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The Effects of Hydrogen Fluoride on the Wooden Surface of Historic Buildings during Fire Suppression Using Fluorinated Chemical Gases

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ABSTRACT

Chinese historic buildings are characterized by a wooden structure, which has caused serious results due to the ravages of frequent fire disasters. After the weathering by the natural environment, the fire risk of wood is highly enlarged. Fluorinated chemical gases are widely used for fire suppression at the early stage of a historic building fire. However, the effects of hydrogen fluoride (HF) released during fire suppression on historic buildings are unavailable. Presently, experiments were performed to study HF effects on the weathered timbers. Five traditional fluorinated chemical gases, H-37, FK-5-1-12, H-1323, H-2402, and H-1301, are employed to suppress a fixed flame. The weathered timber chips, acting as historic building materials, are located in the chamber to detail the influence of HF on the surface. The characteristic of gas production and elements variety of wood surface are discussed by FTIR and SEM-EDX, respectively. It is observed that flame enhancement takes place at the early stage of fire suppression and various types of fire agents. The total amount of HF during 50 min is highly dependent on the physical parameters of fluorinated chemical gases. The SEM results of weathered timber suggest the surface of the wood is porous, resulting in a strong gas absorption performance. The amount of deposited F on the wood surface is found to be positively correlated with the total amount of HF yielded during fire suppression.

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Corrosiveness; fire suppression; hydrogen fluoride; weathered wood; wooden culture heritage

1. Introduction

Historical buildings are an important cultural heritage since the potential historical and cultural values are precious and non-renewable (An and Wang 2016). Unfortunately, many of the historic buildings in the world had been destroyed due to the ravages of fire disasters (Li et al. 2018; Xiaomeng, Biao, and Xiang 2010; Zhou, Zhou, and Chao 2012). For example, the serious fire that has destroyed much of the timber roof and toppled the Gothic spire of the Notre Dame Cathedral took place on 16 April 2019 (Ferreira 2019). The National Museum of Brazil was ravaged by a huge fire on the night of 2 September 2018 (Domingues da Silva 2019). In Japan, a serious fire destroyed all seven main structures of the Shuri Castle World Heritage site on 31 October 2019 (Huang 2020). The fire protection of the historic building is the emphasis of the current stage in the world. It is known to all that China, an ancient country of historical civilization, has retained many historic buildings in the long historical process (Yin and Yamamoto 2013). These days, a great many historic buildings are recognized and recorded in the Chinese official database to strengthen the importance of fire

protection. The damage of historic buildings by fire is devastating in China (Zhou et al. 2019) since it is wooden. This is different from other countries. After the wooden structures exposing to the natural weathering process for a long time, it is much easier to be ignited because of the serious deterioration. The fire extinguishing agents used to put out historic building fires at the early stage of fire are limited. Because the additional damage to historic buildings during fire suppression should be avoided. Although the fire-extinguishing performance of water-mist in historical buildings has been reported (Xiaomeng, Biao, and Xiang 2010), the clean chemical gas agents are needed. The fluorinated chemical gas agents have been widely used in the field of many places, where the cleanliness is necessary, such as super-computer room, historic building and so on (Sheinson, Penner-Hahn, and Indritz 1989). Nowadays, chemical gas has been employed for fire suppression in the field of historic buildings (Piotr and Artur 2017). However, there are still many uncertain issues available. The hydrogen fluoride (HF) released during fire suppression process using fluorinated chemical gas is believed to impact a harmfulness and secondary damages because of corrosiveness.

There have been many attempts in the literature to develop approaches that address the HF generation mechanism during fire suppression. The main emphasis of available HF generation researches is focused on the HF yield mechanism of CF_3H gas (Zhou and Zhou 2011), amount of HF production (Zhou et al. 2011), thermal decomposition properties, and mechanism of HFC-125 (Wang et al. 2016, 2017), thermal decomposition of other Halon alternatives (Yamamoto et al. 1997), HF production using 2-H-Heptafluoropropane (Fu et al. 2011), thermal decomposition of trans-1,1,1,4,4,4-Hexafluoro-2-butene (Zhang et al. 2020), pyrolysis production of HF by difluoromethane (Xue et al. 2013), thermal decomposition of 2-Bromo-3, 3, 3-trifluoropropene (Zhou et al. 2013) and so on. Although the yield of acid gas has been reported to show potential damage on the surface of materials (Zierold and Chauviere 2012b), the comprehensive study of the influence of HF gas released during the fire suppression process on the wooden materials is still beyond the current research yield. Concerning Chinese historic buildings, it is characterized as a wooden structure. In general, most of the previous works were just focused on the mechanism of HF generation from the chemical aspects but not the influence of HF on the Chinese historic buildings. However, a deep understanding of the HF effects on weathered woods is of great necessity since the fluorinated chemical gas is being highly used in the fire protection of Chinese historic buildings. The basic knowledge about it should be conducted before the engineer application.

In this contribution, to achieve a detailed understanding of HF influence on the weathered timber samples, a series of tests are conducted. Authors detail works involve the HF concentration varying releasing time, explore HF generation differing the types of fluorinated chemical gas, and discuss the HF deposited on the weathered timber surface.

2. Experiment and method

2.1. Fire suppression test

The experiment method was based on Figure 1. The fire test facility consisted of a chamber (size in $L \times W \times H = 1.0 \text{ m} \times 1.0 \text{ m} \times 1.0 \text{ m}$), a window opening (size in $L \times W = 1.0 \text{ m} \times 1.0 \text{ m}$), a pool plate (diameter = 0.18 m), a sample plate (diameter = 0.20 m), specimens (weathered timber chips), a chemical gas tank (2 L) and an FTIR (Fourier Transform infrared spectroscopy) gas analyzer (MBGAS-3000). A sketch model of the test is detailed in Figure 1a. The chamber had an inner

surface area of 6.0 m^2 , just as shown in Figure 1b. The fuel of pool fire is gasoline, which is used to provide a fire source for suppression. In the plate, a little water is added before the addition of fuels to control a uniform combustion speed. To avoid the influence of dust, a little water is also added to the chamber. The FTIR gas analyzer includes an MBGAS-3000 and a gas sampling apparatus (see Figure 1c). During the whole test, the gas sampling tube is heated to keep 200°C . The smoke is firstly treated by an adsorption facility and then goes through the gas cell of FTIR. Before the test, calibration of the whole test is conducted. Then the pump of FTIR is used to exhaust the air from the chamber for 2 hours.

The fire extinguishing concentration (V/V %) used in the current work of H-2402, H-1323, H-1301, H-37 and FK-5-1-12 are 5.00%, 3.64%, 5.00%, 8.04% and 5.30%, respectively. The concentration of each fire extinguishing agent is calculated according to the minimum fire extinguishing concentration. The releasing press is 2.0 Mpa. To clarify the effect of hydrogen fluoride released during fire suppression on the historic buildings, a series of weathered cedar chips are used. They are placed on a sample plate. The physical information of cedar could be found in a previous art (Zhou et al. 2019). Description of the accelerated weathering tests of cedar chips has been reported previously (Zhou et al. 2021). The width of the sample is about 2 cm. A camera (Canon Rebel T3i 1080P) is used to record the fire suppression images at 30 frames per second with a spatial resolution of 0.47 mm/pixel. All tests are performed under a hood without ventilation. Ambient temperature and moisture are $19\text{--}24^\circ\text{C}$ and $15\text{--}36\%$, respectively.

2.2. Fluorinated chemical gases

The H-2402 ($\text{C}_2\text{F}_4\text{Br}_2$, CAS No.124-73-2, 99.7%), H-1323 ($\text{C}_3\text{H}_2\text{BrF}_3$, CAS No.1514-82-5, 99.7%), H-1301 (CBrF_3 , CAS No.75-63-8, 99.7%), and H-37 (C_3HF_7 , CAS No.431-89-0, 99.7%) were obtained from Jiujiang Middle Ship Chang'an Fire Equipment Co., Ltd. FK-5-1-12 ($\text{C}_6\text{F}_{12}\text{O}$, CAS No. 756-13-8, 99.7%) was phased from Sinochem Lantian Co., Ltd.

2.3. Test facilities

An MBGAS-3000 FT-IR Gas Analyzer bought from ABB is used. It is sized in a dimension 46 cm (W) \times 70 cm (H) \times 27 cm (D). The sample cell made of aluminum is a 1-liter volume (temperature to 200°C) and with resistant mirrors. It is equipped with a deuterated triglycine sulfate detector (DTGS) detector. The spectral range is $500 \text{ cm}^{-1}\text{--}4500 \text{ cm}^{-1}$. The pump exhaust speed is about 10 L/min.

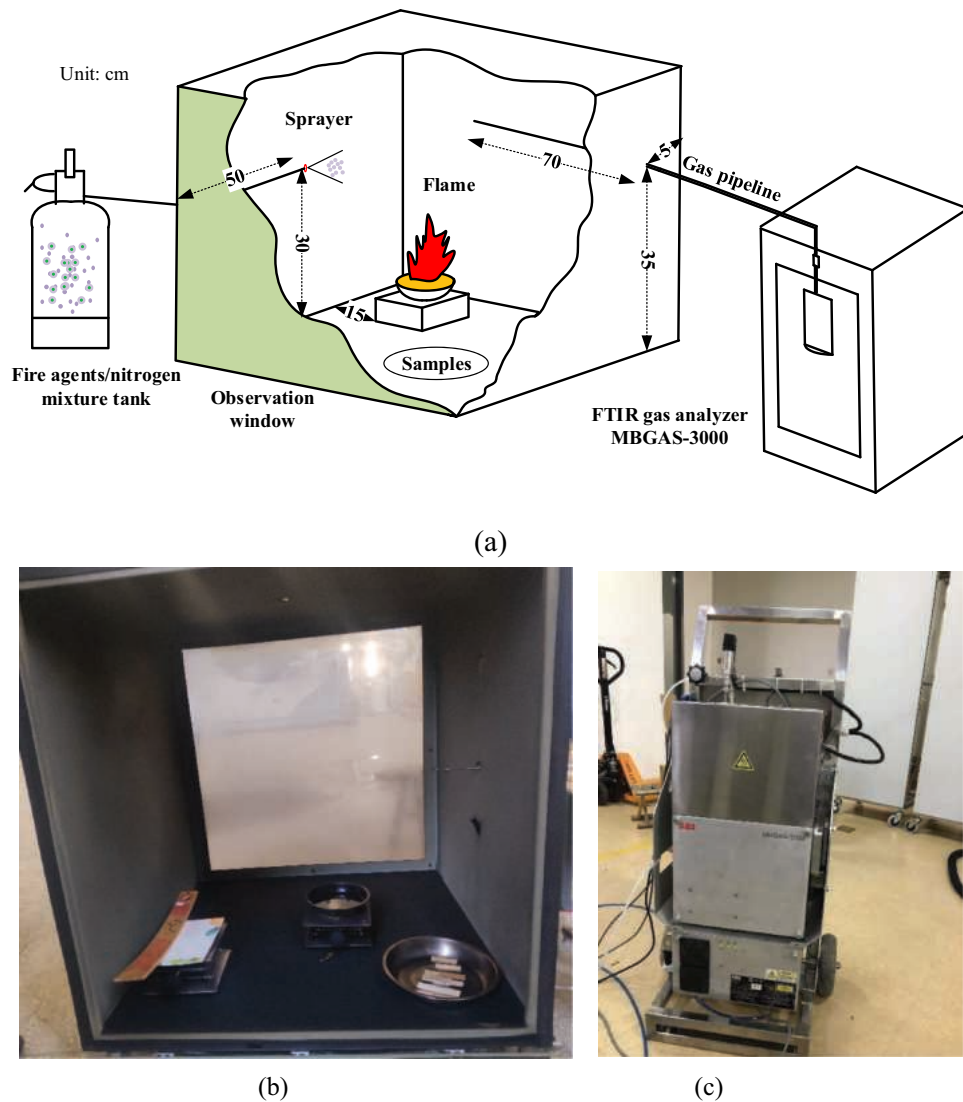


Figure 1. Experimental apparatus description. (a) A sketch model. (b) The experimental apparatus. (c) MBGAS-3000.

The surface morphology of the weathered timber was observed with an FEI Quanta 650 FEG SEM at 30 kV. The EDX analysis is also conducted on it.

3. Results and discussion

3.1. The description of fire suppression using fluorinated chemical gas

In the current work, five typical fluorinated chemical gases are employed to inhibit a fixed fire. It includes H-2402, H-1323, H-1301, H-37, and FK-5-1-12. From the observation of tests, it is found that the fire suppression time of the above five typical chemical gases is 9 s, 9 s, 9 s, 12 s, and 9 s, respectively. The low fire suppression efficiency of H-37 is partly attributed to hydrogen contained in the molecular (Yang, Liu, and Wu 2012). The

description of fire suppression performed by H-37 is shown in Figure 2. After the pool fire keeping for 10 s (shown in Figure 2a), the H-37 is injected from the sprayer. The flame was observed to be enlarged at the early stage of H-37 releasing (Figure 2b). This has consistent with a combustion promotion phenomenon reported previously (Xu, Jiang, and Ren 2016). Then, the H-37 spreads in the chamber to surround the fixed flame, which is shown in Figure 2c. Finally, the flame is suppressed by gaseous chemical fire agents due to the inhibition of radical chain reaction.

It is reported previously that most halogenated extinguishing agents added at sub-suppressing concentration result in a combustion enhancement and lead to an increase in both flame size and fluctuation. It is mainly because of the highly exothermic reactions between fluorine-containing groups and H, OH radicals (Wang

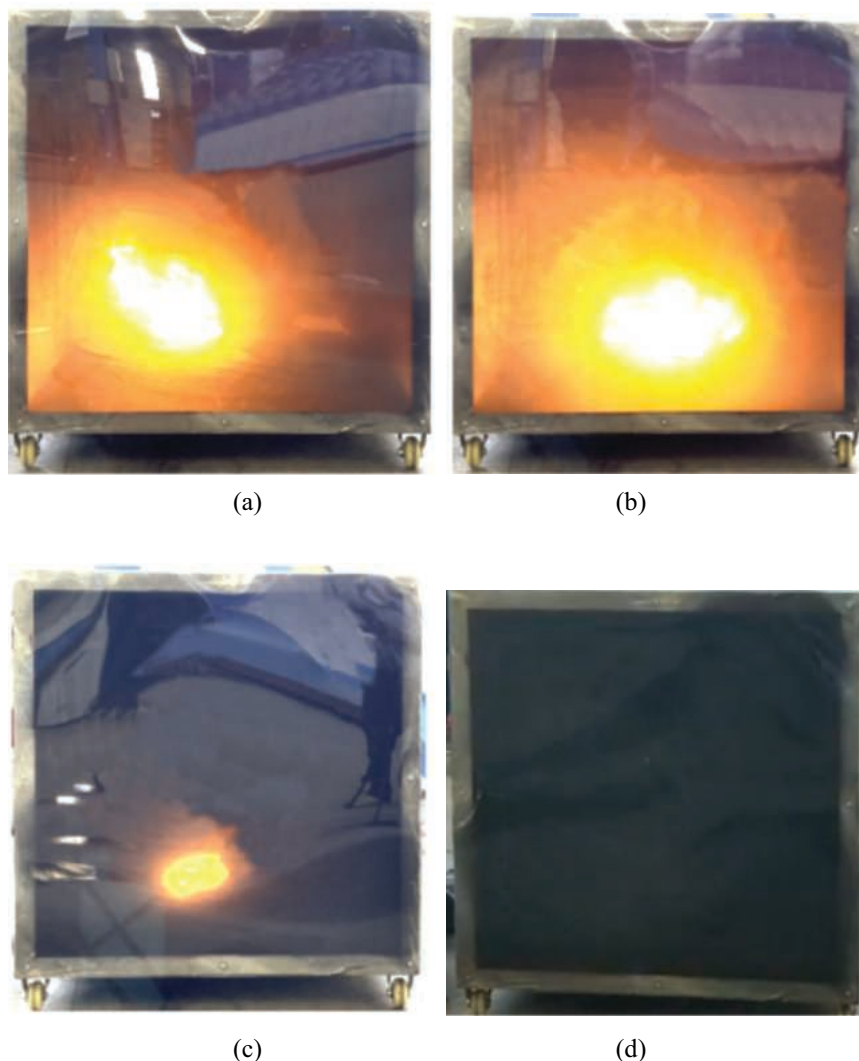
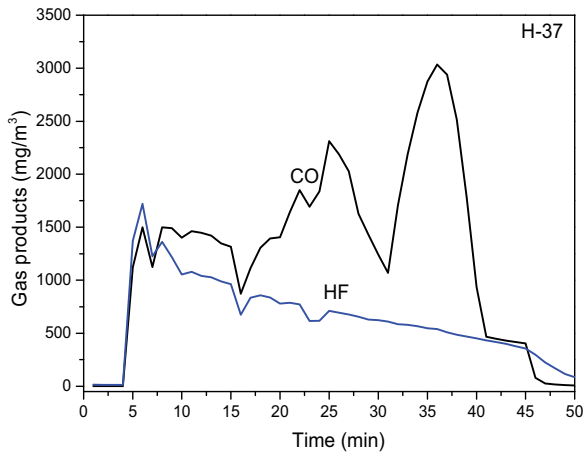


Figure 2. The description of fire suppression using H-37. (a) $t = 0$ s. (b) $t = 3$ s. (c) $t = 9$ s. (d) $t = 12$ s.

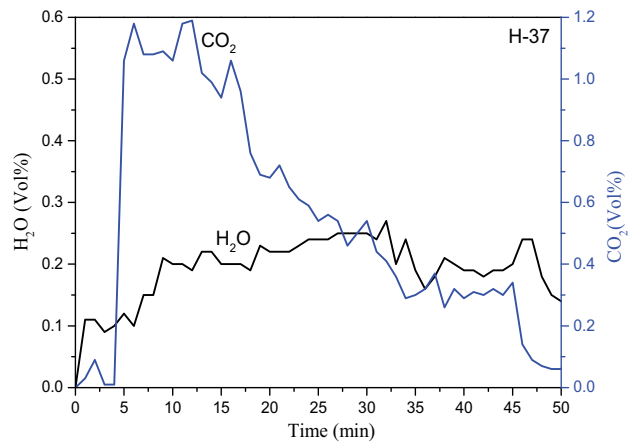
et al. 2020). From the observation, it shows that when the fluorinated chemical gas with a high boiling point is released into the chamber, the flame is much stable than the ones with a low point. For example, the flame exposing to H-2402, H-1323, and FK-5-1-12 gas is much smaller than the flame exposing to H-37 and H-1301 at the same time duration. This could be explained by the low diffusivity due to a high boiling point (Linteris et al. 2013). The reduction of combustion enhancement concerning the utilization of fluorinated chemical gas could be realized by shortening fire extinguishing time. Since the fluorinated chemical gas is widely used in the total floating system, the fluctuation of flame in the size reduces the fire extinguishing efficiency. High fire extinguishing efficiency is believed to be able to protect more historical items from fire. Therefore, the fire-extinguishing time is one of important indexes when the fluorinated chemical gas is under consideration for fire protection of historic buildings.

3.2. Characters of gas products released during fire suppression

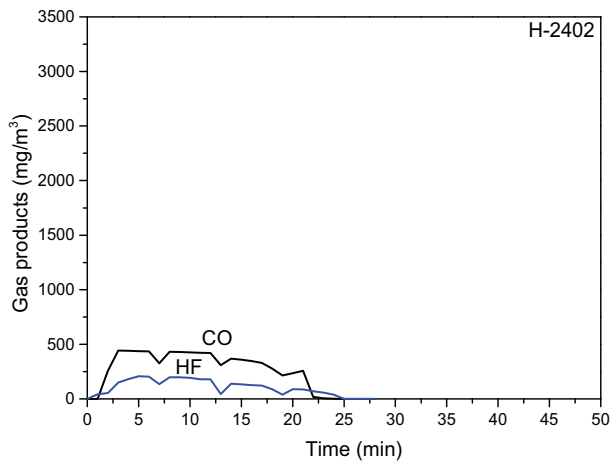
When the fluorinated chemical gas releasing into the chamber and exposing to a fixed flame, a variety of gaseous products are founded. The yielded gases include HF, CO, CO₂, H₂O, and so on. The description of main gas products' varying test times is illustrated in Figure 3. The composition of gas products varies greatly with the changing of fluorinated chemical gas. Regarding the traditional gas, CO, CO₂, and H₂O, it comes from the combustion of fuels. One of the corrosive gas products is HF. The peak of CO in gas products of fire suppression test using H-37, H-2402, FK-5-1-12, H-1323 and H-1301 is 3034.08 mg/m³, 442.49 mg/m³, 3439.43 mg/m³, 4312.6 mg/m³ and 6199.47 mg/m³, respectively. The low CO concentration is founded in the gas production of H-2402 inhibition. The maximum CO would



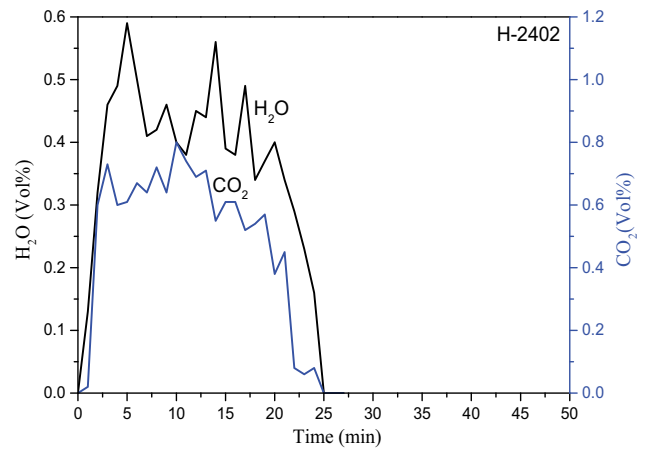
(a)



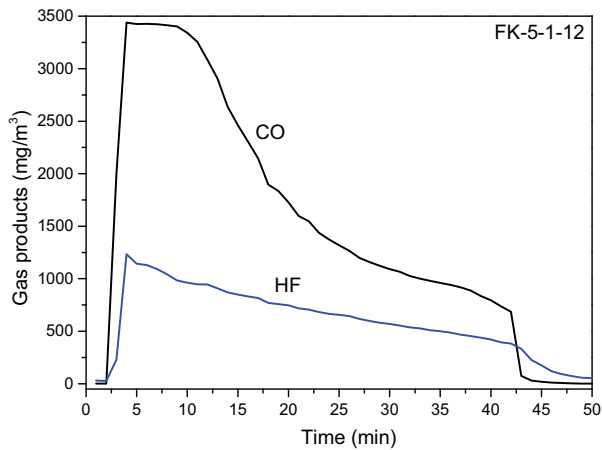
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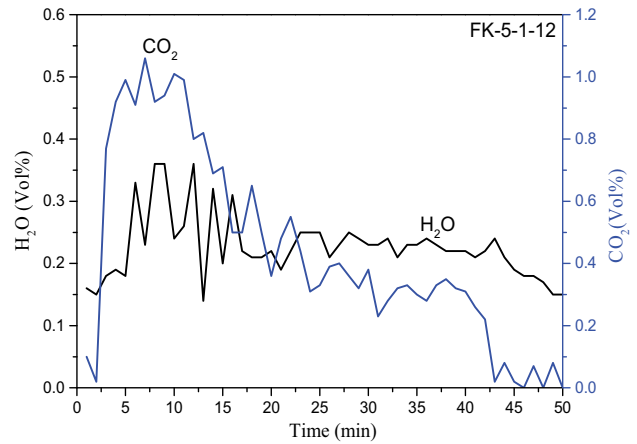
(c)



(d)



(e)



(f)

Figure 3. The gas yield of different test conditions using 5 typical chemical gas. (a) H-37. (b) H-2402. (c) FK-5-1-12. (d) H-1323. (e) H-1301.

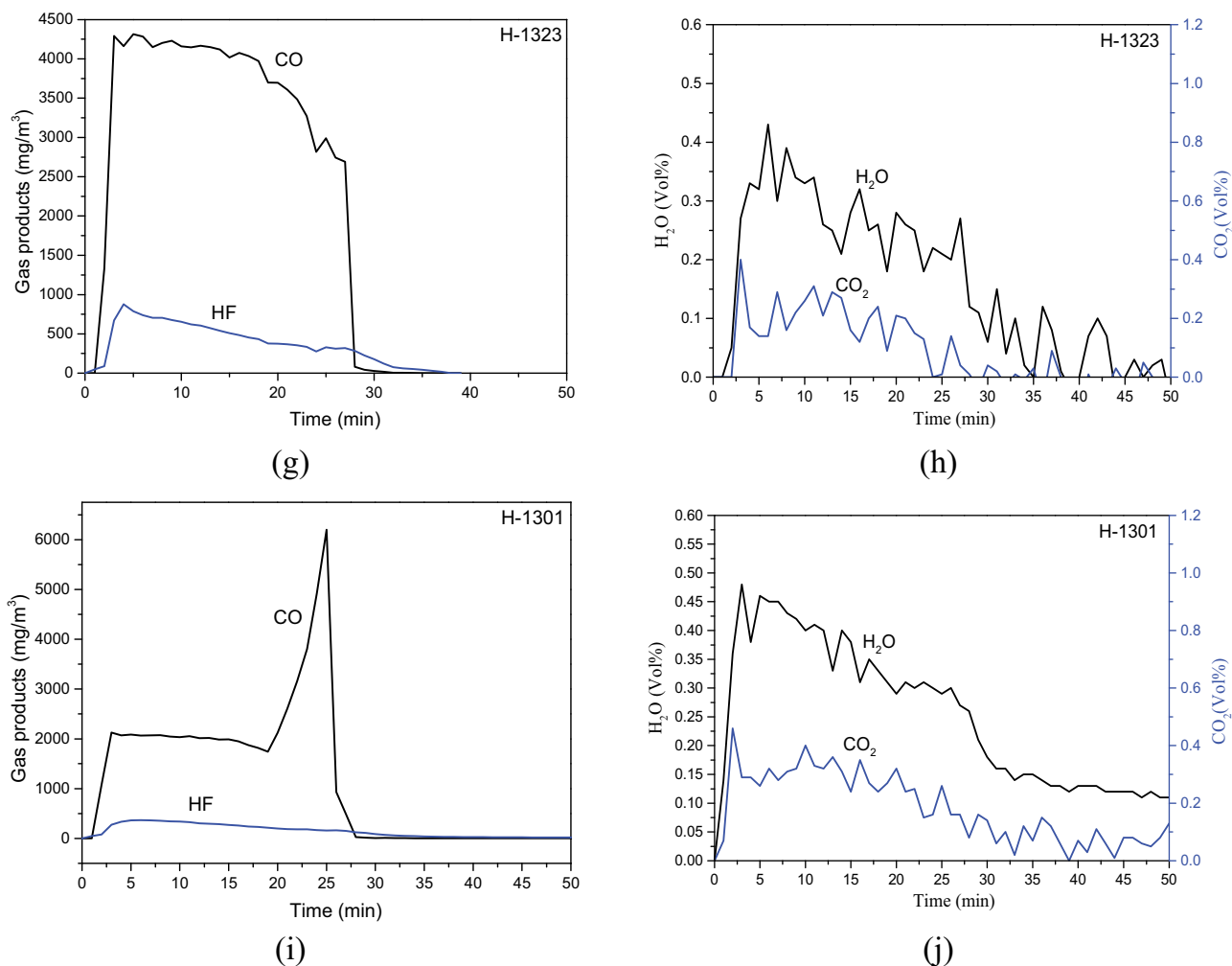


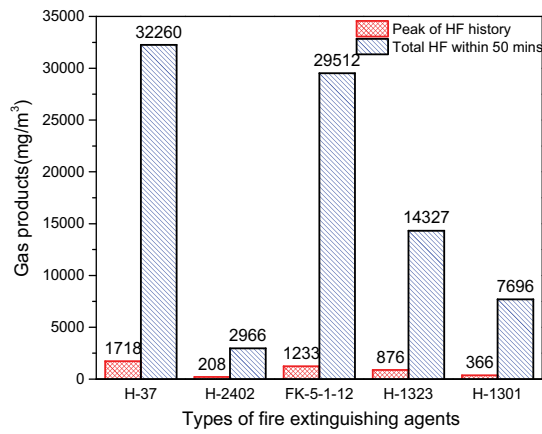
Figure 3.

be produced when the H-1301 is used to extinguish a fixed pool fire. A high yield of CO could impact a potentially harmful hazard to the people who are in the room (Ahmed and Kumar 2020). The total amount of CO during 50 min fire suppression test using H-37, H-2402, FK-5-1-12, H-1323 and H-1301 is 63,018 mg/m³, 7206 mg/m³, 74,238 mg/m³, 96,992 mg/m³ and 59,429 mg/m³, respectively. To reduce the yield of CO within the fire enclosure, the H-1323 is not recommended. Although the peak concentration of CO yield shows a total difference between H-37 and H-1301, the total amount seems the same. A low yield of CO is found when H-2402 is used to inhibit a fixed flame.

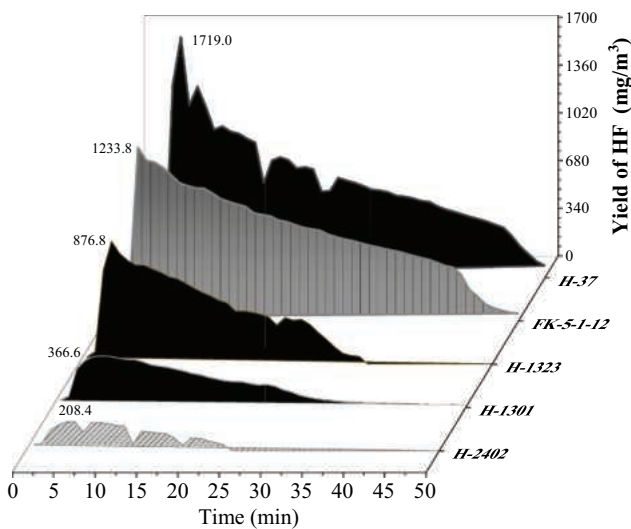
The peak of CO₂ yield in gas products of fire suppression test using H-37, H-2402, FK-5-1-12, H-1323, and H-1301 is 1.2 vol %, 0.8 vol %, 1.1 vol %, 0.4 vol %, and 0.5 vol %, respectively. The low CO₂ concentration is founded in the gas production

of H-1323 inhibition. The maximum CO₂ appears when the H-37 is employed. The total amount of CO₂ during 50 min fire suppression test using H-37, H-2402, FK-5-1-12, H-1323 and H-1301 is 27.5 vol %, 12.6 vol %, 21.9 vol %, 4.0 vol % and 9.4 vol %, respectively. The low yield of CO₂ is obtained when H-1323 is used.

When HF gas is mixed with gaseous H₂O, its corrosiveness is highly improved (Blake, Brauer, and Bachmann 2018). Reduction in gaseous H₂O is a potential method to lower the hazard impacted to the surrounding environment during fire suppression. Therefore, the H₂O concentration is also under discussion in current work. The peak of H₂O yield in gas products of fire suppression test using H-37, H-2402, FK-5-1-12, H-1323 and H-1301 is 0.22 vol %, 0.60 vol %, 0.35 vol %, 0.44 vol % and 0.49 vol %, respectively. The total amount of H₂O during 50 min fire suppression test using H-37, H-2402, FK-5-1-12, H-1323, and H-1301 is



(a)



(b)

Figure 4. The gas yield of different test conditions. (a) peak of HF. (b) HF history varying time.

10.2 vol %, 9.4 vol %, 11.2 vol %, 8.5 vol %, and 13.8 vol %, respectively. The amount of H₂O varies from 8.5 vol % to 11.2 vol %.

Once the anhydrous HF is combined with H₂O, it shows a big hazard to the surrounding materials and people (Zuo et al. 2018). It is reported that after pre-treatment by HF, some morphological changes of wood were observed indicating partial damage in biomasses structure although, their main framework was unchanged (Asadieraghi and Daud 2014). In addition, HF is believed to show considerable results on Si diminution (Asadieraghi and Daud 2014; Dong et al. 2015). The reduction of HF is a continuous goal for various researches. The HF history and peak of the HF profile are summarized in Figure 4. The peak of HF yield in gas products of fire suppression test using H-37, H-2402, FK-5-1-12, H-1323, and H-1301 is 1718.9 mg/m³, 208.4 mg/m³, 1233.8 mg/m³, 876.8 mg/m³, and

366.5 mg/m³, respectively. The total amount of HF during 50 mines fire suppression test using H-37, H-2402, FK-5-1-12, H-1323 and H-1301 is 32,260.4 mg/m³, 2966.0 mg/m³, 29,512.9 mg/m³, 14,327.9 mg/m³ and 7696.8 mg/m³, respectively. It is observed that the HF yield is low when H-1301 and H-2402 are used. To avoid high HF yield, the H-37 is not recommended.

3.3. The analysis of F deposited on the weathered timbers

It is known to all, high concentration of HF yield during fire suppression would show a danger to the surface of surrounding materials. Here, the weathered timbers are employed to be acting as traditional Chinese historic buildings. Then, the hydrogen fluoride effects on the weathered timbers are illustrated. During our test, the weathered timber chips are placed in the chamber for a while after fire suppression, which is consistent with the real situation. Then the characteristic of the specimen surface is identified with the help of SEM. The preliminary results are shown in Figure 5. Three dimensions sizes are employed here, for example, 300 μm, 100 μm, and 50 μm. The dimension size decreases from left-side (just as Figure 5a) to right-side (just as Figure 5c). It is observed that the surface of the wood is full of porous cells, which have a strong ability for gas absorption. The BET surface area of traditional wood could approach 1000 m²/g (Gao et al. 2018). After weathering by natural conditions, the BET surface area would be highly enlarged. It means that much more gases would be absorbed by weathered ones. It is believable that in the fire suppression scene when the weathered wood (acting as a Chinese historic building) is exposed to a variety of gases, the deposition of corrosive gas (HF or HBr) and water (H₂O) would impact a big hazard to the surface of materials. The surfaces of materials would be corroded when they were treated with high concentration acid. In addition, the deposited fluorine is hard to be removed since the wood or paper historic artwork is brittle. The influence of these acid gases deposited on the wood sample remains unclear.

When the weathered timber chips were placed in the chamber for a while, the surface element concentration distribution is evaluated using EDX analysis of SEM. The results are listed in Figure 6. Figure 6a–e indicates the ratio of the main elements of wood surface after fire suppression using five fluorinated chemical gases. It is observed that the deposited F on the wood surface using H-37, H-2402, FK-5-1-12, H-1323 and H-1301 are found to be 1.6%, 0.5%, 1.0%, 1.0% and 0.4%, respectively. This is consistent with the previous result of the total amount of HF, which is

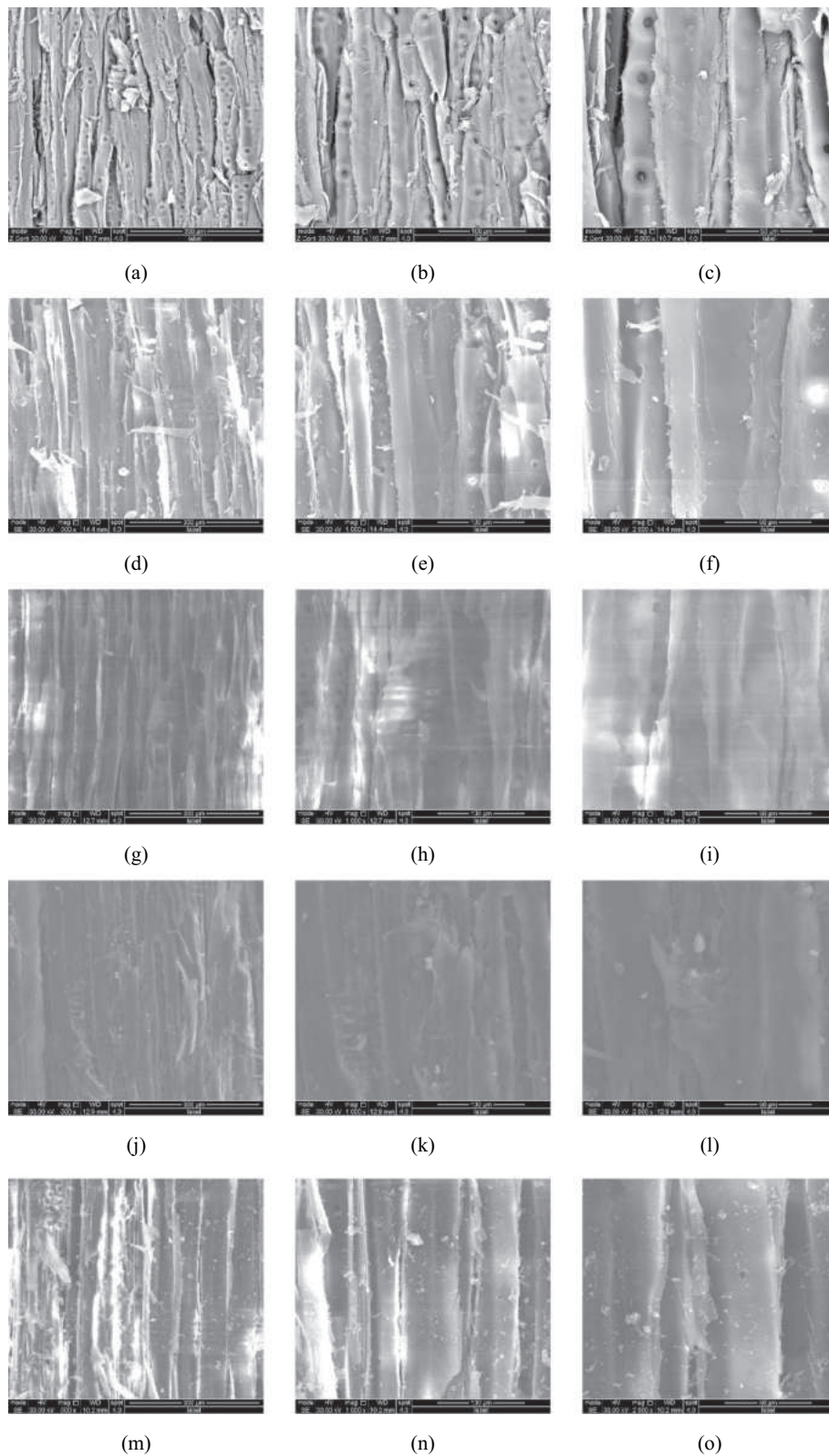


Figure 5. The SEM results of 5 tests. (a) 300 μm , H-37. (b) 100 μm , H-37. (c) 50 μm , H-37. (d) 300 μm , H-2402. (e) 100 μm , H-2402. (f) 50 μm , H-2402. (g) 300 μm , FK-5-1-12. (h) 100 μm , FK-5-1-12. (i) 50 μm , FK-5-1-12. (j) 300 μm , H-1323. (k) 100 μm , H-1323. (l) 50 μm , H-1323. (m) 300 μm , H-1301. (n) 100 μm , H-1301. (o) 50 μm , H-1301.

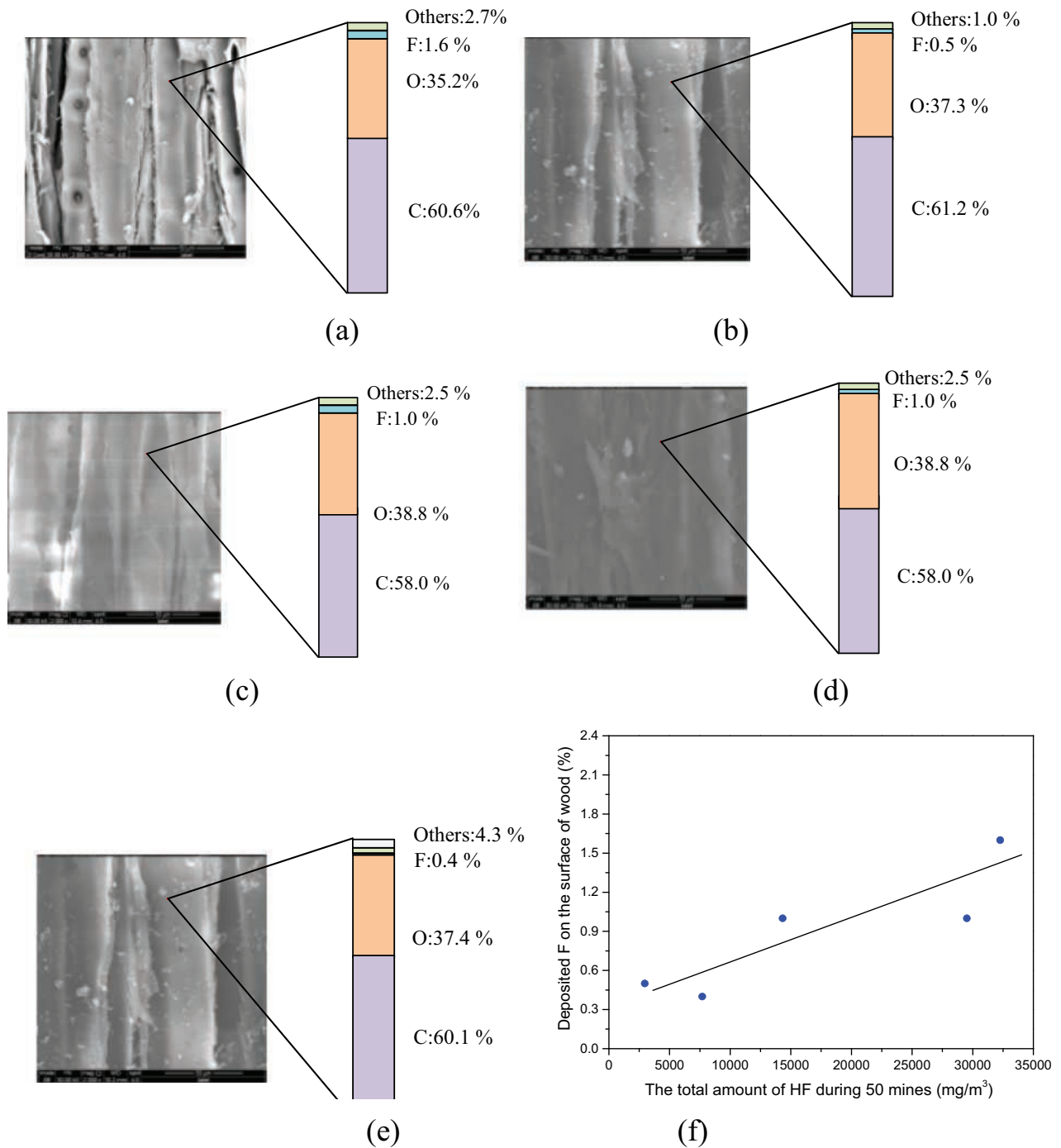


Figure 6. The EDX analysis of weathered timber surface. (a) H-37. (b) H-2402. (c) FK-5-1-12. (d) H-1323. (e) H-1301. (f) the relationship between deposition of F and the total amount of HF yielded during fire suppression.

illustrated in Figure 6f. The amount of deposited F on the wood surface is believed to be positively correlated with a total amount of HF yield during fire suppression. It is concluded that HF yielded during fire suppression could be deposited on the surface of weathered wood chips. To reduce the deposited F on the surface of materials, the H-2402 and H-1301 are recommended. However, these two fluorinated chemical gases have been prohibited to

be widely used commercially due to ill environment-friendliness. Both FK-5-1-12 and H-1323 have the same level of deposited F on the surface. H-37 yields the biggest HF in gas among the five typical chemical gas agents and results in a high ratio of F deposited on a surface. However, the effects of deposited F on the surface change of historic buildings are still need to be made further analysis.

4. Conclusion

In this work, to clarify the influence of acid gas yielded during fire suppression on the surface of historic buildings, a series of fire suppression tests using fluorinated chemical gases were conducted. During the tests, the weathered wood samples acting as the Chinese historic buildings are tested in the chamber. The characteristic of gas production and elements variety of wood surface are discussed by FTIR and SEM-EDX, respectively. Finally, the followings were obtained based on experimental results and detailed analysis:

- (1) The fluorinated chemical gas is effective for flame inhibition under a fixed condition. The flame shape varies the physical parameters of fire agents. When the boiling points of fire extinguishing agents are high, the flame enhancement and enlargement after the injection of chemical gas becomes weak.
- (2) The total amount of HF during 50 min fire suppression test using fluorinated chemical gas is ranked as H-37 > FK-5-1-12 > H-1323 > H-2402 > H-1301.
- (3) The SEM image of weathered timber illustrates that the surface of the wood is full of porous cells, which have a strong ability for gas absorption. The experimental results indicate that the amount of deposited F on the wood surface is found to be positively correlated with a total amount of HF yielded during fire suppression.

However, the effects of deposited F on the surface change of historic buildings should be further studied.

Disclosure statement

No potential conflict of interest was reported by the author(s).

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