

IGNITION AND QUENCHING OF FLAMMABLE *n*-BUTANE-AIR MIXTURE AT VARIOUS INITIAL PRESSURES. CRITICAL PROPERTIES

Nicoleta-Gabriela MUȘAT,^a Valentin MUNTEANU,^a Mihaela PUIU,^a Domnina RĂZUȘ^b and Dumitru OANCEA^a

^a Department of Physical Chemistry, University of Bucharest, 4-12 bd. Elisabeta, 030018 Bucharest, Roumania

^b "Ilie Murgulescu" Institute of Physical Chemistry, Roumanian Academy
202 Splaiul Independentei, 060021 Bucharest, Roumania

Received May 22, 2009

Critical properties like quenching distance, d_q , maximum experimental safe gap, MESG, and minimum ignition energy, E_{\min} , have been determined by either high voltage inductive-capacitive sparks and low voltage break sparks, by testing *n*-butane-air flammable compositions around the stoichiometric mixture, and various atmospheric and below atmospheric initial pressures. The correlation between MESG and d_q has been verified and it resulted that MESG is approximately half of d_q , in agreement with previously reported data. It was also found that the energy requirement for low voltage spark is at least twice higher than that for high voltage spark. The results agree with our previous findings for other hydrocarbon/air systems.

INTRODUCTION

Among the most important characteristics in terms of safety against accidental explosion, the quenching distance, d_q , the maximum experimental safe gap, MESG, and the minimum ignition current, i_{\min} , are frequently measured, discussed and analyzed.¹ These parameters have been experimentally determined at different initial pressures (atmospheric and subatmospheric), and for various compositions of *n*-butane/air mixtures. The specific objectives of this work were the measurement of d_q and MESG and application of the simple correlation model, the measurement of i_{\min} and derivation of the low voltage minimum ignition energy, E_{\min}^{LV} , which is compared with the high voltage minimum ignition energy, E_{\min}^{HV} , obtained via d_q through the means of correlation model, and the evaluation of overall reaction order from the pressure dependence of either d_q and or i_{\min} .

EXPERIMENTAL

Materials: The previously described experimental setups^{2,3} have been used to measure quenching distance and

minimum ignition current variation with initial pressure for *n*-butane-air compositions of 2.6%, 2.9%, 3.12% (stoichiometric mixture), 4%, 4.5%, 5.0%, 5.5%, respectively. Two 10-L steel cylinders have been used to prepare the gas mixtures by partial pressure method, at 500 kPa absolute pressure. *n*-Butane, 99.95%, and compressed air, 5.0 grade, were provided by SIAD RG. The mixtures were allowed at least 24 hours to mix prior to use. A test cell (shown in fig. 1), manufactured at PTB-Braunschweig, was used to directly measure MESG. The test cell, made of stainless steel, consists of two concentric cells that communicate through a variable slit opening which is set to the desired length with a micrometric screw.



Fig. 1 – Test cell for MESG measurement: 1–admission valve; 2–micrometric screw; 3–observation window.

* Corresponding author: valentin.munteanu@g.unibuc.ro

An observation window allows visualization of the flame initiated in the inner cell, by the help of high voltage electric spark circuitry. The maximum experimental safe gap is determined by averaging two consecutive runs, one being the highest slit opening that forbids the flame to pass through and the second being the smallest that allows the flame into the outer cell.

RESULTS AND DISCUSSION

Tables 1 and 2 show experimental quenching distances, minimum ignition currents and maximum experimental safe gaps for the *n*-butane-air compositions taken under study.

Table 1

Quenching distances and minimum ignition currents of selected *n*-butane-air compositions at various initial pressures and ambient temperatures (units: d_q , mm; i_{min} , mA; p_0 , kPa)

	conc. p_0	2.60 %		2.90 %		3.12 %		4.00 %		4.50 %		5.00 %		5.50 %	
		d_q	i_{min}	d_q	i_{min}	d_q	i_{min}	d_q	i_{min}	d_q	i_{min}	d_q	i_{min}	d_q	i_{min}
1	101.3	3.12	188	2.05	182	1.70	138	1.21	124	1.43	93	1.56	108	2.13	112
2	91.3	3.30	203	2.23	198	1.95	145	1.36	137	1.55	102	1.78	123	2.37	122
3	81.3	3.62	228	2.50	202	2.22	162	1.59	146	1.66	108	1.88	127	2.78	128
4	71.3	4.29	242	2.76	218	2.45	182	1.91	162	2.04	118	2.16	136	3.30	138
5	61.3	5.16	263	3.45	253	2.63	198	2.22	173	2.40	132	2.48	153	3.67	153
6	51.3	5.62	272	4.24	268	3.49	212	2.59	200	2.85	148	3.01	160	4.26	167
7	41.3	7.43	295	5.33	288	4.44	238	3.45	211	3.52	170	3.62	278	5.28	198
8	31.3	–	–	7.46	–	5.95	248	4.62	238	4.78	213	4.19	–	7.23	–

Table 2

Maximum experimental safe gaps (in mm) of selected *n*-butane-air compositions at various initial pressures and ambient temperatures

	conc. p_0	2.60 %	2.90 %	3.12 %	4.00 %	4.50 %	5.00 %	5.50 %
		1	101.3	1.52	1.03	0.89	0.63	0.70
2	91.3	1.68	1.12	0.98	0.70	0.91	0.85	1.14
3	81.3	1.83	1.28	1.16	0.77	0.95	1.01	1.36
4	71.3	2.12	1.37	1.22	1.00	1.12	1.23	1.75
5	61.3	2.61	1.73	1.33	1.25	1.29	1.38	1.89
6	51.3	2.83	2.13	1.71	1.47	1.46	1.49	2.14
7	41.3	3.69	2.63	2.23	1.75	1.79	1.87	2.66
8	31.3	3.84	3.88	2.92	2.38	2.13	2.02	3.61

One expected that maximum experimental safe gap is about half of the corresponding experimental quenching distance for the same flammable mixture under the same ambient conditions. In order to verify this assumption the linear regression analysis was applied to 55 experimental pairs. The results come to confirm the validity of this assumption (see the plot of residuals, fig. 2):

$$d_q = (1.98 \pm 0.01) \times \text{MESG} \quad (1)$$

The experimental i_{min} has been further used to compute low voltage minimum ignition energy, E_{min}^{LV} :

$$E_{min}^{LV} = \frac{L \cdot i_{min}^2}{2} \quad (2)$$

where L represents the inductance, in H.

While E_{min}^{LV} was determined exclusively from the characteristics of electrical circuitry, the minimum ignition energies for the ignition by high voltage inductive-capacitive sparks, E_{min}^{HV} , were computed via d_q data. A previously developed correlation model⁴ has been used at this stage. The model takes into account the initial pressure of the mixture:

$$E_{min}^{HV} = k \cdot p_0 \cdot d_q^3 \quad (3)$$

where $k = 0.445$ is a proportionality constant and p_0 is the initial pressure (p_0 , d_q and E_{\min} are expressed in SI units).

The computation of E_{\min}^{HV} as a function of mixture composition shows a parabolic behaviour for either atmospheric or subatmospheric initial pressure. The minimum of this dependence is around 4.0% *n*-butane-air (equivalence ratio 1.3); this minimum is attributed to the most or the easiest ignitable mixture.

Fig. 3 shows the variation of E_{\min}^{LV} with mixture composition, at selected initial pressures. Although the shape of the dependence is somewhat similar to that of E_{\min}^{LV} , there are differences in rich mixture as well as at lower initial pressures. One can observe that parabola is not well defined and the minimum is displaced towards richer domain. The trend is even more significant at lower (*i.e.* 41.3 kPa) initial pressure.

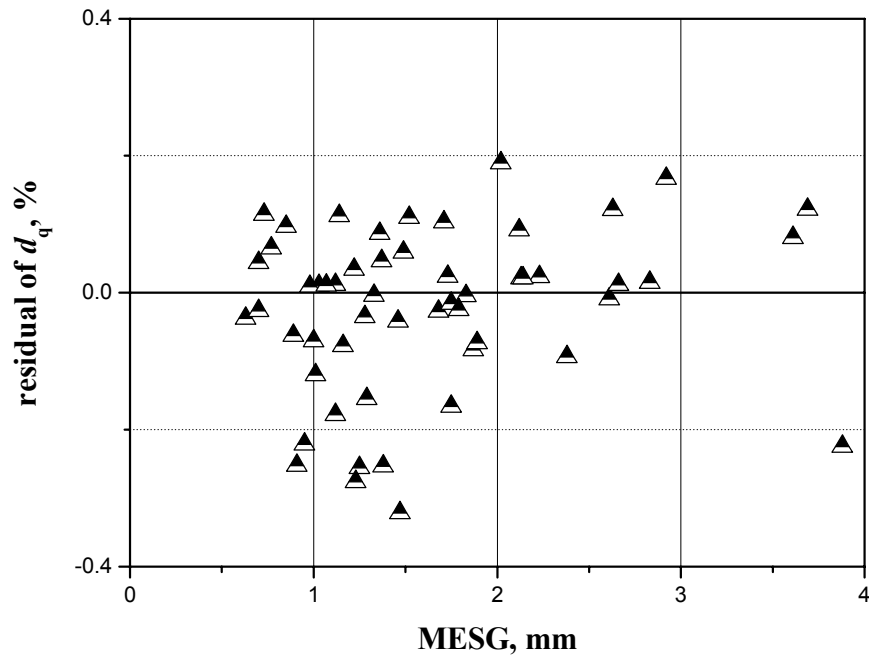


Fig. 2 – Residual plot of the dependence $d_q = f(\text{MESG})$.

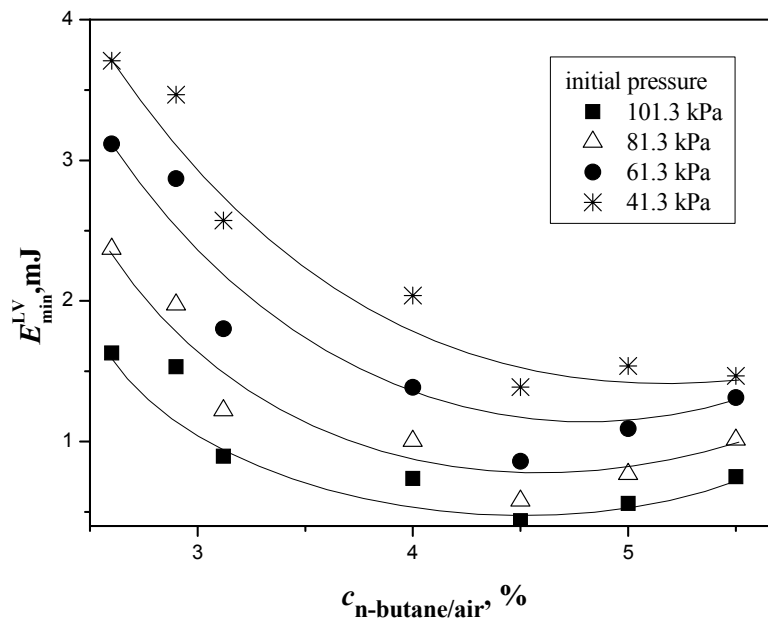


Fig. 3 – Low voltage minimum ignition energy for different *n*-butane/air compositions, at selected initial pressures.

Fig. 4 gives a comparison of the two ignition energies at atmospheric initial pressure. One clearly observes that the minimum ignition energy obtained through low voltage inductive spark is higher than the corresponding high voltage capacitive spark. The increasing factor ranges from 1.7, at the lean or rich limits of the tested compositions, to more than 4 at the region of the most readily ignitable mixture. This observation is also in agreement with our previous findings.^{2, 3} The main cause that E_{\min}^{LV} is greater than E_{\min}^{HV} is strictly related to the geometry of the ignition system: for high voltage spark the electrodes are

separated by a finite distance (known as electrode gap) and the spark is generated inside this gap; the energy deposited in the spark is almost entirely transmitted to the mixture. In the case of low voltage spark, the two electrodes are practically connected. When the electric circuit is broken (disconnection of the electrodes), a very short separation distance appears between the electrodes and an energetic spark is generated. The energy of the spark is in this case partly transferred to the mixture and the rest is lost to the ignition electrodes.

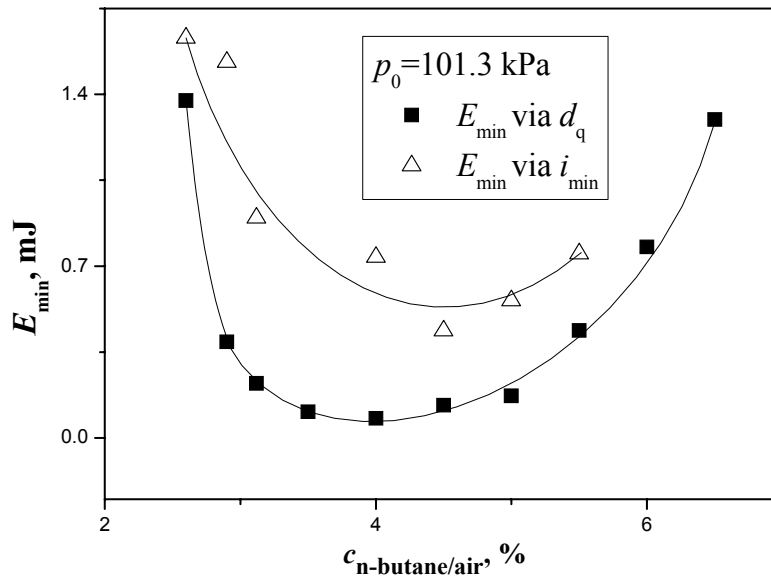


Fig. 4 – Comparison between minimum ignition energies obtained with different ignition systems.

Finally, the pressure dependence of either d_q or i_{\min} allowed the evaluation of the overall reaction order, an important kinetic parameter measuring the reaction rate sensitivity to total pressure changes.

The expressions which gives the physical significance of the overall reaction order in relation to the quenching distance or minimum ignition current are:^{5,6}

$$\begin{aligned} d_q &\approx p_0^{m_1}; & i_{\min} &\approx p_0^{m_2} \\ m_1 &\approx -0.5 \cdot n; & m_2 &= 0.5 \cdot (1 - 3n/2) \end{aligned} \quad (4)$$

where m_1 and m_2 are the baric exponents of d_q , and of i_{\min} , respectively, and n is the overall reaction

order. The values are computed from the slope of the linearized dependency of d_q or i_{\min} on pressure.

Although there is some discrepancy between the results obtained via quenching distance as compared to those obtained via minimum ignition current (see table 3, ranging from 1.48 to 2.14), they are consistent with both theory^{7, 8} and our previous experimental data on hydrocarbon combustion.⁵ Such discrepancy is, however, attributed not only to the difference in experimental techniques, but also to the simplicity of the above expressed empirical models used to evaluate the overall reaction orders, with more accuracy expected to be in favour of the data obtained via minimum ignition current.

Table 3

Overall reaction orders derived from d_q and i_{\min} .

	n , via	<i>n</i> -butane-air concentration, %						
		2.60	2.90	3.12	4.00	4.50	5.00	5.50
1	d_q	2.04	1.84	2.08	2.14	2.06	1.96	2.00
2	i_{\min}	1.56	1.64	1.54	1.53	1.56	1.80	1.48

CONCLUSIONS

Several critical parameters of the *n*-butane-air mixture have been experimentally determined in order to verify empirical correlations between them and to obtain supplemental information about the flammability behaviour of this gaseous mixture when different spark ignition systems are applied. It was verified that maximum experimental safe gap is twice lower than the corresponding quenching distance. Minimum ignition energy shows parabolic dependence on mixture composition but the pattern is disturbed when low voltage spark is used. Moreover, low voltage E_{\min} s are higher compared to high voltage E_{\min} s due to significant energy loss towards the electrodes of the ignition system. The overall reaction orders for *n*-butane-air flame that have been evaluated from the pressure dependence of the quenching distance or minimum ignition current are in agreement with our previous studies.⁵

Acknowledgements: The authors wish to acknowledge the financial support from ANCS-CNCSIS Roumania, through the grant type ID_1008, contract no. 38/2007. Special thanks are

directed to PTB-Braunschweig, dr. Elisabeth Brandes, for providing the test cell to measure the maximum experimental safe gap.

REFERENCES

1. J.M. Martin-Valdepenas and M.A. Jimenes, Exploring MIE as a Safety Indicator Parameter in Practical Applications, in *EU-5th Framework Programme*, EVG1-CT-2001-00042: Experimental and Numerical Study of Reactive Flows with Relevance to Industrial Safety for Explosion Protection, **2003**.
2. D. Oancea, D. Razus, V. Munteanu and I. Cojocea, *J. Loss Prev. Proc. Ind.*, **2003**, *16*, 353-361.
3. V. Munteanu, N.G., Musat, D. Razus and D. Oancea, *Rev. Chim., (Bucharest)*, **2005**, *56* 951-954.
4. D. Oancea, D. Razus, F. Chirila and N.I. Ionescu, *Rev. Roum. Chim.* **1997**, *42*, 571-578.
5. V. Munteanu, D. Oancea and D. Razus, *Ann. Univ. Timisoara*, **2003**, *12*, 239-246.
6. D. Oancea, D. Razus and V. Munteanu, *Proc. 17th Int. Symp. Comb. Proc.*, Poznan, 24-27 September, Poland, **2001**, 191- 195.
7. S.R. Turns, "An Introduction to Combustion. Concepts and Applications", McGraw-Hill, USA, 1996, p.95-97.
8. W.C. Gardiner, "Gas-Phase Combustion Chemistry", Springer-Verlag, NY, 1999, p. 31-40.

