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In a previous communication²⁾ we have shown that when some combustible mixture of H_2 and air is ignited by a small spark, the flame front which is nearly spherical in its initial stage is corrugated by a characteristic pattern of irregular creases. Later, the experiment has been continued with some points of improvements introduced in the method and apparatus of experiment. The combustion chamber in use is of a cylindrical form, 5.0 cm. in length and 6.9 cm. in diameter. The propagation of the flame front is photographed kinematographically on a revolving film by the "Schlieren" method, the intermittent illumination being made by the sparks with a frequency of 100 per sec., fed by a transformer excited with 50 cycle A.C. The mixtures studied are, as before, those near the limits of inflammability, either upper or lower. For filling the combustion chamber with the gas, the use of water was avoided and the repeated previous evacuations of it by Cenco oil pump was preferred.

Those different mixtures of H_2 with different proportions of O_2 and N_2 were tested, which showed nearly equal velocities of propagation, and the factor determining the appearance of the corrugation of the flame front was investigated. It was found that the corrugation is not merely determined by the magnitude of the velocity of propagation. Thus, for example, a mixture with about 72 % H_2 and 28 % air (i.e. 5.9% O_2) shows no sensible corrugation, though its velocity of propagation is much larger than those mixtures with about 12 % H_2 , 10% O_2 and 78 % N_2 which display marked corrugated patterns. On comparing different mixtures with 10–15 % H_2 with different proportions of O_2 and N_2 , it was found that those showing conspicuous corrugations contain some excess of O_2 above the theoretical amount for the complete combustion.

1) The research is continued under the supervision of the Imperial Japanese Navy. We wish to express our best thanks to the Authorities of the Navy for the help given.

2) These Proceedings, **2** (1926), 261.

On the other hand, the mixtures of CO, C₂H₂ or coal gas with air, which are of the same order of the velocity of propagation with the above H₂-air mixtures characterized by marked corrugations, show very smooth spherical flame front. Fig. 1 (a)-(d) will illustrate the different degrees of development of corrugation, in different mixtures with nearly equal flame velocities.

In the case of the mixture with 10% H₂ and 90% air, a reduction of the pressure by about 100 mm. Hg is effective in annulling the intricate corrugation, except some irregularity in the shape of the flame front due to the electrode, meanwhile the flame velocity being reduced to about 2/3 of the initial value for the atmospheric pressure.

The same mixture was dried by passing through H₂SO₄. The effect of this degree of drying also reduced the tendency to form corrugation, accompanied, of course, with the conspicuous diminution in the flame velocity; viz., in an example, to about one half.

Some effect of the nature of the spark gap electrodes is observed. Thus, the corrugation is less with the electrodes consisting of copper wires of 0.8 mm. diameter than with those of brass rods of 3.6 mm. diameter. It is sensibly decreased when the rod electrodes are covered with glass tube except its gap end.

This latter fact suggests that the corrugation is brought about by the shrinkage of the volume of the gas in the inside of the flame front by the rapid condensation of the water vapour formed¹⁾. This suggestion is favourable for explaining the smooth flame front in the case of CO mixture where no water is formed. It is, however, not able to explain the fact that the mixture of 12% coal gas with 88% air, or of 3% C₂H₂ with 97% air, shows a quite regular spherical propagation, though the flame velocity of the either mixture is of the same order as that for 10% H₂ mixture. In the cases of these organic gases the amounts of O₂ are also in sufficient excess, so that the excess alone cannot be considered as the sole determining factor of the formation of the corrugation. This singular anomaly in the form of the flame front characteristic to the low % mixtures of H₂ with air or O₂ with an excess of O₂ might be possibly connected with some peculiarity of the mecha-

1) The fact that such a rapid condensation of vapour might occur has been shown by the present authors in the case of low percentage mixture of H₂ in eudiometer tube, Sci. Pap. Inst. Phys. Chem. Res., **6**, No. 90 (1927), 81-127; also see the previous paper in these Proceedings, loc. cit. That a contraction of volume is actually taking place may be shown by tracing the variation of velocity with time. This latter part of the result will be reserved for a future communication.

nism of combustion and may deserve a more detailed investigation from different sides¹⁾.

The effect was also studied of mixing the vapour of some antiknock substance, such as C_2H_5Br or C_2H_5I to a low percentage mixture of H_2 with air. Comparing the two mixtures with and without the antiknock admixture which are of nearly same velocity of flame propagation, the corrugation is markedly less for the mixture with the antiknock vapour than for that with out the same.

1) The growth of the corrugated creases implies some fluctuation in the radial velocity of propagation. If the velocity V is a function of the curvature k of the flame front, the corrugation will develop when dV/dk is positive, whereas the front will remain smooth if the differential coefficient is negative. The fact that those mixtures with marked corrugations show some decrease of the velocity with time is apparently in harmony with this idea, though the alternative explanation by the contraction of the volume seems more probable for the present.

Another problem of theoretical importance, which is connected with the phenomenon here observed, is whether the volume change due to the chemical combination takes place before or after the evolution of heat. The question may worth notice, though hitherto seldom inquired.

Fig. 1

(a) $\begin{cases} \text{H}_2 & 10.2\% \\ \text{O}_2 & 89.8\% \end{cases}$

(b) $\begin{cases} \text{H}_2 & 10.5\% \\ \text{Air} & 89.5\% \end{cases}$ (O_2 18.8)

(c) $\begin{cases} \text{(b)} & 50\% \\ \text{(d)} & 50\% \end{cases}$

(d) $\begin{cases} \text{CO} & 18.5\% \\ \text{Air} & 81.5\% \end{cases}$ (O_2 17.1)