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TEMPERATURE PATTERN MEASUREMENTS IN BRIGGS-RAUSCHER OSCILLATORY REACTION WITH THE STATE I TO THE STATE II TRANSITION

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ABSTRACT:
Thermal monitoring of a Briggs-Rauscher oscillatory reaction with a phase transition or the state I (low concentration of iodide and iodine) to the state II (high concentration of iodide and iodine) transition with formation of new solid iodine phase is presented. This is the very first time that an oscillatory reaction and the state I to state II transition have been monitored using a thermal camera. It should be pointed out, that the BR reaction solution is not homogenized by stirring, and that only oxygen production influence the solution mixing. Therefore, the analysis was done at three sampling points on the cuvette where temperature change over time was observed, and compared with average temperature distribution. The first point is chosen to correspond to the top of the solution in the cuvette, followed by one in the middle of the volume and one at the very bottom of the cuvette. Although the state I to the state II transition itself is not reproducible (investigated transition exhibits crazy clock behavior), the overall temperature pattern has shown a reproducible character.

Keywords: *temperature distribution, Briggs-Rauscher reaction, crazy clock, solid iodine, nonlinear phenomena*

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1. INTRODUCTION

This article reports the analysis of the results obtained by examining the temperature profile of the Briggs-Rauscher (BR) reaction.[1] This non-equilibrium oscillatory reaction is particularly interesting for this observation angle since it happens with phase transition, e.g. solid iodine formation.[2,3] The phase transition occurs between two BR reaction states: state I (low iodide and iodine concentration) when oxygen bubbles are dominantly visible and state II (high iodide and iodine concentration, with solid iodine) when the coloration of the solution becomes dominant.[4] This analysis enables the correlation of the dynamics of the phase transition and the temperature change using very precise measurements by thermal camera[5]. It should be pointed out, that the BR reaction solution is not homogenized by stirring, and that only oxygen production influences the solution mixing. It is also important to note that the phase transition in BR reaction is characterized by crazy clock behavior, which means that its induction period is not repeated i.e. the phase transition is irreproducible.[2] Presented results are the first of this kind in oscillatory reaction measurements and could have the great importance in understanding local temperature distribution when there is no mixing.

2. EXPERIMENTAL

The phase transition phenomena occurring in the oscillating BR reaction was carried out with concentrations of reactants: $[CH_2(COOH)_2]_0$ = 0.0789 mol dm^{-3}, $[MnSO_4]_0$ = 0.0075 mol dm^{-3}, $[HClO4]_0$ = 0.03 mol dm^{-3}, $[KIO_3]_0$ = 0.0752 mol dm^{-3} and $[H_2O_2]_0$ = 1.269 mol dm^{-3} equal to the ones in the study by Pagnacco et al.[2], but with reaction volume divided by ten. The total volume used for the BR reaction was equal to 2.5 ml and reaction is followed in plastic cuvette. The experiments are done in ambient temperature.
The change in temperature is monitored by a thermal camera "FLIR A65" (640 x 512 pixel, thermal resolution 50 mK, focal length 13 mm, field of view angle 45°x37°). The camera is calibrated before experiments on ambient temperature 26 ^{o}C measured by mercury thermometer. The temperature change was observed at three points during the entire BR reaction. The reference points were chosen to be at the top, i. e., on the surface of the solution, in the middle of the same volume, and at the bottom of the cuvette. The setup used in experiment is presented in Fig. 1.

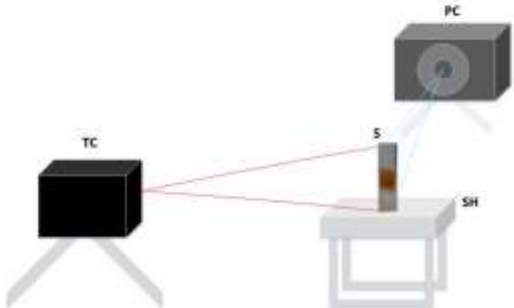

Fig. 1. The setup: photo camera (PC), thermal camera (TC), sample (S): BR reaction in cuvette and sample holder (SH)

3. RESULTS AND DISCUSSION

The temperature profile was observed during the entire reaction in the three previously mentioned points (see Fig. 2). It is important to note that the upper point was chosen as a point on the surface of the solution at the moment of adding the reagent (necessary to cause a reaction), and that during the reaction, the volume of the solution in the cuvette increases, and therefore the surface of the solution is no longer at the same height in the cuvette. This level change occurs when oxygen begins to intensively leave the reaction. Thus, the point chosen at the beginning does not change its place, but at certain moments, it is no longer on the surface of the solution. Another important factor regarding these three positions is that the lowest selected point is positioned at a very small distance from the metal support of the cuvette.

Fig. 2. The three points on the cuvette in which the dynamics of temperature change is observed.

It should be pointed that regardless of the position of the point, the temperature dynamics is almost the same. Only a slight shift in a moment of increasing the temperature was observed, which was attributed to the gradual coloration from top to bottom. This result must be verified by observing a large number of reactions and, if possible, by catching and measuring the reaction with a thermal camera when the coloration moves in the opposite direction.
When attention is paid to the degree of heating, it is observed that on average the cuvette is heated by about two degrees due to the phase change, i.e. the coloring of the reaction (see Fig. 3). The recorded result refers to the BR reaction recorded with a thermal camera for the case when mixing occurs only naturally due to the dynamics of oxygen bubbles.

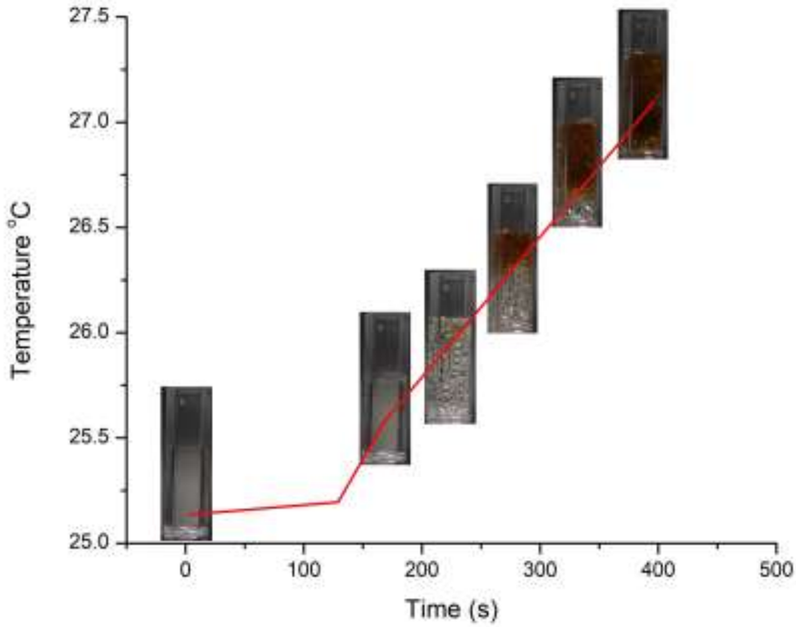

Fig. 3. Temperature profile of the BR reaction.

Fig. 3 shows a rough temperature profile, while for a precise representation it is necessary to analyze and define various factors that affect heating and are not part of the process itself. For example, such a comparative measurement in three points showed that although an almost identical temperature profile is observed, the lowest point still lags behind in terms of temperature increase. The reason for obtained temperature behavior could be cold metal cuvette supporter. It is an important conclusion that metal supporter has significant impact on reaction temperature. This measurement is also related to the conditions found

in the laboratory. For future measurements, it is planned to take into account different external temperatures, as well as different circumstances in terms of air oscillation around the cuvette caused by other laboratory equipment.
The most important conclusion from the obtained measurements is that although a completely non-reproducible reaction was observed, the temperature profile is somewhat reproducible. This point of view opens numerous questions and defines the further course of research.

3. CONCLUSION

The non-equilibrium oscillatory Briggs-Rauscher (BR) reaction was monitored with a thermal camera. The recorded temperature changes were analyzed. Changes were recorded in three key points of cuvette (top, middle and bottom) during BR reaction. Although the state I to the state II transition itself is not reproducible (investigated transition exhibits crazy clock behavior), the overall temperature pattern in three points has shown a reproducible character. Additionally, the upper and the middle spots also have the identical temperature, while the bottom temperature significantly distinguish. The reason for obtained temperature behavior could be cold metal cuvette supporter, indicating the significant impact of laboratory measurement conditions on temperature of BR reaction mixture.

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5. LITERATURE

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