



RF number as a new index for assessing combustion hazard of flammable gases

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Abstract

A new index called RF number has been proposed for assessing the combustion hazard of all sorts of flammable gases and their mixtures. RF number represents the total expectancy of combustion hazard in terms of flammability limits and heat of combustion for each known and unknown compounds. The advantage of RF number over others such as *R*-index and *F*-number for classification of combustion hazard has been highlighted.

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1. Introduction

In order to cope with global environmental problems, chlorofluorocarbon (CFC) replacements are being developed. At the present stage, hydrofluorocarbons (HFCs) with short atmospheric lifetimes are promising as CFC alternatives, and partially hydro-fluorinated ethers (HFEs) are considered candidates for new generation CFC replacements. These compounds contain hydrogen atoms in the molecule, and some of them are flammable. If they are flammable, their use will be strictly controlled by public regulations. Therefore, the flammability study is crucial for the developers and users of alternative refrigerants.

There is no question that the flammability limit is a most widely used index for representing the flammability characteristics of gases and vapors. There is a large volume of

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flammability limits data [1–3]. However, it is not necessarily easy to understand the true flammability characteristics of individual gases solely from the data of flammability limits. For example, the flammability of methane, ethane, and propane are considered to be very similar to each other, but the values of flammability limits are quite different, i.e., methane 5.0–15.0, ethane 3.0–12.4 and propane 2.1–9.5.

Considering this situation, we have recently introduced an index called *F*-number to represent the flammability characteristics of a given gas in terms of one unique number [4]. It is defined by the following equation:

$$F = 1 - \left(\frac{L}{U} \right)^{0.5} \quad (1)$$

Here *U* is the upper flammability limit and *L* the lower flammability limit. *F*-number takes values ranging from zero to unity depending on the degree of flammability. It can be taken as a sort of normalized flammability range. *F*-number is convenient because it can be expressed by an empirical equation in terms of molecular parameters and can be used to predict the flammability limits of unknown compounds [4]. However, *F*-number is not much more than an alternative way of expressing the flammability range, and does not appropriately represent the total combustion hazard of flammable gases.

On the other hand, there is another index called *R*-index which is defined by the following equation [5]:

$$R = \frac{C_{st}}{L} \quad (2)$$

where C_{st} is the stoichiometric concentration and *L* the lower flammability limit. If both the numerator and denominator of the right-hand side of this equation are multiplied by the molar heat of combustion, the numerator represents the maximum heat per unit volume available for a particular fuel–air mixture, whereas the denominator represents the minimum heat per unit volume to maintain the flame propagation in this system. Therefore, it is expected that the stronger is the fuel, the larger the value of *R*-index. In this sense, *R*-index can be taken as another way of expressing the flammable range. It is not very appropriate to be used to make adequate assessment of the potential hazard of flammable gases either.

In this relevance, American Society of Heating, Refrigerating and Air-conditioning Engineers (ASHRAEs) is considering a criterion for classifying refrigerants according to the combustion hazard of refrigerants [6]. The criterion consists of two items. One is whether or not the lower flammability limit of substance is larger than 0.10 kg/m³. The other is whether the heat of combustion is smaller than 19,000 kJ/kg. The refrigerant is assigned to class 2 if both the two items are satisfied. On the other hand, if the lower flammability limit is smaller than 0.10 kg/m³, or if the heat of combustion is larger than 19,000, then the refrigerant is assigned to class 3. Similarly, International Organization for Standard (ISO) is also investigating the method for the classification of refrigerants [7]. Their criterion for the flammability are the same as ASHRAE except that ISO proposes the lower flammability limit of 3.5 vol.% instead of 0.10 kg/m³ for the first factor. The reason why they use the dual factor system for the criterion seems clear. The first factor is concerned with the ignition probability, and the second for the intensity factor. The problem here is whether the two factors have been chosen appropriately.

The purpose of the present paper is to develop a new combustion hazard index to assess adequately the total potential hazard of various flammable gases.

2. Results

The new index for representing the total expectancy of a combustion hazard should contain two factors; one is indicative of the ignition probability and the other is proportional to the potential magnitude of combustion hazard in case of ignition. The ignitability is strongly dependent on the property of substance, while the potential magnitude of a combustion hazard directly depends on the quantity of substance.

In the field of combustion science, the minimum ignition energy and the auto-ignition temperature are two typical quantities related to ignition and/or ignition probability. In order to cause ignition, we have to give an energy that is larger than a certain minimum value to a fuel–air mixture, or the temperature must be raised to a certain threshold limit value. However, it is necessary to cause ignition that the concentration of the mixture be within the flammability range. Otherwise, the heat given to the mixture is used to heat up the spot of the mixture and the combustion never occurs. In this sense, to satisfy the condition concerning the concentration is far more important than the minimum ignition energy or the auto-ignition temperature.

When a leakage occurs, a large concentration gradient is produced at the leak spot and flammable gas mixtures are produced. As a first approximation, the probability of ignition can be considered proportional to the width of the flammability range. On the other hand, it is also true that the smaller is the concentration of the lower flammability limit, the easier the flammable gas mixtures are produced in case of leakage. Therefore, the ignition probability directly depends on the lower flammability limit as well. It may be considered proportional to the reciprocal lower flammability limit at the first approximation. We can postulate that the total ignition probability can be considered proportional to the lower half of the flammability range divided by the lower flammability limit.

$$\frac{(UL)^{0.5} - L}{L} = \left[\left(\frac{U}{L} \right)^{0.5} - 1 \right] = \frac{F}{1 - F} \quad (3)$$

Here U and L are the flammability limits and F is given by Eq. (1).

There are various factors that are related to the potential magnitude of a combustion hazard, e.g., the burning velocity, the flame velocity, the maximum temperature, the maximum pressure as well as the speed of pressure rise if the system is enclosed. Each of these factors may affect certain aspects of the potential magnitude of combustion hazard. Among others, the burning velocity may be one of the most important. On the other hand, it should be noted that the heat of combustion is the one that essentially determines everything related to the combustion phenomenon. In this sense, the heat of combustion should be a better choice than any others including the burning velocity.

Because, the heat of combustion is always the factor of top priority to be considered in the combustion phenomenon, while the burning velocity is important when the gases are well mixed. Secondly, the maximum temperature and the pressure in case of enclosed

which are direct functions of the heat of combustion are at least as important as the speed of temperature and pressure rises (in case of enclosed system) which are direct functions of burning velocity. Thirdly, the burning velocity itself is a direct function of the flame temperature which is determined by the heat of combustion, and therefore, they are more or less correlated to each other. And finally, access to the data of heat of combustion is in general much easier than that of the burning velocity.

In case the data of heat of combustion is not available for new substances, the theoretical calculation can be utilized. The recent development of the computer and the computer software has enabled to predict pretty accurately the values of heat of formation for molecules of moderate sizes. For this purpose, such calculations as Gaussian-2 [8], G2MP2 [9], BAC/MP4 and BAC/MP2 [10] can be utilized. With these methods, it is by no means difficult to predict the heat of combustion as well as heat of formation with uncertainty of less than 10%, which is accurate enough to be used for the classification of combustion hazard of flammables.

In conclusion, the new index for potential combustion hazard of flammable gases can be defined as a product of the ignition probability defined in the above and the heat of combustion per unit mass. Namely,

$$\text{RF number} = \left[\left(\frac{U}{L} \right)^{0.5} - 1 \right] \frac{Q}{M} \quad (4)$$

where Q denotes the heat of combustion in kJ/mol and M the molecular weight of the flammable gas. It can also be expressed as a function of F -number given by Eq. (1)

$$\text{RF number} = \left[\frac{F}{1-F} \right] \frac{Q}{M} \quad (5)$$

Table 1 shows the values of RF number for typical flammable gases. The values for typical fuel gases like hydrocarbons as well as their oxygen and nitrogen derivatives fall between 30 and 150. The values of RF number for saturated hydrocarbons are around 50 or so. RF numbers of extremely strong fuels such as hydrogen, acetylene and silane extend to 200, 300 or more. On the other hand, RF numbers of heavily halogenated combustible gases are very small indeed, e.g., the RF number values of dichloro-ethylene and methyl chloride are 10 or less. Thus, it is noted that the RF number values are much more widely spread than those of F -number and is conveniently be used to assign various gases to different hazard classes.

Incidentally, some of the compounds listed in Table 1 are given safety classification number by ASHRAE [6]. According to ASHRAE, those for which there is no flame propagation are assigned to class 1, lower flammability to class 2, and higher flammability to class 3. In the table, the values of RF number for class 2 compounds ranges from 1.2 for CH_2Cl_2 to 19.3 for HCOOCH_3 . Similarly, the values of RF number for class 3 materials ranges from 39.5 for CH_4 to 401.5 for hydrogen. Thus, the ASHRAE classification of flammability is in perfect agreement with RF number. By using RF number, we may be able to classify materials even in more detail, e.g., those with RF number less than 30 are classified to weakly flammable, those from 30 to 150 as normally flammable, and those higher than 150 are strongly flammable. RF number is suitable to be used to control materials by quantity as well.

Table 1
The values of RF number and the related quantities for typical combustible gases

Chemical formula	Molecular mass	Refrigerant number	Safety group ^a	Flammability limits (vol.%)	Stoichiometric concentration (vol.%)	LFL (kg/m ³)	Heat of formation (kJ/mol)	Heat of combustion (kJ/mol)	Heat of combustion (kJ/kg)	F-number	R-index	RF number (kJ/g)
<i>Methane series</i>												
CH ₄	16.04	50	A3	4.9–15.7 ^b	9.48	0.032	-74.81 ^c	802	50000	0.46	1.94	39.5
CH ₃ F	34.03	41		6.8–20.3 ^b	12.26	0.095	-234 ^d	672	19700	0.42	1.80	14.4
CH ₃ Cl	50.49	40	B2	8.2–20.2 ^b	10.69	0.169	-80.83 ^c	647	12800	0.36	1.49	7.3
CH ₃ Br	94.94			10–16.0 ^b	10.69	0.388	-35.1 ^c	637	6700	0.21	1.23	1.8
CH ₂ F ₂	52.02	32	A2	13.3–29.3 ^b	17.32	0.283	-446.9 ^c	489	9400	0.33	1.30	4.6
CH ₂ ClF	68.48	31		14.4–26.5 ^b	14.35	0.403	-261.9 ^d	495	7200	0.26	1.20	2.6
CH ₂ Cl ₂	84.93	30	B2	14.1–20.8 ^b	12.26	0.489	-92.47 ^c	486	5700	0.18	1.23	1.2
<i>Ethane series</i>												
CH ₃ CH ₃	30.07	170	A3	3.0–12.5 ^c	5.65	0.037	-84.68 ^c	1428	47500	0.51	1.88	49.4
CH ₃ CH ₂ Cl	64.51			3.8–15.4 ^c	6.06	0.100	-112.17 ^c	1251	19400	0.50	1.72	19.6
CH ₃ CH ₂ Br	108.97			6.8–8.0 ^c	6.06	0.303	-64.52 ^c	1243	11400	0.08	0.96	1.0
CH ₃ CH ₂ F ₂	66.05	152a	A2	4.8–17.3 ^f	7.73	0.130	-478.2 ^c	1093	16500	0.47	1.61	14.9
CH ₃ CHCl ₂	98.96			5.6 ^c –13.2 ^b	6.53	0.227	-129.4 ^c	1084	11000	0.35	1.38	5.9
CH ₃ CF ₃	84.04	143a	A2	7.0–19.0 ^b	9.48	0.240	-736.4 ^c	864	10300	0.39	1.35	6.7
CH ₃ CClF ₂	100.50	142b	A2	7.8–16.8 ^f	8.52	0.320	-532 ^c	890	8900	0.32	1.22	4.1
CH ₃ CCl ₂ F	116.95	141b		9.0–15.4 ^b	7.73	0.430	-335 ^c	908	7800	0.24	1.05	2.4
CH ₃ CCl ₃	133.40			7.5–12.5 ^c	7.08	0.409	-142.30 ^d	922	6900	0.23	1.26	2.0
<i>Propane series</i>												
CH ₃ CH ₂ CH ₃	44.10	290	A3	2.1–9.5 ^c	4.02	0.038	-104.5 ^c	2043	46300	0.53	1.92	52.2
<i>Other hydrocarbons</i>												
CH ₃ CH ₂ CH ₂ CH ₃	58.12	600	A3	1.6–8.4 ^c	3.12	0.038	-126.5 ^c	2657	45700	0.56	1.95	59.0
CH(CH ₃) ₃	58.12	600a	A3	1.8–8.4 ^c	3.12	0.043	-134.5 ^c	2649	45600	0.54	1.73	52.9
CH ₃ (CH ₂) ₃ CH ₃	72.15			1.5–7.8 ^c	2.55	0.044	-146.5 ^c	3272	45300	0.56	1.70	58.1
CH(CH ₃) ₂ CH ₂ CH ₃	72.15			1.4–7.6 ^c	2.55	0.041	-153.8 ^c	3265	45300	0.57	1.82	60.2
<i>Cyclic organic compounds</i>												
-(CH ₂) ₃	70.13			1.5 ^c –6.9 ^c	2.72	0.043	-78.4 ^c	3098	44200	0.51	1.81	50.6
<i>Oxygen compounds</i>												
C ₂ H ₅ OH	46.07			3.3–19 ^c	6.53	0.062	-235.10 ^c	1277	27700	0.58	1.98	38.8
(CH ₂) ₃ CHOH	60.10			2.0–12.7 ^c	4.45	0.049	-272.5 ^c	1875	31200	0.60	2.22	47.4
CH ₃ OCH ₃	46.07			3.4–27.0 ^c	6.53	0.064	-184.05 ^d	1328	28800	0.65	1.92	52.4
CH ₃ OCF ₃	100.04			10.5–21.5 ^s	12.26	0.429	-843 ^c	757	7600	0.30	1.17	3.3
CH ₃ OC ₂ H ₅	60.10			2.0–10.1 ^c	4.45	0.049	-216.4 ^d	1931	32100	0.56	2.22	40.1
CH ₃ OC ₂ F ₅	150.05			10.5–13.5 ^s	9.48	0.644	-1310 ^c	950	6300	0.12	0.90	0.9
C ₂ H ₅ OC ₂ H ₅	74.12	610		1.9–36.0 ^c	3.37	0.058	-252.13 ^c	2531	34100	0.77	1.78	114.5
CH ₃ COC ₂ H ₅	72.11			1.4–11.4 ^c	3.67	0.041	-240.8 ^c	2301	31900	0.65	2.62	59.1

Table 1 (Continued)

Chemical formula	Molecular mass	Refrigerant number	Safety group ^a	Flammability limits (vol.%)	Stoichiometric concentration (vol.%)	LFL (kg/m ³)	Heat of formation (kJ/mol)	Heat of combustion (kJ/mol)	Heat of combustion (kJ/kg)	F-number	R-index	RF number (kJ/kg)
HCOOCH ₃	60.05	611	B2	4.5–23 ^b	9.48	0.110	-350.2 ^c	920	15300	0.56	2.11	19.3
<i>Nitrogen compounds</i>												
CH ₃ NH ₂	31.06	630		4.9–20.7 ^e	8.52	0.062	-22.97 ^e	975	31400	0.51	1.74	33.1
CH ₃ CH ₂ (NH ₂)	45.08	631		3.5–14.0 ^f	5.29	0.064	-47.15 ^f	1586	35200	0.50	1.51	35.2
<i>Unsaturated organic compounds</i>												
CH ₂ =CH ₂	28.05	1150	A3	2.7–36.0 ^g	6.53	0.031	52.26 ^g	1323	47200	0.73	2.42	125.0
CH ₂ =CHF	46.04			2.6–21.7 ^h	7.73	0.049	-140.11 ^d	1160	25200	0.65	2.97	47.6
CH ₂ =CHCl	62.50			3.6–33.0 ^e	7.08	0.092	35.6 ^e	1157	18500	0.67	2.15	37.5
CH ₂ =CF ₂	64.03			5.5–21.3 ^h	9.48	0.144	-328.9 ^f	1000	15600	0.49	1.72	15.1
CH ₂ =CCl ₂	96.94			6.5–15.5 ^h	7.73	0.258	2.43 ^e	974	10000	0.35	1.46	5.5
CHCl=CHCl	96.94			5.6–12.8 ^h	7.73	0.222	6.1 ^c	978	10100	0.34	1.69	5.2
CHCl=CCl ₂	131.39			8–10.5 ^h	8.52	0.430	-7.78 ^h	872	6600	0.13	1.19	1.0
CF ₂ =CF ₂	100.02			10.0–50.0 ^g	17.32	0.409	-650.6 ^g	668	6700	0.55	1.73	8.3
CF ₂ =CFCl	116.47			8.4–16.0 ^g	14.35	0.400	-555.2 ^g	630	5400	0.28	1.71	2.1
CH ₃ CH=CH ₂	42.08	1270	A3	2.0–11.1 ^f	4.45	0.034	20.2 ^f	1926	45800	0.58	2.22	62.1
C ₂ H ₂	26.04			2.5–100 ^h	7.73	0.027	226.73 ^h	1256	48200	0.84	3.09	256.8
<i>Inorganic compounds</i>												
H ₂	2.02	702	A3	4.0–75 ^e	29.6	0.0033	0.0	242	119800	0.77	7.40	401.5
NH ₃	17.03	717	B2	15–28 ^e	21.9	0.104	-46.11 ^e	317	18600	0.27	1.46	6.9
CO	28.01			12.5–74 ^e	29.6	0.143	-110.525 ^e	283	10100	0.59	2.37	14.5
SiH ₄	32.12			1.37 ^m –100 ⁿ	9.48	0.018	34.3 ^c	1429	44500	0.88	6.92	335.6

^a The classification is due to ASHRAE criteria: A is lower toxicity, B is higher toxicity, 1 is no flame propagation, 2 is lower flammability, and 3 is higher flammability.

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^g Calculated from the lower flammability limit together with the relationship of Eq. (8) in the test.

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ⁱ Obtained from ab initio MO calculation by BAC-G2MP2 method.

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^k Y. Urano, Private communication.

^l Obtained from ab initio MO calculation by BAC-MP2/6-37G** method.

^m S. Horiguchi, Y. Urano, K. Tokuhashi, M. Iwasaka, S. Kondo, J. High Press. Gas Safety Inst., Jpn. 24 (1987) 143.

ⁿ Assumed; see, S. Kondo et al., Combust. Flame 101 (1995) 170.

3. Discussion

3.1. RF number for blend refrigerants

There is no particular difficulty in determining the value of RF number for mixtures of several flammable gases. The measurement of the flammability limits can be done similarly to pure materials, and the heat of combustion is obtained by a molar weighted average. As for blend gases consisting of non-flammable and flammable components, the measurement of flammability limits can be done similarly, and the heat of combustion is obtained using the heat of oxidation reaction for non-flammable components.

3.2. Predictability of the values of RF number

Eq. (5) shows that RF number is given by a simple function of F -number multiplied by the heat of combustion per weight. The heat of combustion for blend gases can be calculated from ones for pure materials.

A previous paper showed that F -number is expressed by an analytical form in terms of chemical bonds and chemical groups which constitute the molecule, and its value can be predicted for almost any organic compounds [4]. For a blend gas of flammable components only, the values of flammability limits can be predicted from those of component gases. According to Le Chaterier's rule, the lower flammability limit of blend gas is given by the following equation:

$$\frac{1}{L} = \frac{c_1}{L_1} + \frac{c_2}{L_2} + \frac{c_3}{L_3} + \dots \quad (6)$$

Here, c_1 , c_2 and c_3 are the concentrations of component gases whose lower flammability limits are L_1 , L_2 and L_3 , respectively. The same equation is said to be valid for the upper flammability limit as well. The equation for Le Chaterier's rule can be expressed in terms of F -number. If several kinds of fuels whose F -number values are F_1 , F_2 , F_3 , ... are mixed together with concentrations c_1 , c_2 , c_3 , ..., F -number of the resulting mixture may be given as follows:

$$F = c_1 F_1 + c_2 F_2 + c_3 F_3 + \dots \quad (7)$$

For blend gases containing both flammable and non-flammable components, however, it is not easy to accurately predict the F -number value. One should rather determine its value experimentally.

3.3. Advantages and disadvantages of proposed indices

From a combustion point of view, the strongest mixture between fuel and air is the stoichiometric concentration. Ideally, the upper and lower flammability limits should be located at an equal distance from the stoichiometric concentration if it is scaled by the ratio of fuel to oxygen or oxygen to fuel. In reality, however, we have to consider the effect of selective diffusion on the flammability limits. It is known that both the upper and lower

flammability limits are shifted to higher concentrations due to this effect if the fuel molecule is heavier than oxygen molecule, while they are shifted to lower concentrations if the fuel molecule is lighter than oxygen molecule [11]. As a result, the stoichiometric concentration becomes lower than the geometric mean of both limits for fuels with heavy molecules and vice versa for light ones. In order to maintain the similarity condition among the key concentrations of flammability, one should utilize the geometric mean of both limits rather than the stoichiometric concentration. In a previous paper [4], we have found a relationship between the geometric mean of both limits and the stoichiometric concentration, which is expressed by the following equation:

$$(UL)^{0.5} = [1 + 0.00472(M - 32.0)]C_{st} \quad (8)$$

Here M is the molecular weight and C_{st} the stoichiometric concentration. From this equation, we can estimate the geometric mean of both limits from the stoichiometric concentration.

This discussion reminds us of R -index [5]. As stated, the numerator of Eq. (2) represents the maximum heat per unit volume available for a given fuel–air system while the denominator gives the minimum heat per unit volume that enables to maintain the combustion in air. Therefore, R -index is expected to take values larger than 1.0. In reality, however, the flammability limits are affected by the selective diffusion while the stoichiometric concentration is not. If the fuel molecule is heavier than oxygen molecule, the lower flammability limit is shifted to higher concentrations, and the value of the ratio may become smaller than the value without the selective diffusion and vice versa. In order to maintain the similarity condition, we should utilize the geometric mean of both limits rather than the stoichiometric concentration. In this sense, R -index should have been defined as the geometric mean divided by the lower limit.

On the other hand, since F -number is defined in terms of the geometric mean of both limits together with the lower limit, it is free from the effect of selective diffusion. The advantage of using F -number instead of R -index also exists in the fact that F -number takes values ranging from zero to unity, and is particularly suitable for comparison among various substances.

It should be noted, however, that the indices such as F -number and R -index do not directly consider the magnitude aspect of a combustion hazard. It is essential to take into account a quantitative aspect for the index that represents the total potential hazard of combustion, and that is the reason why RF number has been introduced in the present paper.

4. Conclusion

We have proposed a new index called as RF number for addressing the potential combustion hazard of refrigerants. The expression of combustion hazard of refrigerants in terms of RF number is convenient for it can be expressed by an empirical equation in terms of molecular parameters and represents the total expectancy of combustion hazard with respect to flammability limits for each compound. RF number can also be expressed as a simple function of F -number, denoted for flammable characteristics.

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