

# CO/CO<sub>2</sub> Ratios in Fire

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## ABSTRACT

The CO/CO<sub>2</sub> mol ratio has been used to estimate CO generation for hazard assessment in some room fire models. For better estimation of CO, more detailed usage of the CO/CO<sub>2</sub> ratio and more experimental data on their values seem required. CO/CO<sub>2</sub> mol ratios in the literature, measured using small-scale apparatuses and in large-scale experiments, were reviewed. The ratios for different materials at different fire conditions were experimentally measured using an oxygen depletion OSU apparatus and cone calorimeter. CO/CO<sub>2</sub> mol ratios depended on fuel types and fire conditions, but for a type of fuel in one mode of combustion, the ratio was fairly stable. It is suggested to apply different CO/CO<sub>2</sub> values to each fuel block for each stage of combustion in a fire modelling to estimate the CO generation.

**KEYWORDS:** CO/CO<sub>2</sub> mol ratio, CO generation, small-scale tests, cone calorimeter, OSU apparatus, pyrolysis, flaming, glowing

## INTRODUCTION

Generation of CO is the most significant factor for toxicity hazard assessment in building fires and is estimated as a time dependent variable in several computerized room fire models. As a basis for calculating CO generation, several models simply use constant values of CO conversion ratio[1,2,3] or constant values of CO/CO<sub>2</sub> ratio (or inverse)[4,5]. In the latter, multiplying the rate of generation of CO<sub>2</sub>, which is calculated from mass loss rate, the rate of generation of CO is obtained. One of the latest models[6] alternatively requires, as one of options, CO/CO<sub>2</sub> ratio as a time dependent variable supplied by the user of the model. In either case, more CO/CO<sub>2</sub> data are desirable for better estimation of CO generation. The CO/CO<sub>2</sub> ratio is relatively steady in one mode of combustion of a fuel. This is an advantage of using CO/CO<sub>2</sub> ratio for CO estimation. In this paper, applications of CO/CO<sub>2</sub> ratios for each fuel at different stages of combustion in hazard assessment is discussed. CO/CO<sub>2</sub> data

for such application can be produced by small-scale experiments with well defined fire conditions. CO/CO<sub>2</sub> data in the literature are reviewed for the purpose. New experimental data are produced using a cone calorimeter in order to fill the deficiency of existing data.

## BACKGROUND

The relationship between carbon dioxide/carbon monoxide ratio (CO<sub>2</sub>/CO) and oxygen concentration in fire was used to describe complex processes occurring during fire by Woolley and Fardel[7]. The method was adopted and expanded to classify various fires by ISO/TC92/SC3[8]. Recently, more authors prefer to report CO/CO<sub>2</sub> instead of its inverse. In a graphic presentation of data for high CO, which is important for hazard assessment, CO<sub>2</sub>/CO show poor graphic resolution. Also in relating to toxicity hazard, CO/CO<sub>2</sub> is about proportional to the hazard while CO<sub>2</sub>/CO is inversely proportional. In this paper, CO to CO<sub>2</sub> mol ratios are reported and expressed as CO/CO<sub>2</sub> unless otherwise stated. Whenever data in other expressions are referenced they have been recalculated to CO/CO<sub>2</sub> mol ratios throughout the paper for consistency. Some such data obtained from the literature are shown in Table 1[9-20].

TABLE 1 CO/CO<sub>2</sub> in small-scale tests

Apparatus	Fuel	Mode	CO/CO <sub>2</sub>	Reference
NIBS Test	6 materials	Flaming	0.047±0.017	Hirschler[9]
NBS Cup	Douglas fir	Flaming	0.092	Purser[10]
DIN Tube	Fibres	Flaming and non-flaming	0.17 - 0.6	Kallonen[11]
DIN Tube	Wood	Flaming and non-flaming	0.01 - 0.6	Prager[12]
Heated tube	PU	Pyrolysis	2.8, 18	Woolley & Wadley[13]
HRR apparatus	Various	Combustion	0.004-0.11	Tewarson[14]
Cone calorimeter	Various	Combustion	0.007- 0.05	Paul[15]
OSU apparatus	Plywood	Flaming and non-flaming	0.01 - 0.53	Tsuchiya, Mathieu[16]
UPITT	4 materials	Combustion	0.05 - 0.4	Grand[17]
Small chamber	4 liquids	Combustion	0.05 - 0.5	Morikawa[18]
Small chamber	Methanol	Combustion	0.09 - 0.9	Kim et al[19]
Fluidized bed	Peat	Pyrolysis	0.2 - 4	Arpiainen, Lappi[20]

Hirschler[9] stated that small scale tests yield too little CO and could not predict CO yields (CO/CO<sub>2</sub> also) in full-scale fires. He referenced NBS Cup furnace and NIBS test, both of which generated small CO/CO<sub>2</sub> values. DIN tube method yields large CO/CO<sub>2</sub> values but he does not think that it represents any stage of real fire.

Prager and others[12] modified the DIN procedure by increasing air flow rate up to threefold and showed that DIN tube method can simulate both smouldering and flaming fires. Purser stated that existing small scale test protocols do not cover the range of conditions found in full-scale fires, however, non-flaming condition is replicated by a number of small-scale test methods, and well-ventilated fire is replicated by NBS Cup[10]. He recommends the DIN tube method with modified protocols to simulate all types of fire.

In Woolley and Wadley's study[13], flexible polyurethane foams were pyrolysed in a heated tube. The highest CO/CO<sub>2</sub> values were 2.8 for a polyester foam and 18 for a polyether foam. Pyrolysis of oxygen containing polymers could yield high CO/CO<sub>2</sub> values because oxygen atoms in molecules may be insufficient to oxidize carbon atoms to CO<sub>2</sub>.

Grand[17] used a modified University of Pittsburgh Toxicity Test apparatus. Four materials were tested at two levels of heating rate and three levels of air flow rate. Effects of these experimental conditions on CO/CO<sub>2</sub> ratios were not obvious except for douglas fir which flamed at the higher heating rate but did not at the lower heating rate. These results may suggest that the CO/CO<sub>2</sub> value varies abruptly, rather than smoothly, with the mode of combustion.

In large scale fire experiments, CO/CO<sub>2</sub> ratios are reported or can be calculated from CO and CO<sub>2</sub> data. Some studies in the literature are listed in Table 2[21-27]. CO/CO<sub>2</sub> ratios can be related to air/fuel equivalence ratio or O<sub>2</sub> concentrations as in Table 2. Usually CO/CO<sub>2</sub> values increase by decreasing O<sub>2</sub> concentration.

TABLE 2 CO/CO<sub>2</sub> in large-scale tests

Fire Type	Fuel	CO/CO <sub>2</sub>	O <sub>2</sub> %	Reference
18 room fires	Fibreboard cribs	0.37±0.19		Gross,Robertson[21]
37 room fires	Wood cribs	0.35±0.19		Tewarson[22]
16 room fires	Cribs, panels etc.	0.03-0.63	19.5-3	Budnick[23]
Low-vent. fire	Wood	0.36	3.6	Woolley & Fardel[7]
A room fire	Wood	0.13	0.01	Portier et al[6]
A room fire	Wood, low & high ventilation	0.33, 0.07	4, 9	Purser[10]
A room fire	PU	0.07	5.8	Alpert et al[24]
1/2 scale house	Fuel oil	0.01-0.037	14-12	Kaya & Itaya[25]
Dormitory room	Furniture	0.05	9-6	Kim[26]
Two bldg.fires	Furniture	0.33, 0.53	3.1, 2.8	Morikawa et al[27]

## EXPERIMENTAL MEASUREMENTS OF CO/CO<sub>2</sub>

In reviewing data in the literature, it seemed necessary to measure CO/CO<sub>2</sub> values with clear ideas of which mode of fire is represented by the experimental conditions. Some of the data in the literature are averaged values in more than one modes of combustion. For some charforming polymers, CO/CO<sub>2</sub> values vary several hundred-fold by the mode of combustion as shown later. Averaged data for such materials did not seem useful for the present purpose. Values of CO/CO<sub>2</sub> in combustion of various fuels at different conditions of combustion were measured using a cone calorimeter. In the method, incident heat flux on the surface of a specimen can be defined. The author and a coworker previously measured CO generation from wood using an OSU apparatus under depleted O<sub>2</sub> atmosphere[16]. CO/CO<sub>2</sub> have been calculated from the previous experimental data. In the OSU apparatus, surface incident heat flux can be defined as in the cone calorimeter. DIN apparatus was not preferred mainly because of the restricted combustion in the narrow tube and also temperature instead of heat flux is defined in the procedures.

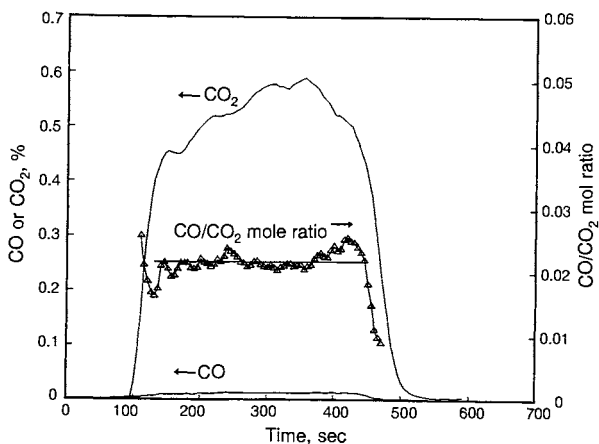


FIGURE 1 CO/CO<sub>2</sub> mol ratios in combustion of non-charforming polypropylene

TABLE 3 CO/CO<sub>2</sub> in combustion of liquids and non-charforming plastics

	kW/m <sup>2</sup>	CO/CO <sub>2</sub>
Methanol	5	0.0002
Ethanol	5	0.0004
i-Propanol	5	0.0020
s-Butanol	5	0.0025
n-Hexane	5	0.013
n-Heptane	5	0.010
Benzene	5	0.065
Xylene	5	0.056
Ethyleneglycol	30	0.0006
Polyethyleneoxide	30	0.0007
PMMA	30	0.0056
PMMA	50	0.0063
Polypropylene	30	0.024
Polypropylene	50	0.025
Polystyrene	25	0.046
Polystyrene	40	0.051
ABS	30	0.056
ABS	50	0.055

#### CO/CO<sub>2</sub> in Combustion of Liquids and Non-charforming Plastics

Concentrations of CO and CO<sub>2</sub> were measured during combustion under normal air in the cone calorimeter. In this condition, values of CO/CO<sub>2</sub> were almost constant during the major part of combustion for each fuel. An example on polypropylene is shown in Fig. 1. Results for various materials are summarized in Table 3. CO/CO<sub>2</sub> depended on the chemical

composition of fuel. Alcohols and oxygen containing fuels had low CO/CO<sub>2</sub>. Hydrocarbons had higher CO/CO<sub>2</sub> especially aromatics. Non-charforming polymers get molten surface and the melt behaves as liquid in combustion. Incident heat flux did not have significant effect on CO/CO<sub>2</sub> of non-charforming polymers. Under oxygen depleted conditions available experimental data on CO/CO<sub>2</sub> ratios are limited. By decreasing oxygen supplies, CO/CO<sub>2</sub> increased in Morikawa[18] and Kim and others[19] studies in which liquids were burnt in small chambers with varying levels of oxygen supply.

### CO/CO<sub>2</sub> of Wood Combustion

Fig. 2 (a) and (b) show CO, CO<sub>2</sub> and CO/CO<sub>2</sub> of combustion of 6 mm plywood in a cone calorimeter under 40 kW/m<sup>2</sup> incident heat flux. The CO/CO<sub>2</sub> varied in three phases during combustion. The 1st stage is pre-flaming pyrolysis, in which, the rate of generation of CO and CO<sub>2</sub> steadily increased by time. The rate of CO generation was the highest among the all three phases and also CO/CO<sub>2</sub> was the highest and nearly unity. The 2nd stage is flaming. As soon as the wood specimen ignited, CO/CO<sub>2</sub> rapidly fell to the lowest value of 0.002. The 3rd stage is char combustion or glowing, in which CO/CO<sub>2</sub> value was about 0.2. There was 500 fold difference in CO/CO<sub>2</sub> values at different stages in combustion of wood (one of charforming polymers).

These three stages in combustion of plywood were further examined in repeated experiments on 6 mm and 12 mm plywood under heat fluxes from 20 to 60 kW/m<sup>2</sup>. The effect of specimen thickness or incident heat flux levels was not significant. The results are presented in Fig. 3. Means of CO/CO<sub>2</sub> in each of the three stages were calculated and the results are presented in Table 4. Other charforming polymers also had these three stages of combustion.

TABLE 4 CO/CO<sub>2</sub> in combustion of wood

Mode	kW/m <sup>2</sup>	CO/CO <sub>2</sub>		
Pyrolysis	20-50	0.90	n=15	σ=0.17
Glowing	25-60	0.25	n=21	σ=0.072
Flaming	25-60	0.0035	n=23	σ=0.001

(a) Pre-flaming pyrolysis. When a plywood specimen was subjected to incident heat flux, both CO and CO<sub>2</sub> increased together as shown in Fig. 2 and when concentrations of pyrolysis products reached sufficiently high level flaming took place and rapidly CO<sub>2</sub> increased and CO decreased. This was true for the irradiance of 25 to 60 kW/m<sup>2</sup> that were experimented. At a lower irradiance, 20 kW/m<sup>2</sup>, concentrations of product gases were too low for flaming and CO/CO<sub>2</sub> was not determined accurately. There is an experimental result, that may be relevant to the present discussion, reported by Pitts and others[28]. In an 2/5 scale ISO/ASTM standard room, that was lined with plywood at the upper part, fire started with natural gas. Within 200-400 seconds into the test, CO vol% reached at 14% (one of the highest values ever observed in room fires) and soon later fell to a normal value

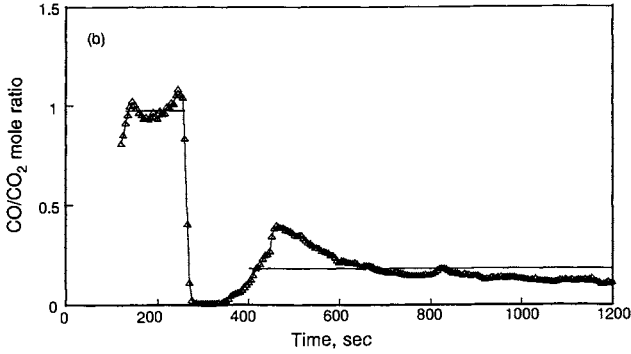
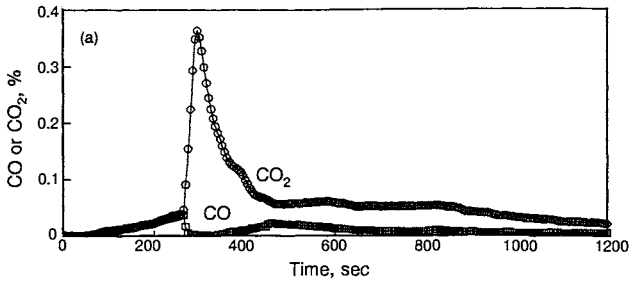


FIGURE 2 Variation of (a) CO% and CO<sub>2</sub>%, and (b) CO/CO<sub>2</sub> mol ratio in combustion of plywood under 40 kW/m<sup>2</sup> incident heat flux

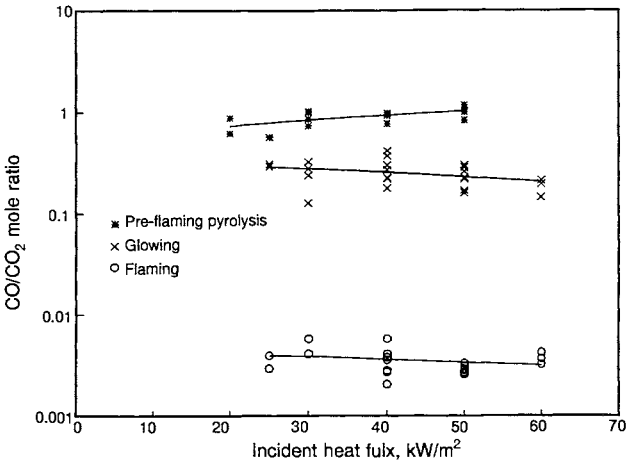


FIGURE 3 CO/CO<sub>2</sub> mol ratios in three stages of wood combustion

of about 2%.

(b) Flaming and non-flaming. In flaming of wood, generation of CO was minimal and the value of CO/CO<sub>2</sub> was several 100s fold smaller than that in the pre-flaming pyrolysis and 50 to 100 fold smaller than that in glowing combustion in the cone calorimeter experiments. In the previous experiment using an OSU apparatus with depleted oxygen atmosphere[16], CO generation or the value of CO/CO<sub>2</sub> varied 50 fold abruptly at the point where flaming/non-flaming transition occurred. The transition mainly depended on the oxygen concentration and slightly depended on the incident heat flux. The flaming occurred when O<sub>2</sub>% was higher than that shown by the following empirical equation:

$$(O_2, \%) = 19.4 - 0.19 \times (\text{Heat flux, kW/m}^2). \quad (1)$$

In large-scale room fire experiments, CO generation or values of CO/CO<sub>2</sub> increases with decreasing O<sub>2</sub> concentration. The change is not abrupt because the reported data are averaged values in the room. Each fuel block distributed in the room, may be experiencing different stages of combustion (and having different values of CO/CO<sub>2</sub>) at one time point. The proportion of the fuel involved in different modes of combustion may depend on location, time, temperature and O<sub>2</sub> depletion.

(c) Glowing. The glowing combustion of wood occurs in the later stage of combustion when remaining volatile products are insufficient to sustain flaming combustion. This is different from non-flaming discussed above and not related to air supply. In the free combustion in the cone calorimeter, air supply is unrestricted, but glowing combustion always occurred. The transition from flaming to glowing was obvious by observing the combustion or from slopes of mass loss -time curve, or by the shape of heat release curve. It occurred at a certain proportion of mass loss. Fig. 4 shows the percent of mass remaining at the transition for the combustion of fir plywood.

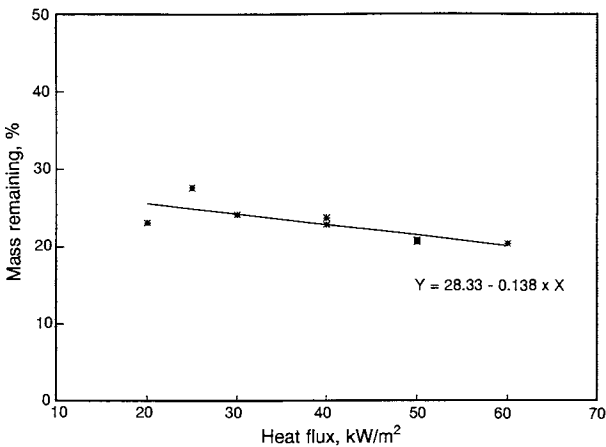


FIGURE 4 Mass remaining at the transition from flaming to glowing

## CO/CO<sub>2</sub> in Oxidative Pyrolysis of Polymers

TABLE 5 CO/CO<sub>2</sub> in oxidative pyrolysis of polymers

	kW/m <sup>2</sup>	CO/CO <sub>2</sub>		
PMMA	20-30	not measurable		
Polypropylene	30	not measurable		
Nylon	40	0.09		
FRP	50	0.61		
Polyethylene oxide	20	0.60		
PVC	30	0.31		
Wool	30	0.11	n=2	σ=0.02
Phenolic resin	60	0.14		
Wood	20-50	0.9	n=15	σ=0.17

Table 5 shows CO/CO<sub>2</sub> in oxidative pyrolysis of polymers. Values are generally larger than in other modes of combustion except for wool.

## CO/CO<sub>2</sub> in Combustion of Charforming Polymers

Table 6 shows CO/CO<sub>2</sub> in combustion of charforming polymers. FRP and Nylon 6 are pseud-charforming in a sense that their chars did not persist under incident heat flux. Wood is shown here again for convenience of comparison. Three polymers had distinct glowing phase of combustion in which CO/CO<sub>2</sub> were much higher than in flaming.

TABLE 6 CO/CO<sub>2</sub> in combustion of charforming polymers

	Mode	kW/m <sup>2</sup>	CO/CO <sub>2</sub>		
Nylon 6	Flaming	40	0.01	n=2	not stable
FRP	Flaming	50	0.020	n=3	σ=0.0018
PVC	Flaming	50	0.28	n=2	σ=0.014
Wool	Flaming	50	0.37	n=2	σ=0.14
	Glowing	50	2.2	n=2	σ=1.1
Phenolic resin	Flaming	60	0.015		
	Glowing	50	0.11	n=2	σ=0.008
Wood	Flaming	25-60	0.0035	n=23	σ=0.001
	Glowing	25-60	0.25	n=21	σ=0.072

## USE OF THE CO/CO<sub>2</sub> RATIO FOR HAZARD ASSESSMENT

Before discussing the use of CO/CO<sub>2</sub> in hazard assessment, usages of CO/CO<sub>2</sub> in existing major fire models are briefly reviewed. In FIRST (Mitler and Rockett[1] 1987) CO/fuel and CO<sub>2</sub>/fuel mass ratios are input as constants. An example for PU foam shows



that the ratios are 1.504 for  $\text{CO}_2$  and 0.0133 for CO respectively. Then  $\text{CO}/\text{CO}_2=0.0139$ . In another example for PMMA,  $\text{CO}/\text{CO}_2=0.0082$ .

In BRI2 (Tanaka and Nakamura[2] 1989), a fixed percent of fuel carbon is assumed to be converted to CO. The default value for wood ( $\text{CH}_{1.6}\text{O}_{0.71}$ ) is 0.005 and for urethane foam ( $\text{CH}_{1.8}\text{O}_{0.3}\text{N}_{0.06}$ ), 0.01. These values may be relevant to the flaming phase of well ventilated fires only.

In Hazard I, yields of CO and  $\text{CO}_2$  for post flashover for an enclosure fire of wood burning are respectively 0.3 and 1.1 g/g fuel. The values correspond to  $\text{CO}/\text{CO}_2=0.43$ . These values were recommended by Mulholland[3]. In FPETOOL (Nelson[4] 1990), two levels of  $\text{CO}/\text{CO}_2$  ratio, one in the lower smoke layer and the other in the higher smoke layer, are assumed. When there is an excess of oxygen, the ratio is 0.005 and insufficient oxygen, 0.5. These values are based on Mulholland's study[29].

In the NRCC model (Takeda and Yung[6] 1992) for polyurethane fires, CO concentrations in flaming fire and in smouldering fire are calculated separately. In flaming fires, the mass ratio of  $\text{CO}_2$  to CO is assumed to have an empirical linear relationship with the weight concentration of oxygen. In smouldering fires, quantities of CO and  $\text{CO}_2$  are considered proportional to the quantity of total products of combustion (CO,  $\text{CO}_2$  and  $\text{H}_2\text{O}$ ). In the present author's expression,  $\text{CO}/\text{CO}_2 = 0.602/Y_{\text{O}_2}$ , for flaming, where  $Y_{\text{O}_2}$  is the weight percent of  $\text{O}_2$ . In normal air,  $\text{CO}/\text{CO}_2 = 0.026$  (flaming). In smouldering,  $\text{CO}/\text{CO}_2 = 0.140$ .

In CFAST ver. 1.6 (Portier and others[7] 1992), the mass ratio CO to  $\text{CO}_2$ , among other time dependent variables, can be specified for each object being burned. This provides flexibility but the user of the program is responsible for supplying that information.

In room fire models, estimated mass loss rate is often taken as the basis for calculation. Oxygen consumption rate may be an alternative. A hazard assessment calculations may follow the steps:

1. mass loss rate or oxygen consumption rate
2. rate of  $\text{CO}_2$  formation
3. rate of CO formation using empirical  $\text{CO}/\text{CO}_2$  values or equations
4. toxicity hazard assessment using estimated CO

Fuels in a room or rooms are divided into blocks by the location and chemical composition. This is already performed in some models[2,7]. Each block go through three stages of fire namely pyrolysis, flaming and glowing. For each stage different  $\text{CO}/\text{CO}_2$  values are applied. The pyrolysis stage is not neglected because charforming polymers could generate the highest rate of CO in this stage. For transition from pyrolysis to flaming, i.e. ignition, a requirement is a sufficiently high concentration of combustible gases near the surface of fuel. Other factors are the  $\text{O}_2$  concentration and availability of an ignition source. The last one is the most unpredictable; a sort of assumption is necessary in modelling.

The 2nd stage, flaming, requires sufficiently high concentrations of combustible gases and ignition source. In addition,  $\text{O}_2$  concentration and incident heat flux are other two factors and depending on these two, flaming transfers to non-flaming. This is well

understood but empirical data are insufficient. It is seriously needed to produce data on the conditions of transition from flaming to non-flaming and CO/CO<sub>2</sub> ratio of various fuels using a small-scale apparatus with well defined heat flux level and O<sub>2</sub> concentrations such as the OSU apparatus or the enclosed cone calorimeter.

Transition from flaming to glowing for charforming polymers is a relatively simple phenomenon, yet experimental data are lacking for various fuels. CO/CO<sub>2</sub> values in glowing combustion of charforming polymers may be applied in non-flaming combustion discussed above. Values obtained on non-flaming of wood in the previous study[16] were similar to data on glowing obtained in this study.

Whether or not the CO/CO<sub>2</sub> ratio changes after leaving burn room while fire gases are still hot is an important question relating to toxicity hazard of adjacent or remote room in the same building. The change could be a decrease by oxidation of CO to CO<sub>2</sub> or an increase by oxidation of unburned hydrocarbons to CO. There are indications of unchanging ratio by Mulholland[3].

In this paper discussion was focused only on CO which is the primary cause of toxicity hazard. Discussion on HCN may be added as a next step in the similar methodology as CO. Discussion on HCl may be much simpler in the generation process but removal process by sorption would add difficulty. Other toxicity components such as oxides of nitrogen and sulphur and organic components are at secondary importance.

The major efforts in toxicity hazard assessment up to present time were directed towards establishing fire models and animal models (such as efforts of ISO/TC92/SC3). Using a fire model that best represents full-scale fire, materials are burnt and combustion products are applied to an animal model (or chemical analysis is used as a substitute for the animal model). Many fire models have been devised, but most people agree that any single model would not successfully represent all phases of a real fire. The approach described in this paper is to measure a hazard assessment parameter, that is CO/CO<sub>2</sub> mol ratio in this paper, using a test method that represent one phase of fire. Values of the parameter in different phases for different materials are separately measured and will be input into a computer room fire model to estimate the hazard. This approach may be called the digital approach as opposed to the analog approach of most other methods. The digital approach is still in its infancy and present room fire modelling programs are not ready to fully utilize this method and experimental data for input into the program are insufficient. The deficiency will hopefully be corrected by generating more data by small scale test, improving computer models, and checking validity of this approach by comparing with full-scale fire tests.

## CONCLUSION

To estimate generation of CO for hazard assessment in room fires, CO/CO<sub>2</sub> mol ratios can be used effectively since the ratio often showed a steady value within a mode of combustion of a material. The ratios for non-charforming fuels largely depended on the chemical composition. Charforming fuels generally went through three stages of combustion, namely, oxidative pyrolysis, flaming (and non-flaming which is mainly caused by low O<sub>2</sub> concentration), and glowing. In the oxidative pyrolysis, CO generation rate and CO/CO<sub>2</sub> mol

ratio were the highest of the three stages and the mol ratio was some 100s fold higher than in flaming. The CO/CO<sub>2</sub> mol ratio in glowing was the 2nd high. In estimating CO generation, it is suggested to use the following methodology. Fuels are divided into blocks by the location in a room or rooms and chemical composition and each block undergoes the three stages of combustion separately and appropriate CO/CO<sub>2</sub> mol ratios are applied for each stage. Some of CO/CO<sub>2</sub> values were presented in this paper for such usage. More data are required for better estimation of CO generation. Such data can be produced using a small-scale apparatus with well defined O<sub>2</sub> concentrations and incident heat fluxes.

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