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Assessing levels of hydrogen cyanide in fire experiments using a generalized correlation

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Abstract
A generalized relationship between the normalized yields of carbon monoxide and hydrogen cyanide for nitrogen-containing materials has recently been derived. This correlation is used in the current study to analyze experimentally derived hydrogen cyanide data from three sets of fire tests. For a reduced-scale compartment fire test, the yields of hydrogen cyanide with varied equivalence ratios and the transient hydrogen cyanide concentrations are estimated; for a series of room–corridor sofa fire tests, the extremely high hydrogen cyanide level observed is demonstrated to be a realistic result and a hydrogen cyanide yield value of 0.047 g/g is suggested for this sofa in post-flashover fires for fire safety assessments; and finally, for a series of smoke chamber tests with polyurethane, possible causes for the failure to detect hydrogen cyanide are suggested.

Keywords
Fire toxicity, carbon monoxide, hydrogen cyanide, yield, flashover

Introduction

Carbon monoxide (CO) is regarded as the dominant toxicant leading to incapacitation and deaths in fires. This results in a wide range of experimental studies on

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the yields of CO from small-scale to full-scale fire tests [1–4]. It has been found that the yields of CO in the post-flashover fire stage are approximately 0.2 g/g with very little material dependency [5,6]. This finding provides practical guidance in assessing the measured CO data in fire tests. Hydrogen cyanide (HCN) is another significant toxic species in fires involving fuels containing the element nitrogen [7]. Experimental investigations show that in many cases HCN is the most toxicologically significant fire species [8]. Similar to CO, experimental studies on the yields of HCN have also been undertaken for various materials [8–11]. Data of this type relating to toxic gases are important in engineering calculations of available safe egress time in fire enclosures. Unlike for CO, a reference yield for HCN in flashover fires has not been observed in experimental studies. The lack of such a simple guidance on the yield of HCN implies that experimental studies on materials with unknown toxicity properties are necessary for fire safety assessments. Furthermore, the lack of a representative reference yield sometimes makes it difficult to assess the reliability of the measured HCN levels from fire tests.

It is therefore desirable to introduce a practical method for assessing experimental HCN data and a general way for estimating yields of HCN in flashover fires. Purser [11] has found that yields of HCN show similar relationships to equivalence ratios as those for CO. He also found that close correlations exist between the conversion efficiency of element N to HCN (normalized yields of HCN) and the conversion efficiency of C to CO (normalized yields of CO) for each investigated material. In a recent study [12], it has been shown that the correlation found by Purser can be linked by the stoichiometric oxygen to fuel mass ratio (SOFMR) of the material of interest. A general formula representing the correlation between the normalized yields of CO and HCN for various materials has been derived using SOFMR. With this formula, the yields or concentrations of either HCN or CO can be approximately determined from the other in toxicity assessments if only the data for one of the two species are measured in fire experiments.

This study is an application of the generalised relationship between the normalised yields of CO and HCN derived in [12]. These applications include (1) estimating the yields of HCN and HCN concentrations in fire experiments, in which only CO yields or concentrations are measured; (2) deriving a general formula for estimating the yields of HCN at flashover stage for various materials; (3) interpreting and analysing measured HCN data in experiments. The generalized relationship and the formulae for the HCN calculation are presented in the next section. Three sets of experimental data are then assessed using these formulae. Based on this assessment, a set of conclusions are then drawn.

Equations for the calculation of HCN

Denote y as the yield (g/g) of one species and let f represent its normalized yield. Equations (1) and (2) give the expressions for the normalized yields of CO and

HCN based on their maximum theoretical yields:

$$f_{CO} = y_{CO}/y_{CO}^{max} \quad (1)$$

$$f_{HCN} = y_{HCN}/y_{HCN}^{max} \quad (2)$$

A generalised relationship between f_{HCN} and f_{CO} is given in the earlier study [12] as:

$$f_{HCN} = (\mathrm{TANH}(3.8 \times (y_{SOFMR} - 1.7) - 2.0)/1.3 + 1.2) \times f_{CO} \quad (3)$$

where y_{SOFMR} is the stoichiometric oxygen to fuel mass ratio. Its value for a given material is calculated under an assumption that the combustion products are CO_2, H_2O, NO_2, HCl, HBr and SO_2. The equations used in the analysis presented in this study are the correlation of HCN and CO yields (see equation (4)); the HCN yield in flashover fires (see equation (5)), the HCN concentrations (see equation (6)) and the concentration ratio of HCN to CO (see equation (7)). All these calculations use the measured CO data from fire tests. Substituting equations (1) and (2) into (3) provides the yield of HCN as a function of CO yield as shown in equation (4)

$$y_{HCN} = (\mathrm{TANH}(3.8 \times (y_{SOFMR} - 1.7) - 2.0)/1.3 + 1.2) \times y_{CO} \times y_{HCN}^{max}/y_{CO}^{max} \quad (4)$$

Corresponding to the yield of 0.2 g/g for CO in the flashover stage, a likely yield of HCN under the same conditions can be calculated as shown in equation (5)

$$y_{HCN} = 0.2 \times (\mathrm{TANH}(3.8 \times (y_{SOFMR} - 1.7) - 2.0)/1.3 + 1.2) \times y_{HCN}^{max}/y_{CO}^{max} \quad (5)$$

As seen from equation (5), the yields of HCN in post-flashover fires depend on the SOFMR of the material and the maximum theoretical yield of HCN and CO, which are determined by the molecular structure of the material of interest. This is quite different from the yields of CO in post-flashover fires, which has very little dependence on material properties. While the relative toxicity of HCN compared to that of CO is dependent on both exposure time and gas concentration, HCN is more than an order of magnitude more toxic than CO [7]. Taking a relative toxicity of HCN as 35 times that of CO [13], a yield of 0.0057 g/g HCN is equivalent to the toxic potency contributions from CO corresponding to the CO yield of 0.2 g/g in post-flashover fires. Therefore, if a calculated yield from equation (5) is greater than 0.0057 g/g for a material, HCN contributes more toxic potency than CO from the burning of the material.

As seen in Reference [12], the concentrations of HCN can be estimated from measured CO concentrations with equation (6) below:

$$X_{HCN} = 1.037 \times X_{CO} \times y_{HCN}^{max} \times (\mathrm{TANH}(3.8 \times (y_{SOFMR} - 1.7) - 2.0)/1.3 + 1.2)/y_{CO}^{max} \quad (6)$$

From equation (6), the concentration ratio of HCN to CO is obtained by equation (7)

$$R_{\mathrm{HCN,CO}} = 1.037 \times y_{\mathrm{HCN}}^{\max} \times (\mathrm{TANH}(3.8 \times (y_{\mathrm{SOFMR}} - 1.7) - 2.0)/1.3 + 1.2)/y_{\mathrm{CO}}^{\max} \quad (7)$$

Equations (4)–(7) are all derived from equation (3), which has been validated with experimental data for 13 different materials in Reference [12]. These equations will be used to assess the toxic hazards in various fire tests in the next section.

HCN experimental data assessments

In this section, experimental data from three sets of fire tests with nitrogen-containing fuels are assessed. All fuels in these tests are mainly polyurethane (PU) foam but with different molecular structures. The first set of data is from a reduced-scale compartment fire test that did not involve the measurement of HCN concentrations. The second set of data is from a series of room–corridor fire tests with much higher observed HCN yields than those normally found in other experiments. The third set of data is from a series of smoke chamber fire tests in which the measurement of HCN was undertaken but surprisingly no HCN was detected.

Reduced-scale compartment test without HCN measurement

Gottuk experimentally studied the correlation between the yields of toxic gases and global equivalence ratios (GER) with a reduced-scale fire room [14]. This type of data is useful in fire modeling for predicting the generation and transport of toxic gases in enclosure fires [15]. However, rather than HCN, only the main toxic gas, CO was measured in the experiments. One of the tested materials is PU ($C_1H_{1.74}O_{0.323}N_{0.0698}$). The lack of HCN data will affect the accuracy in toxicity assessment for this material. As suggested in [12], one of the applications of equation (3) is to estimate the level of HCN in fire engineering assessments of toxicity, in which only CO is measured in fire experiments or predicted in complex CFD fire simulations. In the following analysis, the level of HCN in this test is simply approximated using equations (4)–(6).

Figure 1(a) shows the measured yields of CO as a function of GER. Based on the molecular structure of this PU, $C_1H_{1.74}O_{0.323}N_{0.0698}$, the maximum theoretical yields of CO and HCN and the SOFMR are 1.41 g/g, 0.095 g/g and 2.16, respectively. The calculated yields of HCN using equation (4) are depicted in Figure 1(b). As seen in Figure 1, the average of measured CO yields at the post-flashover stage, with GERs greater than 1.0, is 0.25 g/g. The average of the calculated yields of HCN at this stage is 0.017 g/g. As HCN is more toxic than CO by an order of magnitude, HCN contributes far more toxic potency than CO. Figure 2(a) shows the measured CO concentrations as a function of time and Figure 2(b) is the

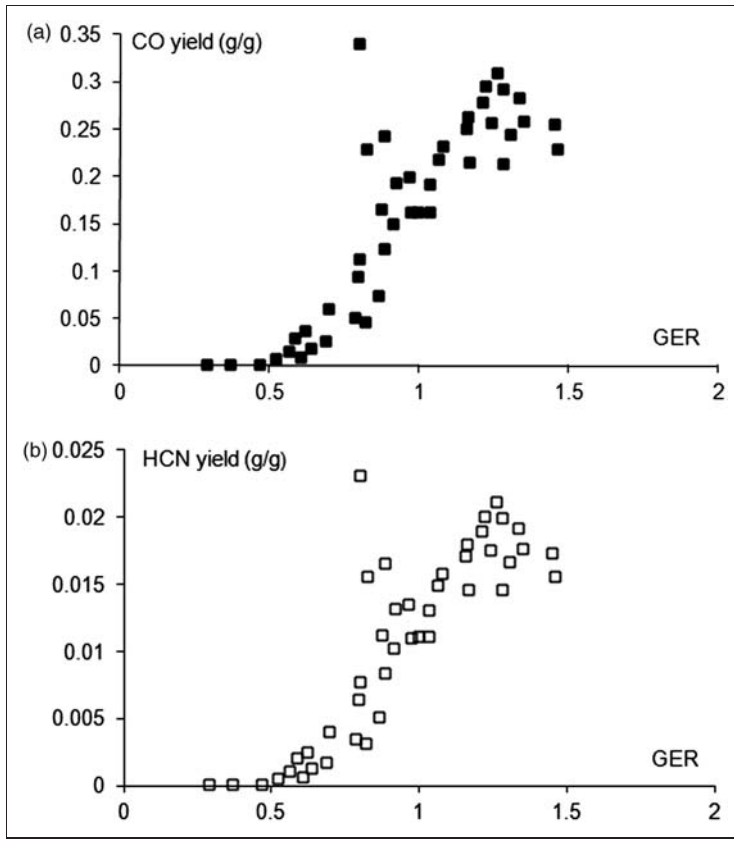

Figure 1. (a) Measured yields of carbon monoxide (CO) [14] and (b) calculated yields of hydrogen cyanide (HCN) as a function of global equivalence ratios (GER).

calculated HCN concentrations using equation (6). The measured CO concentrations reach 4% while the calculated HCN concentrations are as high as 0.28% between 300 and 400 s. Both these levels of CO and HCN can cause incapacitation within just a few seconds [7]. The HCN analysis in this case demonstrates that relationship (3) is a useful tool in toxic potency assessment for materials in which only the CO data are available from fire experiments.

Equation (3) can also be of assistance when planning experiments by determining whether it is necessary to measure the HCN concentrations for a given fuel. For example, 0.2 g/g is usually observed for the CO yield in the post-flashover stage of fire development. From equation (5), the estimated HCN yield of the material in the aforementioned experiment corresponding to this level of CO yield is 0.013 g/g, which is much higher than 0.0057 g/g, an indicator of equal toxic potency contributions by CO and HCN. This indicates that HCN measurements should be undertaken for this material to allow a full analysis of its toxic potency.

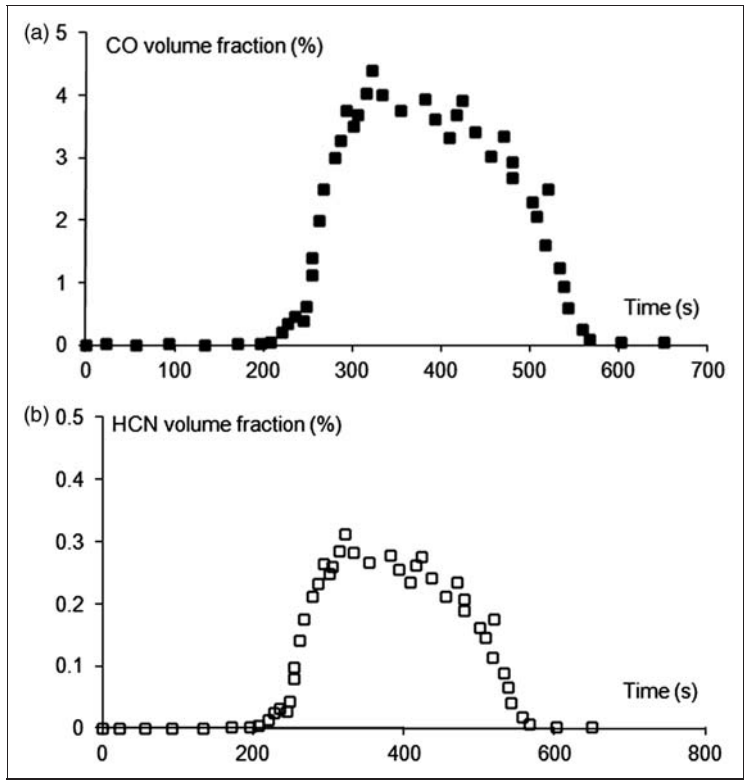

Figure 2. (a) Measured concentrations of carbon monoxide (CO) [14] and (b) calculated concentrations of hydrogen cyanide (HCN) as functions of time.

Room–corridor fire tests with high HCN yields

A series of fire tests were conducted in a room–corridor assembly shown schematically in Figure 3 [16,17]. The interior of the burn room was 2.44 m wide, 2.44 m high and 3.66 m long. The attached corridor was 9.75 m long and of the same width and height as the burn room. A doorway of 0.76 m wide and 2.0 m high was centered on the wall connecting the room and the corridor. The numbers 1–4 in Figure 3 indicate four gas sampling locations for the measurements of carbon dioxide (CO_2), CO and HCN, etc. Location 2 was on the central line of the corridor, approximately 0.3 m below the ceiling and at 1 m outside the burn room doorway. The concentrations and yields of toxic gases analyzed in this study are based on the measurements using Fourier transform infrared spectroscopy (FTIR) at location 2. Four tests with a sofa as the fuel, SW10–SW14, are investigated here. Approximately 80% of the sofa mass consisted of the PU cushion foam and the other 20% was the cushion fabric. The chemical compositions of the sofa and the PU foam are listed in Table 1.

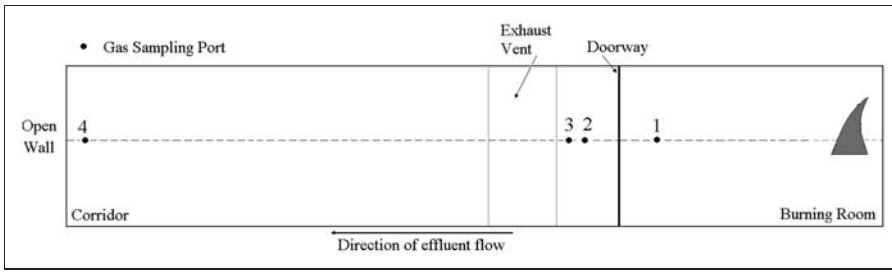

Figure 3. Schematic (plan view) of the room–corridor test fixture [16].

Table 1. Mass fractions of the elements in the materials[a]

	C	H	N	O	Cl	P
Sofa [16]	0.545	0.08	0.1	0.267	0.0068	0.0015
PU cushion foam [16]	0.5636	0.0853	0.1251	0.2156[a]	0.0085	0.0019
PU foam [18]	0.6475	0.0661	0.0636	0.2153	0.0137	0.0012

PU: polyurethane.
[a] $1 - \Sigma$ (mass) of listed elements.

The measured yields of CO and HCN based on the measurements at location 2 in all of the selected tests are shown in Table 2. Flashover was noted to have occurred in these selected fire scenarios. Due to the secondary burning of the combustion products after the gases exited the fire room, the yields of CO in post-flashover fires are much lower than the expected value of 0.2 g/g. On the other hand, the post-flashover concentrations of the measured HCN were much higher than those found in most other studies. To put the HCN measurements into perspective, Hirschler [6] compared them with those taken in a full-scale PU foam fire test carried out by NIST [18]. For the PU foam used in [18], it was found that the concentration ratio of HCN to CO was 0.068. By contrast, the average concentration ratio of HCN to CO found in the post-flashover fire tests with sofas in [16] and [17] is 0.34.

The unexpected low yields of CO in the fire tests (due to the noted secondary burning) resulted in a suggestion in [16] and [17] that the CO yield value of 0.2 g/g should be used, instead of the measured CO yields, in fire safety analysis for post-flashover fires. However, no indication was given as to the appropriate level of HCN, other than the measured data, that should be used in the same type of analysis. In the remainder of this section, equations (4)–(7) are used to assess the measured HCN data in [16] and [17].

Firstly, the unusually high level of HCN observed in the tests is analyzed. The calculated yields of HCN from equation (4) for tests SW10–SW14 are 0.003–0.0066 g/g (column 4 in Table 2), which are much lower than the corresponding measured data. It seems to corroborate Hirschler's claim that the measured HCN

Table 2. Yields of carbon monoxide (CO) and hydrogen cyanide (HCN)

	Species yields (g/g)			
	Measured CO [16]	Measured HCN [16]	Calculated HCN based on sofa composition	Calculated HCN based on PU cushion foam composition
SW10	0.0575	0.0186	0.0066	0.0135
SW11	0.0264	0.0084	0.0030	0.0062
SW12	0.0554	0.0172	0.0064	0.0130
SW13	0.0455	0.0166	0.0051	0.0104
SW14	0.0592	0.0164	0.0068	0.0139

PU: polyurethane.

data are unusually high. However, the calculations have not taken into account the burning behavior of the sofa in the fire tests. It was reported in [16] that the cushion fabric of the sofa was not generally burned away well before the PU foam was. Thus, perhaps only part of the covering was consumed by the fire in the early stages, exposing the PU foam which was then the main fuel that was burnt during the peak period of the fire. Taking into account this fire behavior and the large mass fraction of the cushion foam (80%), it is appropriate to assume that the main burning fuel at the peaks of these fires was the PU cushion foam. With this assumption, the calculated yields of HCN from equation (4) in these tests are between 0.0062 and 0.0139 g/g, which are close to the measured yield range of 0.0084–0.0172 g/g. Therefore, the high yields of HCN are not considered to be unrealistic as is suggested in [6].

In addition, from equation (6), the HCN concentrations from the sofa fires can be estimated from the measured CO concentrations. The comparisons between the measured and calculated HCN concentrations with the chemical composition of the PU cushion foam taken into account are presented in Figure 4. The calculated HCN concentrations in all of the five tests with sofas essentially follow the measured trends. The peak values are under-predicted by relative errors of 18%–40%. This demonstrates that the measured HCN concentrations are comparable to those calculated. It further substantiates that the unexpected high concentrations of HCN measured in [16] are not unreasonable.

The under-prediction of the peak HCN values may be caused by both the complexity of the composition of the PU foam considered and the simplicity of a model that is not capable of handling the complex nature of the material composition. The PU foam was described as a flexible polyurethane formulation containing melamine and a chlorinated phosphate ester fire retardant [16]. Melamine itself can contain as much as 66.7% nitrogen and the role of melamine in the combustion of the PU foam composite is not fully understood. Both the pure PU foam and

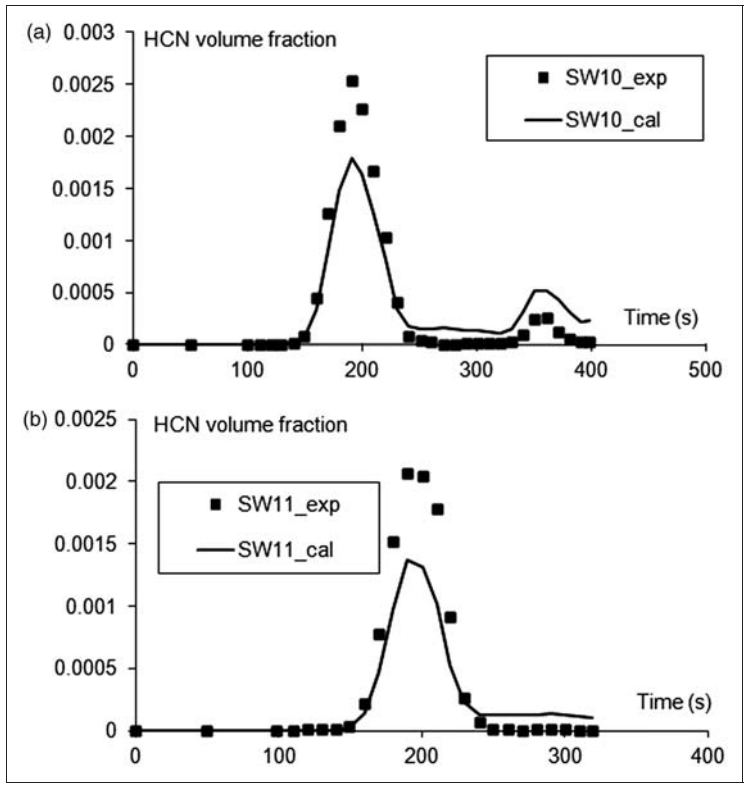

Figure 4. Measured [16] and calculated hydrogen cyanide (HCN) concentrations for the tests with sofas (a) SW10, (b) SW11, (c) SW12, (d) SW13 and (e) SW14.

melamine contribute to HCN generation, but they may have very different mechanisms of HCN formation when burnt. However, in the HCN calculation, the two materials are treated as a simple composite in which each nitrogen-containing component produces HCN in the same manner.

Secondly, the discrepancy between the ratios of HCN to CO in [16] and [18] observed by Hirschler is explained. From equation (7), the concentration ratios of HCN to CO are calculated. As seen in Figure 5, the estimated concentration ratios of HCN to CO are 0.087 for the PU foam in [18] and 0.244 for the cushion foam in [16], respectively, which represents a difference similar to the measured ratios in the two tests. Note that the estimation of the ratio of HCN to CO is determined only by the molecular composition of the material of interest. It indicates that the discrepancy in the two HCN to CO ratios is very likely caused by the difference in the element compositions of the cushion foam used in [16] and the PU foam used in [18]. As seen in Table 1, the element compositions of the two types of PU foams are quite different. The mass fraction of

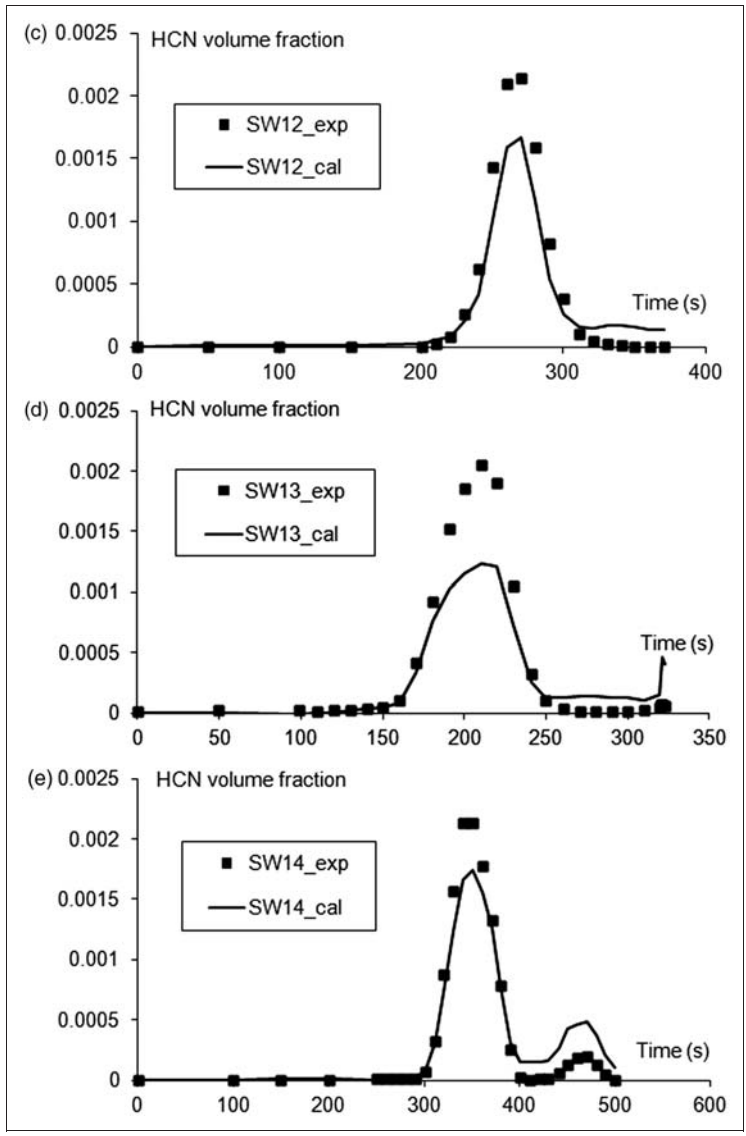

Figure 4. Continued.

N in the cushion foam is 0.1251, which is almost twice of that in the PU foam in [18]. Therefore, the discrepancy between the HCN to CO ratios of the two room-scale fire scenarios does not necessarily indicate that one is more reliable than the other.

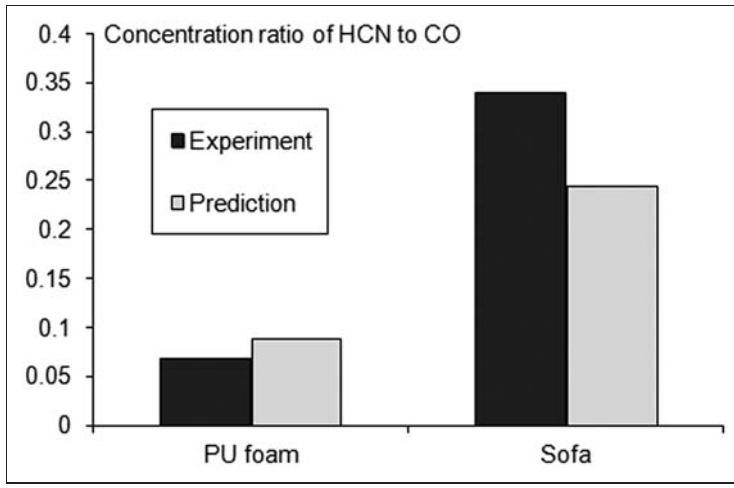

Figure 5. Measured and calculated concentration ratios of hydrogen cyanide (HCN) to carbon monoxide (CO).

Thirdly, an appropriate level of the HCN yield for the sofa in post-flashover fires is suggested for fire safety assessment. Corresponding to the expected yield of CO of 0.2 g/g in the post-flashover stage (approximately four times the value measured in the tests), the yield of HCN for this sofa with the chemical composition of the PU cushion foam is estimated to be 0.047 g/g using equation (5), which is approximately three times the value measured in the tests. The estimation, while very high, should not be surprising as it corresponds to the expected yield of CO. Furthermore, the estimated value is very close to 0.045 g/g, the average HCN yields in vitiated nylon fires under a temperature of 825°C [8]. The similarity of the HCN yields of the two different materials is not unexpected according to equation (5) because the investigated PU foam has a similar molecular composition to nylon. The HCN yield of a material can be estimated from the maximum theoretical yields of CO, HCN and the SOFMR for the material. These values for the cushion foam (1.487 g/g, 0.241 g/g and 2.254, respectively) are similar to those (1.315 g/g, 0.239 g/g and 2.263 respectively) for nylon.

Smoke density chamber test with undetected HCN

In an experimental study in [19], the generation of toxic products from burning a PU foam was measured in the NFPA/ISO smoke density chamber. The PU foam used in these small-scale experimental tests was the same one listed in Table 1 for the sofa mock ups in the full-scale fire tests in [16]. It was already known from the full-scale fire tests that high levels of HCN would be generated for this material (Figure 4). However, for these small-scale fire tests, "*Most surprisingly, there was no HCN detected in any of the polyurethane*

foam experiments" [19]. Those conducting the experiment stated that no satisfactory explanation was found for the failure of the detection of HCN in these tests and that further investigation for this failure would be expected. For a material like this PU foam with known HCN toxicity potency, the failure to detect HCN is readily noticeable. However, a failure of this kind for materials with a less well-known toxic fire hazard would lead to a significant underestimation of their toxic potency.

In this study, a possible cause for the failure to detect HCN in the smoke chamber tests is provided using the generalized relationship between HCN and CO. The measured peak concentrations of CO are no more than 190 ppm in all the PU experiments in [19]. Corresponding to the CO level of 190 ppm, the peak HCN concentration for this PU foam would be at most 46.3 ppm, estimated using equation (6). However, the instrument used for the measurement of toxic gases in the experiments in [19] had a minimum detection limit of 50 ppm for HCN, which represents the lowest concentration that could be measured. Therefore, the fact that the maximum of HCN concentrations might be less than 46.3 ppm indicates that the HCN levels from the burning of the PU foam would be too low to be detected with the instrument used in the tests.

From equation (7), the calculated concentration ratio of HCN to CO for this material is 0.244. As HCN is approximately 35 times more toxic than CO [7,13], HCN contributes much more toxic potency than CO does for this material. The undetected HCN in the tests in [19] does not imply that the presence of HCN for these polyurethane fires can be neglected. The use of equation (7) is helpful for deciding whether to measure HCN and for choosing the correct accuracy level of an instrument for such a measurement. This is especially of importance for testing new materials whose toxicity contribution by HCN is little known.

Conclusions

The generalized relationship between the normalized yields of HCN and CO derived in an earlier work has been used to assess three sets of fire test data in this study. It has been demonstrated that the derived relationships are of assistance in making reliable toxic hazard assessments for fires with fuels containing the element nitrogen by

- providing a reference for the material-dependent HCN yields in post-flashover fires, based on the guidance of a CO yield of 0.2 g/g for most materials;
- providing estimations of yields and concentrations of HCN if only the CO data are available;
- interpreting unexpected experimental results for certain materials, for example, extremely high or low HCN levels in fire experiments;
- determining the need to make HCN measurements and selecting instrumentation with appropriate accuracy levels for fire tests.

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