Detection of Source of Low Mass Peaks in UF_6 Gaseous Sample Using Quadrupole Mass Spectrometer

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(Received 20th February, 1993)

Summary: To determine the source of low mass peaks (0-45 amu) in gaseous Uranium Hexafluoride sample, a qualitative study was made using a Quadrupole Mass Spectrometer QMS 420 equipped with SEM and UHV facilities. The experimental findings indicates that some of these low molecular weights peaks are part of the sample and few fragments of hydrocarbons are added from oil of the diffusion pump. NO^+, a product of the ion source is also detected. Many of these peaks also interfere with each other. An attempt is made to determine the origin of these interfering species in the spectrum.

Introduction

The level of impurities in industrial materials plays an important role in determining the quality and usefulness of the final products. For such materials, trace impurities analysis is performed prior to processing and to check the level of impurities. Uranium hexafluoride used in enrichment is one of the above mentioned material. Its impurity levels should be known and kept under specified limits is very crucial for the nuclear industry [1].

A programme was started to determine the impurities present in the uranium hexafluoride sample. Qualitative determine was made using Quadrupole Mass Spectrometer. A companion paper [2] describe the results of the study made in high mass region. In the present manuscript we have investigated qualitatively the source of low molecular weight peaks in gaseous uranium hexaflouride sample to establish:

i) these low mass peaks are part of uranium hexaflouride, or

ii) these traces are transferred to the analyzer of the instrument from the oil of the diffusion pump.

Results and Discussion

Comparing the results obtained in the study under different experimental conditions (Table 1) and Figure 2. We restrict our discussion specifically on the appearance of low mass peaks in the spectrum and their variations in intensity.

Table 1: The peaks obtained at different amu using different experimental conditions.

m/z	Experimental Conditions					
		1	2	3 (mv)	4	5
2	H_2^+	700	700	830	800	700
9.5	F^{2+}	-	-	70	60	-
12	C^+	60	60	200	120	-
14	N^+	60	70	67	80	-
15	CH_3^+	50	20	20	40	-
16	O^+	400	100	200	200	60
17	OH^+	150	43	40	30	-
18	H_2O^+	100	60	60	58	20
19	F^+	730	600	1000	1000	700
20	HF^+	62	70	5000	5000	600
28	N_2^+, $C_2H_4^+$, CO^+	600	800	1000	1000	600
30	NO^+	10	-	-	-	-
32	O_2^+	80	250	100	350	75
40	Ar^+, $C_3H_4^+$	-	35	20	40	-
44	CO_2^+	80	70	200	200	90

1. H_2^+, HF^+, F_2^+

Main source of hydrogen ion in the UF_6 sample is HF and H_2O. Study Table 1 and comparing the results of experimental conditions No.2 and 3 an increase in intensity of hydrogen peak is observed when the analyzer side is connected with the empty expansion tank. This indicates the presence of HF in the tank. Supporting evidence came from the simultaneous increase in the intensity of HF^+, F^+ and F_2^+ (Table 1 and figure 2). It is expected that HF which is

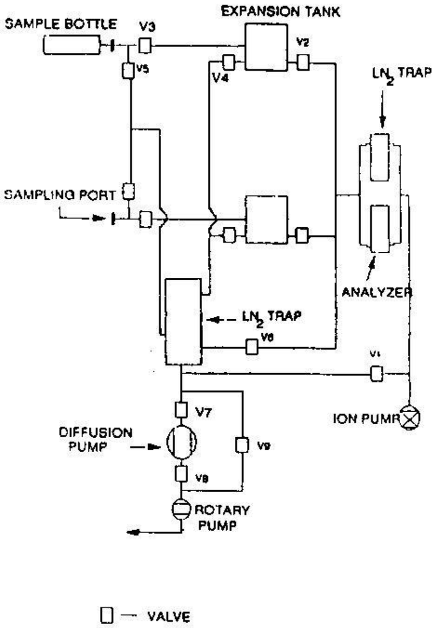

Fig. 1: Flow diagram of Quadrupole Mass Spectrometer QMS 420 (reproduced from ference 11).

a part of the system was adsorbed on the walls of the expansion tank.

The F_2^+ peak at 9.5 m/z (mass to charge ratio) further support our argument that HF is present in the expansion tank of the QMS 420 instrument in the pre-adsorbed state. It is well documented in the literature that adsorption of gases on metals is a physical and the chemical process [3]. It could be noticed that F_2^+ peak does not appear in the study using experimental condition no. 1 and 2. This peak could be seen under experimental condition 3 and 4, where HF^+ intensities are considerably higher. It appears that UHV conditions in the analyser and high HF concentration are the possible cause. This promotes the formation of doubly ionized species, hence resulting the appearance of F_2^+ peak in the spectrum. In experimental condition No. 5 when the sample was introduced in to the analyser, HF peak intensity dropped as compared to experimental conditions 3 and 4. This prohibits the formation of F_2^+.

2. C^+

The main source of carbon in the sample studied is the oil of the diffusion pump and carbon dioxide from background. Studying experimental conditions 1 and 2; a constant intensity at amu 12 is observed. In cases 3 and 4 where empty expansion tank is connected with analyzer, an increase of 2-3 fold in the peak height is observed. We assume that oil vapors from the diffusion pump enter the expansion tank through back streaming during the evacuation of the gas handling section. This oil evaporates under high vacuum condition and reach analyser section. Another support to this argument comes from the study of experimental conditions No. 5, where the introduction of uranium hexafluoride in the analyser through the expansion tank resulted in diminishing of C^+ peak in the spectrum. Here the sample is kept at 0.3 mbar tank pressure and at such a pressure the oil vapors deposited on the wall of the tank do not vaporize to reach the analyser [2].

3. N_2^+, N^+, NO^+

Traces of nitrogen are usually present in the mass spectrometer background resulting in the appearance of N_2^+ and N^+ peaks. On the other hand NO^+ could be a result of recombination process between background oxygen and nitrogen [4]. Various ions are formed in the analyser section by ionisation in the ion source by electron impact [5] and new species form by recombination of these ions. The low level of $^{15}N_2$ peak intensity may also be contributing to this peak.

4. CH_3^+

This is a very common hydrocarbon contamination which is present in almost all the metals, alloys and in gaseous samples produced through chemical process [6]. The presence of CH_3^+ peak could be due to the diffusion pump oil vapors which reach the ion source [7,8].

5. H_2O^+, OH^+, O_2^+, O^+

All these low mass peaks are mainly the product of oxygen and water vapors in the system. These water vapors are introduced during sample changing. In experimental condition No. 5 a reduction in the peak intensities (Table 1) of these ions are observed which we attribute to the vigorous chemical reaction of water vapors with uranium hexafluoride [9].

$$UF_6 + 2H_2O \longrightarrow 4HF + UO_2F_2$$

6. CO_2^+, CO^+

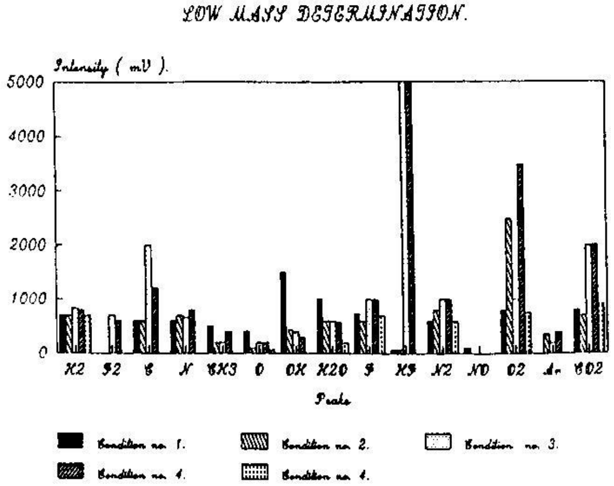

Fig. 2: UF_6 spectrum obtained under different experimental conditions.

The data indicates that CO_2 is present in the background but its intensity is increased upon connecting the empty tank to the analyser. The increase in intensity is an indication that CO_2 and oil adsorbed on the expansion tank wall, reaches the ion source.

7. Ar^+, $C_3H_4^+$

The peak at m/z 40 is due to Ar^+ and a large fragment of oil $C_3H_4^+$ [10]. We suggest that main ion responsible for this peak is the oil fragment. The data also suggest that this ion further fragments and finally disappear when all the valves leading to the analyser section are closed.

Experimental

A Quadrupole Mass Spectrometer QMS 420 equipped with U.H.V and SEM facility was used for the study (Figure 1). The natural uranium hexaflouride sample was collected in a sample bottle from the feed cylinder using the following procedures.

The sample bottle was mounted with the prepared sampling system which contains a vacuum pump, a flowmeter (for measuring the gas glow), a baratron and a pirani gauge for monitoring the pressure inside the port. The system was connected with the main feed cylinder, evacuated to 2^*10^{-2} mbar pressure. The sampling bottle was dipped into the liquid nitrogen. The controlled flow of gas from the feed cylinder was collected in the sampling bottle at the liquid nitrogen temperature. About 2.0 gms gas was collected. The system was the flushed with dry Argon. Any light gases that might have entered during sampling were removed from the sample at -70°C. The sample bottle was removed and mounted to QMS 420 instrument for analysis (Figure 1). The instrument was calibrated for the most intense UF_5^+ peak. All the data then reference to this peak.

The data was collected in the following sequence:

1) background spectrum with ion pump (value v_1 closed).
2) background spectrum with diffusion pump and ion (value v_1 and v_7 open).
3) residual spectrum of the empty expansion tank after evacuation to 10^{-7} mbar pressure (value v_1, v_7 and v_2 open).
4) residual spectrum of the empty expansion tank with diffusion pump connected (value v_1, v_7 and v_2 open).
5) sample scan using a computer program from amu 0-45 with the expansion tank filled with gas (value v_1 closed, v_2 open).

The spectrum obtained in the study is presented in figure 2 and in Table 1.

Conclusion

The results obtained from the study indicates the possible sources of low molecular weights peaks in the uranium hexaflouride. On the basis of the experimental findings it could be concluded that:

(i) CO_2 and N_2 are part of UF_6 gaseous sample.

(ii) Low molecular weights hydrocarbon species such as $C_3H_4^+$, $C_2H_4^+$, CH_3^+ and C^+ originates from the diffusion pump oil vapors.

These species enter the system during evacuation.

(iii) NO^+ is neither a part of sample nor comes from oil but form by the recombination process in the analyzer.

(iv) HF is present in UF_6 sample and its peak height indicates other contribution as well from the analyser system.

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