

Letters to the Editor

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The Thermal Decomposition of Acetaldehyde

In the note which I contributed on behalf of the workers in my laboratory to the discussion at the meeting of the Royal Society on May 10, it was stated that our results showed that the rate of thermal decomposition of acetaldehyde was influenced by the state of the surface of the silica, which was contrary to the conclusion arrived at by Hinshelwood and Hutchins¹, who had stated that the process was "practically entirely homogeneous". When I had read my note, Mr. Hinshelwood commented on my statement, and according to the report of the meeting which has just been published said,—"In our experiments the decomposition of acetaldehyde is an absolutely homogeneous reaction unaffected by the character of the vessel." A reply demanded reference to the actual facts recorded by Hinshelwood and Hutchins, and I proposed to look up the paper and submit one in writing, but I was not permitted to do so. As Mr. Hinshelwood built up his hypothesis on the results of these and similar experiments, it is worth while considering how far his deductions are legitimate, and I take this opportunity of making a few observations regarding them.

In the paper to which I have referred, it is stated that in two experiments carried out at 518°, in which two different silica bulbs were used, values of the velocity constant were obtained, (I) 0.333 0.352, (II) 0.350 0.349, showing a maximum difference of only 3 per cent. When one of these tubes was packed with silica chips, the values of the constant obtained were 0.461 0.477, the maximum increase being 43 per cent. In experiments at 464° the value for the constant with the empty bulb was 0.049 and for the packed bulb 0.066, the increase being 35 per cent. The authors argue that because the surface of the silica chips represented an area of some twenty times that of the empty bulb, the increase in the velocity constant is negligible. They assume that the effect of unit area of the broken silica and of the fire-polished surface of the tube are identical, which is not in accordance with experience. These experiments merely show that with different tubes treated in a similar manner one may obtain almost identical results, but that the effect of surface is not negligible. With this we are in perfect accord. Certainly the data do not justify the statement that the thermal decomposition of acetaldehyde is "absolutely homogeneous".

Mr. Hinshelwood's suggestion that the effect of surface would only be observed at low temperatures is negated by subsequent experiments in my laboratory. His second suggestion that our experiments were affected by the condensation of oxygen on the walls of the reaction tubes is not supported by the facts. It is our invariable practice to fill a reaction tube with hydrogen, and to heat it overnight to about 600°. It is then exhausted while hot, cooled, and the measured quantity of acetaldehyde vapour is condensed in it, after which it is sealed.

There is no chance of oxygen coming into contact with the surface. The removal of oxygen from liquid acetaldehyde, in which the gas is very soluble, is a very difficult matter. It was effected by distilling the liquid *in vacuo* at low temperature through a series of vessels sealed together, and finally condensing it in the filling apparatus without contact with air.

The pre-treatment of the tubes with hydrogen for some hours enables one to obtain concordant results in the case of most reactions when using the same tube, but only in the case of very few reactions with all the tubes used. Indeed, out of a large number of reactions which we have studied, only in the case of the pyrolysis of ethane-ethylene-hydrogen equilibrium mixtures does the effect of surface appear to be practically negligible. Even in this case I should not care to use the word "absolutely", even in the broadest and most popular sense. The criteria of surface action are very obscure, and two or three experiments with similar tubes, treated in an identical manner, are insufficient to determine whether the influence of surface is material or not. The experiment described by Dr. A. Farkas at the recent meeting of the British Association, in which D₂ was replaced by H₂ by heating it in a silica tube which was supposed to be completely degassed by the usual high temperature and high vacuum treatment, suggests caution in dealing with the results of gas reactions carried out in silica apparatus.

Actually the main difference between our experimental results and those which I have discussed lies in the fact that, in one case the reaction rates are measured by observing the rate of formation of methane, which requires detailed analysis, and in the other they are deduced from the total rise of pressure, from which it is not possible to eliminate the effect of side reactions.

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¹ *Proc. Roy. Soc., A*, 111, 384.

Human Daily Requirements of Dietary Ascorbic Acid

EARLIER investigations¹ on insane, but physically healthy, experimental subjects, who have been forcibly tube-fed owing to their refusal to take nourishment, have established certain relations between the protective dose of an antiscorbutic required by man and the protective dose of the same antiscorbutic required by a guinea pig, on the view that the degree of susceptibility to microscopic scorbutic alterations in the teeth in guinea pigs and the pre-scorbutic reduction in the strength of the cutaneous capillaries in man are approximately the same. On the basis of this result, further experiment showed that, in order barely to protect himself against the earliest onset of a shortage of vitamin C, an adult weighing 60 kgm. requires a daily dose of an antiscorbutic (fresh orange juice) fourteen to twenty times as large as a guinea pig weighing one third of a kilogram requires barely to protect itself against scorbutic alterations in the teeth which can be determined microscopically. In a further investigation, these facts have been utilised to establish indirectly man's daily requirements of ascorbic acid by means of experiments on guinea pigs.

Of thirty guinea pigs with initial weights between