

## How to produce and emit pollutants from uranium enrichment plant

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The only important materials that will be found in uranium enrichment plant include uranium hexafluoride, uranium tetra-fluoride, and uranium fluoride. UF<sub>6</sub> is around solid phase in both gas and liquid states react with existed water and water vapor around reaction quickly, and the result of this reaction will be hydrogen fluoride. Since estimation the resulted dangers by UF<sub>6</sub> and help to protect the employees in nuclear centers and surrounding dwellers have been always mentioned by specialists. Therefore, essential information was provided about how to emit these materials for people dealing with them or specialists in nuclear safety and the environment to prevent from probable dangers, because the maximum permitted concentration of HF gas in environment mustn't be more than 2-3 ppm, while 10ppm HCN concentration was discerned in the threshold of cyanide toxicity. This issue claims that the importance of emitting HF gas is high in environment and the resulted dangers in the inhaling air is not compensable or negligible. How to emit or transmit the existed pollutants in surrounding environment in various conditions is such cases have been studied and more simply, it must be known transmitting or emitting pollution such as UF<sub>6</sub> in installation space, reactions, and following products in each part of factory, in further place, and substance concentration, and finally changes of dangerous and toxic effects according to distance. The aim of this study is conclusion and analysis of issue and comprehensive and useful data about the manner of emitting pollutants and their performance in environment that are different in different climates and geographical conditions.

**Keywords:** UF<sub>6</sub> emission, HF gas, uranium enrichment contaminators

### INTRODUCTION

The working material in uranium enrichment plant is UF<sub>6</sub> (Uranium Hexafluoride). Whenever this material is beside humidity produces a dangerous toxic materials called HF which makes working with UF<sub>6</sub> difficult. Although, HF usage in industry is high, one of the main problems of uranium plants is its emission in environment that is considered dangerous material for other people, and it can be surely stated that the resulted danger by HF emission in environment and space around systems is much more dangerous than radiation of radioactive materials in location [1-3].

If sulfuric acid contact with hand skin or any other parts of body, it will burn skin significantly. If it is hydrolyzed, the caution is the precondition of working for being toxic. It is more than 200 years that halogen compounds have been mentioned for production and their properties according to scientific and practical view both experimentally and really. The maximum HF gas concentration mustn't be more than 2-3 ppm, while the 10 ppm emitted HCN concentration is detected as the threshold toxicity of cyanide. This issue shows that the importance of emission and releasing HF gas in results in the release of radioactive materials in the

environment is so high, and the resulted dangers in the inhalation air are irreparable and non-negligible [4, 5].

All types of gaseous, without water, or aqueous HF have good reactivity, and metals, glasses, and other compounds such as silicon, leather, and natural plastic will react with HF. In addition, both aqueous and gracious forms of HF are significantly corrosive and cause severe irritation to skin, eyes and olfactory mucosa, as well as causing severe irritation of the respiratory tract. Intensive exposure to great amount of fluoride gases and particles cause nausea, vomiting, loss of appetite and diarrhea or constipation. Fluoride poisoning or other adverse effect can be resulted from being significantly exposed to it. It must be mentioned that leakage of any radioactive material to environment may have chemical dangers besides radiological dangers. The possible mechanisms to produce HF are the main chemical contaminations in uranium processing and enrichment plants and needed equations will be examined to determine concentration and properties of this material in the present article.

This article analyzes some experimental environment and under different conditions such as snow and rain or firing accidents. Since there are many conditions according to climate and building while radioactive emission in environment, high reactivity of radioactive materials with air

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humidity, and erosive properties of some materials such as HF, examining the present condition will have significant role to know why each ones influence on emission and also climate of emission place in dilution and next effects.

### RADIOLOGICAL DANGERS

Each radioactive material that is emitted to environment has series of radiological dangers that people exposure to irritation is different depending on amount of the emitted material and contact with them. Radiological dangers and materials can be divided to three different classes for easiness as following [6, 7]:

- First class: ones with direct effects
- Second class: ones with indirect effects
- Third class: dangerous materials

Now, each class is described:

#### *First class:*

These have direct effect on humans. These materials are divided to three sets depending on amount of their danger:

- 1- Materials producing a total effective equal dose about 1 Sv for employees and who are in contact with materials.
- 2- Materials producing a total effective equal dose about 0.25 Sv for people out of control level.
- 3- Absorbing 30 gr or more solvable uranium by anyone out of control level.
- 4- Acute radiation by used permitted materials or durable process products with undesirable effect on health of people out of control level.

Materials with direct dangers but more balanced than previous class include the following cases:

- 1- Materials producing a total effective equal dose about 0.05 Sv or more for each person out of control level
- 2- Materials producing a total effective equal dose about 0.25 Sv for people in contact.
- 3- 24-hour leakage of radioactive materials out of control level.
- 4- Acute radiation for each person from radioactive materials to their products with long-term effects or soft effects on each one out of control level.

Materials of the second class: materials with reaction

These materials don't have radioactive properties themselves, but they can reaction with other materials and make dangers of the first class materials.

Materials of the third class: materials that may make danger.

These materials have accidental dangers and potential power for dangers. Consequently, they are

harmful for job affairs and must be examined, but they can't be controlled.

### CHEMICAL DANGERS

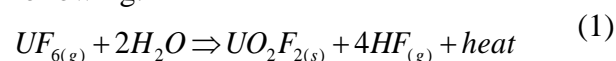
This part describes chemical reactions among various classes and its radiological dangers. In addition, analysis of reactions and probability of emission and exposure to radiation of dangerous chemical materials such as HF will be discussed, then escaping ways of these materials to environment around plant will be stated.

#### *Chemistry and Chemical Reactions*

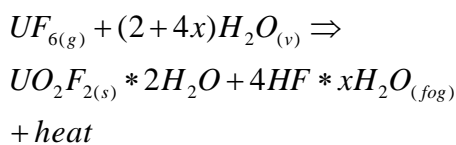
Isotope separation is a physical practice rather than a chemical process. Physical methods without chemical reactions are used by any techniques in uranium enrichment. Of course, the objective is not preparation input feed to plant, but it includes uranium enrichment operations; UF<sub>6</sub> behavior in all possible phases as liquid, solid, and gas can be so dangerous against water. This material has great reactive intention to water and will have exothermic reaction with water and water vapor in the atmosphere. The product of UF<sub>6</sub> hydrolysis is materials such as UO<sub>2</sub>F<sub>2</sub> in solid form and HF in gas form. Therefore, chemical reaction and producing undesirable products can be completely covered in installations and prevent from its contact to humidity. Other chemical reactions in enrichment plant and nuclear fuel production will happen in decontamination process. The reactions can make toxicity and danger for employees, damage metal installations, and make corrosion in levels.

#### *HF uranium reaction with water*

UF<sub>6</sub> in both gas and liquid forms that are usually around solid react with water and vapor around quickly and produce HF without water if there is enough water vapor and its low amount also has corrosion for pipes and installations. Therefore, enrichment system must be excavated from humidity and air before HF gas injection. Overall reaction of HF with water is as following:



If UF<sub>6</sub> is emitted in room temperature and humidity in air in that temperature, the reaction products include UO<sub>2</sub>F<sub>2</sub> hydrate and also HF-H<sub>2</sub>O mass that are seen as white cloth. Reaction with extra water is summarized in the equation:



If the humidity is low, so the color is not white cloud and form solid particles of  $UO_2F_2$ .  $UO_2F_2$  has solubility in water and is yellow color solid whose color exactly depends on its size and hydration degree. The produced heat in the first reaction is 288.4 kJ/kg of  $UF_6$  gas; the produced heat by reaction with extra water, if  $UO_2F_2$  is hydrated and HF- $H_2O$  cloud is also made, will be 2459 kJ/kg of  $UF_6$  vapor.

If this reaction is formed in gas phase and high temperatures, it will be so spontaneous and sudden. The rest of reaction and progress between solid surface and extra water vapor will happen slowly, and uranyl fluoride layers will be formed. This reaction is formed on  $UF_6$  surface and prevents from reaction progress.

#### *Interaction of $UF_6$ with various materials*

Effects of  $UF_6$  according to properties and abilities of fluoride ion compounds and also their oxidation properties are so significant. Many chemical properties of  $UF_6$  are attributable to stability of  $UO_2^{2+}$  ion that let to react with water, oxides, and salts including oxygen ion such as  $SO_4^{2-}$ ,  $NO_3^{2-}$  and  $CO_3^{2+}$  without molecules of releasing  $O_2$ .

$UF_6$  reaction was undesirable with hydrocarbons and can happen intensively. Gaseous  $UF_6$  reacts with hydrocarbons and makes a black substance from carbon compounds and uranium. If hydrocarbons are mixed in liquid phase in high temperature with  $UF_6$ , they can be oxidized combustible; therefore, no trace of grease must remain in parts of  $UF_6$  flowing or solid and any other phase in contacting with hydrocarbon when lubricating in system.

The vacuum pumps are usually lubricated by perfluoropolyether grease and its trade name is Fomblin grease. The mentioned grease was non-flowable and is fully fluorinated. Therefore they don't react with  $UF_6$  in many usual working with it. The small particles of uranium compounds with hydrocarbons may exist in Fomblin grease, or  $UF_6$  or even  $UO_2F_2$  may penetrate in grease, or reduce capacity and efficiency of vacuum pumps by sediment on flow path.

Reusing Fomblin grease by separating compounds and particles from grease are possible by operations such as filtration, sedimentation, and centrifugation. Anhydrous sodium carbonate is added to fomblin

grease and uranium compounds produce uranyl sodium carbonate after reaction with the mentioned sodium carbonate, this product is so unsolvable and precipitative; therefore, the obtained sediment is separated by filtration and centrifuge. The remained effect of hydrocarbons is removed and heat is produced by adding active carbon in fomblin grease. Meanwhile the added carbon will precipitate after a while and remove from the down of container.

#### *$UF_6$ reaction with building materials*

Corrosion of metal equipment and removal of nonmetal sealing are prevented by special insulation materials according to construction compounds and purification. A layer is usually formed on metal by reaction with metal that may reduce or even remove the next reactions. The non-metal materials damage these materials for being exposed to process fluid and its condition. The used materials in piping sealing, flexible pipes, and other sealing materials that must be exchanged after sometimes are in this group.  $UF_6$  and some reaction products have potential of corrosion such as HF.  $UF_6$  has a fluorinating factor that can react with most metals. The reaction between  $UF_6$  and metals such as nickel, copper, and aluminum make a protective fluorinated film on metal preventing from more reactions. These materials are somehow inefficient against  $UF_6$  corrosion and are less impressible. Therefore, they are proper in working with  $UF_6$ . Aluminum is used in  $UF_6$  in pipes compounds, because it can be a good insulator against corrosion  $UF_6$ . Carbon steel and stainless steel at high temperatures may be incompatible with  $UF_6$ , but they are not influenced by it in common temperature and its functional temperature.

Light gases such as HF and air are separated in  $UF_6$  process. Although, HF is an intensive corrosive material in aqueous solution with hydrofluoric acid, it make metal corrosion slowly, because reaction between  $UF_6$  and water is done fast, the remained HF changes in non-aqueous form. The metal corrosion rate with  $UF_6$  in uranium enrichment plant is shown in the following Table 1.

Resistant metals such as stainless steel is used for pipes and valves, and aluminum is used to cover seams and leaking spaces as washer. The used non-metals in plants and  $UF_6$  systems are used in structure of the valves and equipment. All materials used in manufacturing washers and cans must be confirmed. Butyl rubber, Keaton and Kel-F are examples of sealed materials against  $UF_6$ .

**Table 1.** Corrosion rate of UF<sub>6</sub>

Materials	The corrosion rate at 20 °C/year	The corrosion rate at 100 °C/year
Aluminum	6.6 E-7 mm	8.4 E-5 mm
stainless steel	1.4 E-4 mm	0.03 mm
Copper	1.2 E-4 mm	3.3 E-3 mm
Nickel	< 0.05 mm	< 0.05 mm

Used materials in UF<sub>6</sub> installations are shown in Table 2. The used tanks in plant have individual standards for transportation. The nominal and the minimum essential thickness for walls are listed in the above Table, and this Table is about the related standards.

The output materials of waste system are relatively neutral in reaction to UF<sub>6</sub> and their corrosion rates are shown in the previous Table. It can be concluded that these materials are more

proper for having longer life time for UF<sub>6</sub> services. According to Table 2, it is also found out that used cylinders in maintenance and transportation UF<sub>6</sub> are made in steel carbon. The uranium byproduct cylinders (UBCs) are also exposed to air and humidity and their corrosion from humidity and impurities must be considered. The corrosion rate on external surface of steel carbon cylinders are varied, and steel carbon maintaining tanks can be painted to be prevent from corrosion.

**Table 2.** Used installations material in UF<sub>6</sub> system

Installations	Material	The nominal wall thickness	The minimum wall thickness
Feed tanks UF <sub>6</sub> ( 48X, 48Y And (UBCs 48Y	Steel Carbon ASTM A516	16 mm	12.7 mm
Product cylinder UF <sub>6</sub> ( 30B )	Steel Carbon ASTM A516	12.7 mm	8 mm
Sampling bottle ( 1S )	Nickel /Monel ASTM B162	1.6 mm	1.6 mm
Sampling bottle ( 2S )	Nickel / Monel ASTM B162	2.8 mm	1.6 mm
UF <sub>6</sub> pipes	Aluminum and stainless steel	3.7 mm	Unknown
UF <sub>6</sub> valves	Aluminum and stainless steel	Greater than 3.7 mm	Unknown
Cooling trap	stainless steel	8 mm	Unknown

External corrosion may happen on the outer surface of the cylinder and in points between them which are blocked by indentation. The mean corrosion rate was obtained less than 0.051 mm/year for UBCs and mostly happens in external surface of steel carbon. On the other hand, the external corrosion of steel carbon was reported 0.041 mm/year for tanks contacting to ground surface in a different report. These values show that service life time is more than 50 years. Two products are obtained in uranium enrichment process:

*First product:* it has more uranium 235 and is called enriched uranium. It consists of 5% of uranium 235.

*Second product:* it is called remained. It is poor according to 235 and has mean concentration of 0.32%.

UF<sub>6</sub> are brought in enrichment plant by 48X or 48Y cylinders. Hot air around tank is circulated and

makes gaseous UF<sub>6</sub> from its solid phase. Gas flow is controlled to be under atmosphere for separation in waterfall systems.

**Table 3.** Specifications of the used cylinders

Length	Diameter	Gross weight	UF <sub>6</sub> weight	Cylinder type
2070 mm	762 mm	2912 Kg	2277 Kg	30 B
3020 mm	1220 mm	11580 Kg	9539 Kg	48 X
3728 mm	1232 mm	14860 Kg	12501 Kg	48 Y

### USED COMPOUNDS AND PROPERTIES IN EXAMINATION THE EMISSION PROCESS

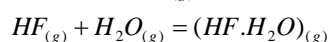
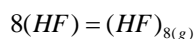
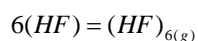
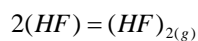
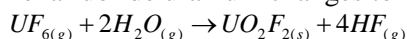
UF<sub>6</sub> can be gas or combination of solid and gas suspended in air under the specific temperature and pressure, when UF<sub>6</sub> is solid, it change to gas is done by sublimation process. It must be mentioned that the produced HF has various compounds which

must be mentioned in sediment process. HF polymers are seen greatly in vapor phase and the chemical reaction produces the following materials:

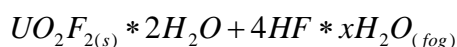
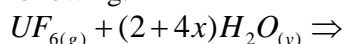
Vapor:  $H_2O$ ,  $(HF)_8$ ,  $(HF)_6$ ,  $(HF)_2$ , HF,  $UF_6$   
 $HF.H_2O$ , and dry air

Solid:  $UO_2F_2$ ,  $UF_6$

When  $UF_6$  penetrate out, it can be stated that all hexafluoride uranium changes to FH and  $UO_2F_2$ .



Free HF compounds themselves have toxic effects and the maximum permitted level for 8h/day inhalation is 0.1 ppm, because exposing more than 8 h make a great volume of this materials to find way to livers and damage its cells (about 22 liter/min air). If  $UF_6$  permeation is in room temperature and presence of air humidity in that temperature, reaction includes hydrated  $UO_2F_2$  and mass  $HF-H_2O$  that are seen as white cloud. The reaction with extra water is summarized as following:



+ heat

If humidity is low, the white cloud isn't made and solid  $UO_2F_2$  particles are made.  $UO_2F_2$  has solubility in water; the yellow solid whose color exactly depends on its size and hydration degree. The produced heat in the first reaction is 288.4 kJ/kg from  $UF_6$  gas and the produced heat in reaction with extra water is 2459 kJ/kg of  $UF_6$  vapor if  $UO_2F_2$  is hydrated and  $HF-H_2O$  cloud is made.

#### CONTAMINATOR EMISSION IN PRESENCE OF VARIOUS CONSTRUCTION MATERIALS

Contaminators emission can be influenced by various obstacles in air stream or reactive materials path. Therefore, various states happen in these conditions that each one can significantly influence on materials distribution in environment. Hence, first various emission conditions on building are examined without considering other climate conditions of the region or sophisticated events that are mentioned in the following: (Perkins, 1974).

##### *Effects of buildings and obstacles on ground*

$UF_6$  emission occurs near building and may be influenced by the effect of various obstacles in

path. These obstacles change emission streams and deviate boundary layer. Therefore, dilution, emission, and distribution of  $UF_6$  will change. The influencing factors to notice the effect of these obstacles are as following;

- Buildings may be high and their distance is short, so this condition traps  $UF_6$  between walls.
- $UF_6$  emission maybe from tanks, pipes, other other installations inside building and penetrate to out through side air among windows or chimneys and roof of the building.
- Emission from a short chimney can guide  $UF_6$  toward ground and near buildings, so its concentration will increase in region.

It is obvious by what was stated that the effect of building and obstacles on ground in emission path can have significant effect on stream pattern and change its concentration. Answering to the following questions will teach us to use which formulas and regulations to get contaminator concentration in various conditions and distances (Danvers, 1995)

- Where is the resource of contaminator?  
Is this resource in wind counter direction toward building or not?  
Is this resource in on building?  
Is this resource in wind direction toward building or not?
- Where is the contamination receiver system?  
Is it on building surface?  
Is it near or back of building?  
Is it near building in wind direction?
- How much error is made by predicted changes of contaminators concentrations for presence of building obstacles?
- Which level of buildings sophisticated are the formulas incorrect, and how near are the conditions and assumptions to the reality?
- Is there efficient data to be used in essential formulas?

Three samples of emission positions are studied individually in this research as following;

- 1- Trapping and emitting contaminator mass among building and the related effects.
- 2- Emitting contaminator on building or through chimney
- 3- Emitting contaminator mass form chimney and it stream to downward.

#### EXAMINING CONTAMINATORS EMISSION IN DIFFERENT CLIMATES

When contaminators penetrate around systems in nuclear plants, they mix with the surrounding air fast and will distribute in gaseous form in space.

One condition that can significantly influence on amount and type of each contaminator or even on sediment and removal of contaminators from surrounding space is raining. Raining and snowing wash contaminators including  $UF_6$  (by reaction with them), increase the HF concentration on ground, and prevent from its emission by air. The contamination mass includes mixture of gases, solid, and suspended particles in air. Other smaller particles react with materials and obstacles on the ground in stream path and will be removed from atmosphere by plant coverage.

#### *Removing pollutants by gravity and sediment process*

The sediment rate by gravity will be about 10 cm/s for the bigger emitted particles such as  $UF_6$  in air whose diameter is bigger than  $50\mu m$ . In this case, removing toxic materials will happen by gravity. Except big particles of  $UF_6$ , other products of hydrolysis reaction such as  $UO_2F_2$  have small diameter about  $1\mu m$ . Other materials like  $UF_6$  or gaseous HF, and also  $HF.H_2O$  won't be evaluable by gravity. The  $HF.H_2O$  compound is considered gaseous and will be evaluated by dry sediment formulas. The discussed particles and gases can be separated from the emitted mass and then precipitated by rain, snow, fog, or cloud by two methods:

- In fog and cloud: by small particles of fog and clouds
- Under the cloud: by precipitation though descending drops of snow or rain on ground among a contamination mass.

#### **RADIOACTIVE MATERIALS EMISSION BY EXPLOSION**

The most concerns about radioactive materials are after explosion in an extensive area. In such cases, more volume of radioactive materials can exposed people to mortality level. The most popular scenario of attacking to uranium enrichment plant or feed or plant products containing vehicles. The great emitting resources such as initial feeds and the enriched uranium products won't have easy transportation, and constructing proper protection is not simple. However, chemical reactions with the present water vapor in air occur quickly and contaminators distribute in extensive area for being floating in air by a missile attack or bombardment of tanks, environmental degradation, and releasing  $UF_6$  in environment; in this case, special filters and proper ventilation can't prevent from emitting materials to environment in plant space.

Emission the resulted contaminations by explosion depends on explosive strength, adhesion and weather conditions, portability of radioactive particles by air particles, and explosive materials. Each emitted gas emit radioactive materials besides HF, but finally sediment on materials and grounds surfaces as mental particles and makes environmental contamination. The most probable case in this state is just small areas in region are contaminated, but chemical contaminations must be scrutinized not to distribute by water flows and wind blowing in other areas.

Radioactive gases, liquid, and suspended particles are internal and external radiation resources. Distance reduces dangers and irritation exposure. Areas with high risk can be explosive to several kilometers. An explosion with lower amount of radioactive materials can contaminate great area as big as city or small area as big as several kilometers.

#### **EMISSION THE MAIN MATERIAL OF URANIUM ENRICHMENT PLANT ( $UF_6$ ) BY FIRING**

It is assumed that cylinder with  $UF_6$  is damaged by fire accidents and consequently  $UF_6$  of these tanks will be emitted to environment. The assumption of this study is the presence of tanks with excavated  $UF_6$ . It means the output of centrifuge which was poor of  $U_{235}$  isotope and is the working basis of non-suspended distribution model (Monte Carlo Lagrange diffusion model). Accidents and events leading to  $UF_6$  emission and hydrolysis reaction of products will be in one fire accident that are divided to three total classes:

First class: during which the cylinder is in the fire, finally the tank pressure increases by high heat of fire and  $UF_6$  evaporation in cylinder, so it damages and emit of  $UF_6$ .

Second class: during which cylinder is broken and  $UF_6$  goes into fire.

Third class: in includes accidents happening after extinguishing the fire and the cooling stage.

Each occurred situation has its actions to reduce contamination and specific formulas to examine contaminator concentration. The obtained concentration in this part is based on time function above ground level in favorite internals. The  $UF_6$  concentration in studies and also hydrolysis products such as HF and  $UO_2F_2$  has been obtained in two different climates:

- 1) Stable D condition with wind speed about 4m/s
  - 2) Stable f condition with wind speed about 1m/s
- The determined concentration in this field will be maximum that an individual can receive in his/her

position. This issue is so important for individual health and hygiene.

*Description the excavated UF<sub>6</sub> emission in fire accidents*

When UF<sub>6</sub> containing cylinders are transported from one location to another in inside or outside of plant many be floated in fire in one of firing probabilities events. This accident my happen in vehicle that work with gasoline fuel. Therefore, the related fire is also provided by this fuel. In this condition, fire heat increases the tank inside temperature, so sublimation, and emission of UF<sub>6</sub> to surrounding. This sublimation can break walls and emit hot UF<sub>6</sub> to environment by increasing tank internal pressure, and its hydrolysis with water happens quickly. The continuity of firing will guide amount of UF<sub>6</sub> to above fire and being hydrolyzed quickly. Finally, UF<sub>6</sub> emission will continue to 30 min after fire exhaustion or its suffocation. Flotation reduces and emission stops after increasing mass density.

In addition, it is assumed that UF<sub>6</sub> react immediately after going out of tank, and produces hydrolysis products such as HF and UO<sub>2</sub>F<sub>2</sub> with 1 μm diameter. Therefore, this part just evaluates concentrations of these two materials. UO<sub>2</sub>F<sub>2</sub> for high molecular weight can have very low rate and precipitate on ground, but HF is gaseous and non-precipitated, but can be absorbed by plant coverage. (Role of plant coverage is evaluated in this article). However, physical and chemical properties and

conditions are two important issues that must be considered.

Two general regulations are determined in next studies for the resulted dangers by emitting toxic products of UF<sub>6</sub>. First class is accidents with irreversible potential effects, and another class is harmful potential effects. The threshold of harmful potential dangers for UO<sub>2</sub>F<sub>2</sub> is 10 mg and for HF is determined 1 h.

*Flotation effects of spring*

Two equations classes are used to consider mass flotation condition in resulted accidents by fire: one id for fires and another is for moment heats. In order to obtain the proper formulas to determine hydrolysis products concentrations of UF<sub>6</sub>, dominant boundary conditions on problems must first be identified. It is also assumed that when mass is raised from ground level by buoyancy force, it won't be settled on it again, actually it won't go to the lower boundary layer.

*Uf<sub>6</sub> mass emission*

There are three classifications for each condition of UF<sub>6</sub> transmission in text:

- State based on non-flotation of mass.
- state such as fire smoke
- State of heat floating

Table 4 shows 5 evaluated accidents in this article. These accidents include three types of initial accidents with 2 sub-set encompassing all existed cylinders in UF<sub>6</sub> emission [5].

**Table 4.** Description of 5 evaluated accidents in fire

Accident	Description	Chemical form	LB value	Time (min)	Emission surface
Fire of vehicles, three tanks filled with UF <sub>6</sub> and an 48G	Three filled tanks with UF <sub>6</sub> will be damaged by firing for fuel combustion or other combustible materials in vehicles hydraulically	UF <sub>6</sub>	0	0-12	ground
			11500	12	
			8930	12-30	
			3580	30-121	
Fire of vehicles, three tanks filled with UF <sub>6</sub> and an 48Y	Three filled tanks with UF <sub>6</sub> will be damaged by firing for fuel combustion or other combustible materials in vehicles hydraulically	UF <sub>6</sub>	0	0-24	ground
			18000	24	
			2770	24-30	
			8010	30-236	
Fire caused by small aircraft collided, two tank filled with UF <sub>6</sub> and an 48G	1 UF <sub>6</sub> full tank is damaged by airplane collide. Another tank is hydraulically damaged by fire resulted from airplane fuel.	UF <sub>6</sub>	0	0-12	ground
			3840	12	
			2980	12-30	
			1190	30-121	
			4240	0-30	
Fire caused by small aircraft collided, two tank filled with UF <sub>6</sub> and an 48Y	1 UF <sub>6</sub> full tank is damaged by airplane collide. Another tank is hydraulically damaged by fire resulted from airplane fuel	UF <sub>6</sub>	0	0-24	ground
			6020	24	
			920	24-30	
			2670	30-236	
			3210	0-30	
Firing the vehicle, 2 waste tanks	Vehicle had an accident and two waste tanks are damaged or the fuel of vehicle made UF <sub>6</sub> combustion and emission.	UF <sub>6</sub>	39	30	ground
			10	30	

The first method is when tank is full of UF<sub>6</sub> has been immersed in whirlwind of flames. Vehicle may lose its control in this accident and diesel fuel in vehicle tank is poured out and makes fire for 30 min. it must be mentioned that vehicle tank capacity and type make difference in size and time of firing. It is assumed for more examinations that the three tanks full of UF<sub>6</sub> are excavated in vehicle. If tank is not broken by this collide, firing will eventually break it. As soon as breaking, UF<sub>6</sub> emits out of tank and mixes with surrounding air. This makes a little floatation. Since three involving cylinders in firing will not be damaged simultaneously, UF<sub>6</sub> emission will happen permanently and gradually for each cylinder. In addition, it is assumed that there must be at least 30 min interval for tank break. 48Y and 48G tanks were studied in this article.

The second method in this text is collide of a small airplane with tank full of UF<sub>6</sub>. It is assumed in this accident that collide breaks the first cylinder in the first moment and the second cylinder will be damaged in fire. Description the elements of accidents are similar to the first accident. In this case, just two sets are considered, one is two 48G cylinder, and another is two 48Y cylinder.

Third method doesn't have sub-set and irradiation from waste cylinders is mentioned. Accidents by both airplane collide or firing, include hydraulic damage or cooling stage. These tanks are small and have a little UF<sub>6</sub> amounts. It is assumed that all UF<sub>6</sub> will be emitted by firing in 30 min.

*UF<sub>6</sub> emission and temperature data*

**Table 5.** Summary of 6FIRE model results.

	Thermal degradation		Primary deficit	
	48G	48Y	48G	48Y
The initial crime UF <sub>6</sub> , lb	28000	28000	28000	28000
Time destroyed, min	12.2	23.9	0	0
The combination of demolition, lb (%)	24163 (86.3)	21984 (78.5)		
Solid	3523 (12.6)	4744 (16.9)		
Fluid	314 (1.1)	1272 (4.6)		
Steam				
Reservoir pressure during demolition, psia	670.9	231.8		
Liquid temperature during demolition, o F	284.9	260		
Steam temperature at the time of the destruction, o F	611.5	672.7		
The average temperature in the destruction of the shell, o F	1085	1125		
Initial Release, lb	6016	3837	0	0

*Climate conditions*

Two climates were studied for each determined conditions previously that the first condition is related to the F stability with wind rate of 1 m/s and the second condition is related to D stability with

In this part, 48G and also 48Y cylinders with 14 ton capacity were studied that were excavated from UF<sub>6</sub> which were floated in fire of 1475°F for 30 min. two emission types were considered for 48G and 48Y cylinders. [2, 6]

Tank was put in fire without being damaged. In this accident, 48Y and 48G cylinders will be damaged in 24 and 12 min, respectively after being floated in fire. However, cylinder damage is by increasing its internal pressure; UF<sub>6</sub> is poured out of tank suddenly in this time. After tank damage, if it is still in fire, and UF<sub>6</sub> emission continues with steady rate. Fuel of vehicle or other combustible materials will finish after 30 min, stop and cool. In this step, emission rate reduces and temperature reaches to 133°F. Cool emission step for 48G cylinder is about 91 min and for 48Y cylinder is 206 min. (time is more for 48Y cylinder, because it has more capacity)

Second method is similar to the previous method by this difference that, it is assumed cylinder has deficit from the beginning (hydraulic damage isn't formed by pressure). This method is based on air plane collide with cylinder that first collide happens. The initial emission rate is zero at the beginning, but it will increase by time increase. When emission in cooling step happens after 30 min, conditions will be similar to the first method.

Summary of 6FLRE and SUBLIME model results for the first method for 48G and 48Y cylinders are shown in Tables 5 and 6. Related data to emission rate and temperature are observed in Tables 7 and 8.

wind rate of 4m/s. for F stability state, roughness length was 10cm, friction rate ( $u_*$ ) was 0.02 m/s and 5m length was considered for Monin-Obukhov. The boundary height was considered 15m in this condition. For D stability state, friction rate was



considered 0.32 m/s, Monin-Obukhov length was about 150 m, and the length of temperature

inversion was considered 500m. The summary of these assumptions are shown in Table 7:

**Table 6.** Summary of SUBLIME model results.

	Thermal degradation		Primary deficit	
	48G	48Y	48G	48Y
Fire destroyed until the end of time, min	17.8	6.1	30	30
Sublimation value at the time of the fire, lb	2975	924	4240	3213
Steam temperature at the end of the fire, ° F	912	882	912	905
Sublimation steam after the fire, lb	1192	2670	1192	2733
Release time after the fire, min	91.4	205.7	91.4	201\6.1
All UF <sub>6</sub> released, lb	8004	9610	5432	5946
% UF <sub>6</sub> primary	(28.6)	(34.3)	(19.4)	(21.2)

**Table 7.** Summary of climate conditions

Condition	Wind speed m / s	Friction speed m / s	Length, m (Monin-Obukhov)	Mixing height M
Stability F	1	0.02	5	15
Sustainability D	4	0.32	-150	500

#### EMISSION STEP BEHAVIOR

In this step, emission step behavior includes hydraulic damages and emission in fire then cooling step. For each emission explained in previously, several cylinders are involved, but it is assumed that each one act individually and so each cylinder condition must be studied individually. As it was described previously, UF<sub>6</sub> react immediately after emission and produce HF and UO<sub>2</sub>F<sub>2</sub> after hydrolysis. These materials with surrounding air produce flotation mixture.

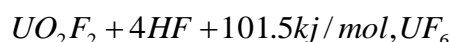
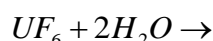
#### Emission by resulted hydraulic damages from heat

As it was mentioned previously, 48Y and 48G cylinders put in fire after 12 and 24 min will be damaged. As it was mentioned previously, products of each damaged cylinder are formed individually and each one will move separately.

While cylinder is broken, UF<sub>6</sub> will emit as solid, liquid, and gas. UF<sub>6</sub> will change to vapor and solid for reducing pressure. The obtained vapor reacts with the present water vapor as a mixture of air-UF<sub>6</sub> and hydrolyzes the products. Movement and rotation of these materials depend on mass concentration and its flotation. Main reasons that play role in mass flotation will be as following:

- UF<sub>6</sub> is a very condensed gas at standard temperature and pressure (relatively about 14 kg/m<sup>3</sup>). This density is so effective on flotation and reduces it. Density increase and concentration accumulation near ground are observed without increase in temperature while UF<sub>6</sub> emission or mass temperature reduction.

- Heat will enter to the initial mass, because UF<sub>6</sub> reaction with water vapor is an exothermic reaction:



Two mole water is needed for each UF<sub>6</sub> mole in this reaction. Water vapor for this reaction is provided by the obtained gases from combustion or environment air permeation inside UF<sub>6</sub> mass. It must be mentioned that the provided water vapor by combustion gases will be negligible in comparison to the vapor in air, because the produced vapor from combustion is about 1 kg/S.

- the obtained gases from combustion having high temperature around cylinders and finally mixing with UF<sub>6</sub> mass increase mass temperature and consequently add on flotation state.

It is observed in each resulted combustion that about 50 to 75 m<sup>3</sup> air in the initial seconds of emission are mixed with each m<sup>3</sup> UF<sub>6</sub> gas and penetrates in UF<sub>6</sub> mass. It is assumed in this article that each 75m<sup>3</sup> air penetrates in 1 m<sup>3</sup> of UF<sub>6</sub>. Of course, this assumption will be so conservative, but since the transferred concentration by wind will increase penetration, it is a good assumption for other safety considerations. If penetration amount is considered low, it means mass is condensed in this time and will be precipitated on ground. Then, ratio is considered 75 to 1. Temperature and flotation will be determined by writing a mass and enthalpy balance. Enthalpy changes will be resulted by the following cases in calculation flotation:

- Penetration of combustion gases

- Hydrolysis reaction
- Sublimation of solid to vapor UF<sub>6</sub>

The assumptions in 48G tank calculations are listed in following. Similar assumptions can be considered for 48Y cylinder:

- Total emitted UF<sub>6</sub> is 1744 kg. This amount is 143 kg vapor in 828K and remained 1601 kg was liquid. This liquid will change to the solid-vapor mixture immediately in 330K. Finally, 75.5% of total input mass of emission will be vapor and the rest will be solid.
- In the initial seconds of combustion, 500m<sup>3</sup> combustion air enters to mass in 1475F and also 10300m<sup>3</sup> environment air with 300K. This amount of entering combustion air to UF<sub>6</sub> mass is relatively 4 seconds volume of combustion by combustive material.
- Hydrolysis reaction produces 101.5 kJ/mol heat; therefore, 502886 kJ heat will be produced by this assumption that the mentioned reaction happens in initial seconds after emission.
- Changing 24.5% of UF<sub>6</sub> that was emitted in soiled form to gaseous form with energy consumption of 47.6 kJ/mol of solid UF<sub>6</sub>.
- According to assumptions, mass will express as a cylinder with 16m radius and height. The volume and temperature of this mass includes HF-air-UO<sub>2</sub>F<sub>2</sub> mixture about 344K and 1.138 kg/m<sup>3</sup>. (density of surrounding air is 1.174 kg/m<sup>3</sup>)

#### *Emission in fire*

After hydraulic destruction of tank, UF<sub>6</sub> is sublimated and changes to vapor. The produced vapor changes to HF and UO<sub>2</sub>F<sub>2</sub> quickly and enters to fire. The entering mass to fire is floated and the following assumptions are considered for this step:

- Fire diameter of 5m
- Fire temperature of 1075K
- Froude number of 0.8

The negative flotation of UO<sub>2</sub>F<sub>2</sub> in comparison to high flotation of combustion will be neglected. (Fire produces 100m<sup>3</sup>/s combustion gas) combustion air flotation is about 0.75 and according to UF<sub>6</sub> emission velocity in fire, it will be 1.7 kg/s, therefore, the flotation of the produced mixed mass reaches to 0.74 for heavy particles of UO<sub>2</sub>F<sub>2</sub> that has 1% reduction than the previous state.

#### *Emission in cooling duration*

After fire stopping, UF<sub>6</sub> emission continues permanently like emission from cold cylinder. This emission is called "cooling-down" step. The mentioned emission for 48G and 48Y cylinders will takes time 91 and 205 min.

## DISCUSSION AND CONCLUSION

The only important radioactive materials in uranium enrichment plant are Uranium hexafluoride, uranium tetra-fluoride and uranium fluoride. The both liquid and gaseous forms of UF<sub>6</sub> that are usually with its solid form, will react with water and water stream immediately in adjacency to them and produce HF reaction. Of this reaction is done in gaseous form and in specific high temperatures, it will be fast and spontaneous. The most important issue about HF is its reaction and mixture ratio with water. It must be mentioned that this molecule can solve in any ratio in water and doesn't have any restriction about ratio of solving in water.

This article studied the methods of UF<sub>6</sub> emission in environment and contaminations were analyzed in all conditions in various intervals. The obtained summaries are in the following Table. First an accident is described that 48G cylinder was fired and damaged hydraulically, and F and D stability conditions are mentioned. In the next parts, the related summary of results is stated in other emission methods. For 5 emission methods, the statements are mentioned as T1 to T5 in Table 8 and letters "a" and "b" shows results for UO<sub>2</sub>F<sub>2</sub> and HF.

The emitted mass by fire in high temperature and also affluent combustion gases has high flotation and rising. This mass won't contact with ground, while the resulted emission in cooling step make small mass contact ground.

According to the mentioned issues, the dominant conditions on UF<sub>6</sub> emission in enrichment plants that are damaged in accidents are described, and it must be noticed that all described results and formulas in text are experiment and were introduced by references and were tested by assumptions. The experiments and UF<sub>6</sub> hydrolysis products measurement were so difficult and need specific measuring instruments that are not accessible for public, and they can just be adapted to the obtained results by scientists according to the condition of plants of Iran and formulas to test their accuracy.

The permitted values necessary for personal health that are in contact with the emitted materials are shown in Table 8. The mean duration that was determined for toxicity is 1-60 second. As it was discussed, the worst conditions to combine average time and concentration are mentioned in wind direction as follows.

**Table 8.** Five proposed accidents specifications

<i>statement</i>	<i>Accident type and involving materials</i>
T1a	Vehicle fire, full of three-cylinder type 48G , $UO_2F_2$
T1b	Vehicle fires, three-cylinder full of 48G, HF
T2a	Vehicle fires, three-cylinder full of type 48Y, $UO_2F_2$
T2b	Vehicle fires, three-cylinder full of type 48Y, HF
T3a	Aircraft collision and fire, two cylinders of the type 48G, $UO_2F_2$
T3b	Aircraft collision and fire, two cylinders of the type 48G, HF
T4a	Aircraft collision and fire, two cylinders of type 48Y, $UO_2F_2$
T4b	Aircraft collision and fire, two cylinders of type 48Y, HF
T5a	Vehicle fires, two cylinders waste, $UO_2F_2$
T5b	Vehicle fires, two cylinders waste, HF

**Table 9.** Toxicity values for HF and  $UO_2F_2$

Average time Min	<i>HF</i>		<i>UO<sub>2</sub>F<sub>2</sub></i>	
	Potential irreversible effects (mg/m <sup>3</sup> )	Potential adverse effects (mg/m <sup>3</sup> )	Potential irreversible effects (mg/m <sup>3</sup> )	Potential adverse effects (mg/m <sup>3</sup> )
1	57	5.7	1600	520
2	57	5.7	780	260
3	57	5.7	520	170
4	57	5.7	390	130
5	57	5.7	310	100
6	52	5.2	260	86
7	48	4.8	220	74
8	45	4.5	190	65
9	42	4.2	170	58
10	40	4	160	52
11	38	3.8	140	47
12	37	3.7	130	43
13	35	3.5	120	40
14	34	3.4	110	37
15	33	3.3	100	35
16	32	3.2	97	32
17	31	3.1	91	30
18	30	3	86	29
19	29	2.9	82	27
20	28	2.8	78	26
21	28	2.8	74	25
22	27	2.7	71	24
23	26	2.6	68	23
24	26	2.6	65	22
25	25	2.5	62	21
26	25	2.5	60	20
27	24	2.4	58	19
28	24	2.4	55	18
29	24	2.4	54	18
30	23	2.3	52	17

**Table 10.** Central line UO<sub>2</sub>F<sub>2</sub> concentrations in wind direction for T1 and F1 stability conditions

<i>Space In wind direction, m</i>	<i>Density (Mg / m<sup>3</sup>)</i>	<i>Average time (Sec)</i>	<i>Increase the scope irreversible effects</i>	<i>Greater than the range of harmful effects</i>
does not exist	does not exist	3600	0.1	100
does not exist	does not exist	3600	0.07	150
does not exist	does not exist	3600	0.04	250
does not exist	does not exist	3600	0.07	305
does not exist	does not exist	3600	0.2	350
does not exist	does not exist	3600	0.29	368
does not exist	does not exist	3600	0.78	450
does not exist	does not exist	3600	1.6	550
does not exist	does not exist	3600	2.6	650
does not exist	does not exist	3600	4.7	750
does not exist	does not exist	3600	6	805
does not exist	does not exist	3600	6.6	840
does not exist	does not exist	3600	6.4	850
does not exist	does not exist	3600	7.5	920
does not exist	does not exist	3600	7.9	1210
exist	does not exist	3600	8.7	1350
exist	does not exist	3600	9.2	1450
exist	does not exist	3600	14	2414
exist	does not exist	3600	13	4023
exist	does not exist	3600	10	6532
exist	does not exist	3600	8.8	7241
does not exist	does not exist	3600	5.1	12068

**Table 11.** Central line HF concentrations in wind direction for T1 in F1 stability conditions

<i>Space in wind direction, m</i>	<i>Density (Mg / m<sup>3</sup>)</i>	<i>Average time (sec)</i>	<i>Increase the scope irreversible effects</i>	<i>Greater than the range of harmful effects</i>
does not exist	does not exist	3600	0.026	100
does not exist	does not exist	3600	0.018	150
does not exist	does not exist	3600	0.011	250
does not exist	does not exist	60	0.46	305
does not exist	does not exist	60	1.8	350
does not exist	does not exist	60	2.8	368
does not exist	does not exist	60	6.8	450
does not exist	does not exist	60	13	550
does not exist	does not exist	60	16	650
does not exist	does not exist	60	30	750
does not exist	does not exist	60	33	805
does not exist	does not exist	60	35	840
does not exist	does not exist	60	36	850
does not exist	does not exist	60	41	920
does not exist	does not exist	60	28	1210
exist	does not exist	60	28	1350
exist	does not exist	60	26	1450
exist	does not exist	750	15	2414
exist	does not exist	1200	8.5	4023
exist	does not exist	1800	4.6	6532
exist	does not exist	2100	3.2	7241
does not exist	does not exist	3600	1.3	12068

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