ANALYSIS OF BY-PRODUCTS OF N_2–SF_6 GAS MIXTURES SPARKED UNDER INHOMOGENOUS FIELD CONDITIONS

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1. INTRODUCTION

A vast data on formation of by-products due to arc and spark decomposition of SF_6 is available in literature [1-10]. But experimental work relating to N_2 – SF_6 gas mixtures is limited. In view of increasing application of gas mixtures, it is essential to understand the nature of by-products formed in gas mixtures. Many recent studies have highlighted the advantages of using N_2-SF_6 gas mixtures as a replacement to SF_6 gas for High Voltage application, with a view to reduce emission of SF_6 gas into the atmosphere. Majority of these studies are aimed at determining the basic characteristics of N_2-SF_6 gas mixtures. However, this study attempts to understand the nature and quantum of different species formed in N_2-SF_6 mixtures in the presence of insulating spacers when sparked under inhomogeneous field conditions.

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2. GC-MS APPARATUS AND THE METHOD

For the determination of N_2 - SF_6 gas ratio, a gas chromatograph (GC) was used and for identification of the species formed during the discharge processes, the Mass Spectrometer (MS) was used. The GCMS analysis was carried out with HP model 5890 series II equipment. Throughout the studies, Thermal Conductivity Detector (TCD) and Mass Selective Detector (MSD) were used. Analysis of sparked SF_6, SF_6-N_2 gas mixture and SF_6-N_2 gas mixture with six different spacer materials have been carried out and reported in this paper. A needle - plane electrode configuration was used and the by-products formed due to spark discharges under these conditions have been compared. The duration of sparking was extended beyond 24 hours depending upon the nature and concentration of by-products formed and their time-dependence. The electrode gap distance for gas gaps was 10 mm and the discs of spacer material of this thickness were inserted into the gap for studying the influence of different materials like PTFE, Polyester, Polypropylene (PP), Nylon, Cycloaliphatic resin and Polymethyl-methacrylate (PMMA).

3. BY-PRODUCTS WITH SF_6 AND N_2/SF_6 GAS MIXTURES

In case of pure SF_6 gas the abundance of SF was 2.4% while SF_4 accounted for 1.75%. Even the percentage of SF_2 was comparatively less, about 2.33. Further, SF_3 is seen to the extent of 14% much higher than all other species like SF, SF_4 and SF_2. On the other hand, SF_4 is also very significant by its presence and its abundance is 6.2%. These results are repeatable and the variation in relative abundance (%) is within ±3%. Thus under non-uniform field conditions the most significant by-product formed is SF_3.

Table 1. Relative abundance of by-products formed after different durations of sparking

Species	Duration of sparking		
	4 hours	6 hours	10 hours
SF	2.4	7	4.7
SF4	1.75	1	4.4
SF2	2.33	6.4	4.4
SF3	14	30	18
SF4	6.2	11	7

With the continuation of sparking at 30 kV for 6 hours it will be interesting to see if there is any increase or otherwise in the concentration of the constituents formed. This result is shown in table 1. SF is observed to the extent of 7% while SF_4 is present marginally at 1%. SF_2 is significant and has a relative abundance of 6.4%. The presence of SF_3 is significant since its relative abundance is as high as

30%. On the other hand, SF_4 is seen to the extent of 11% and SF_3 complex has a relative abundance of 65.4%.

With the continuation of sparking for 10 hours, the presence of SF_5 is seen in larger proportion and the presence of SF_3 is significant in addition to SF_2. The abundance of SF is 4.7% and of SF_2 is 4.4%. On the other hand, SF_3 is significant with the relative abundance percentage of 18.11%. In addition to these ions, complexes involving SF_4 account for 0.8% and even SF_4 is also present and its percentage is 6.8. SF_4 complex is observed in a very low concentration of 0.9%.

4. BY-PRODUCTS IN 80:20 N_2 - SF_6 GAS MIXTURES

In this case, SF and SF_2 have a relative abundance of 4% each after 10 hours of sparking at 30 kV under point-plane field configuration. In fact, there was no difference in the species and their relative concentrations after 6 and 10 hours of sparking at 30 kV. As in the earlier cases, SF_3 has the highest relative abundance of 17.45% and that of SF_4 is 7%. Complexes involving SF_2, SF_3 and SF_4 or similar species are observed in relative concentrations varying from 1 to 4.75 %. Thus the comparison of by-products of SF_6 and SF_6 - N_2 gas mixtures show similar trends with SF_3 being the major constituent.

5. BY-PRODUCTS WITH CYCLOALIPHATIC RESIN

In this case, SF has a relative abundance of 4.3%, SF_4: 3.1% and SF_2: 4.09%. However, SF_3 is a major constituent with a relative abundance of 22%, followed by SF_4 with 8%. Complexes involving SF_4 are seen to the extent of 0.81 %; those involving SF_3 are about 4%. These complexes are also significant when sparking is continued for 8 hours. After 12 hours of sparking there is a slight increase in abundance of SF_5 (5.5%), SF4 (4.7%), SF_2:(4.38%), SF_3 (23.25%), and SF_4 (8%). Complexes of SF_3 and SF_5 are less than 1%. Thus the presence of CAR does not result in formation of new species as compared to the results in SF_6 and SF_6/N_2 gas mixture (20:80). Thus CAR is observed to be stable and does not contribute any new species when sparked in N_2-SF_6 gas mixture.

6. BY-PRODUCTS WITH POLYESTER

After 6 hours of sparking with polyester, it is observed that all the species reported earlier are formed are in a very low concentration. With 12 hours of sparking, there is a significant change in the concentration of various constituents formed. After 16 hours of sparking, there is a further decrease in relative concentration of different species. Complexes involving SF_5 (lower), SF_5 (higher), SF_4+SO_2F and SF_5NF_3 are also observed in a very low percentage.

Though these complexes are present, they do not seem to increase in concentration with duration of sparking.

7. BY-PRODUCTS WITH NYLON

In presence of nylon, in 80:20 N_2 - SF_6 gas mixture the relative abundance of SF is 7.6%, SF_4 6%, SF_2 6%, SF_3 27%, SF_4 9.35% respectively. The complex ions involving SF_3 has a relative abundance of 1.53%, SF_5 (lower): 1% and SF_5 (higher): 4.6%. Thus the trend seen in N_2 - SF_6 mixture is also evident in nylon. The presence of SF_3 to the extent of 27% is a significant factor, which is observed in other cases, except polyester.

The sparking was continued for 12 hours and the relative abundance of SF was 5%, SF_4^{++} is 3.8%, SF_2 is 4.36%, SF_3 is 21.8% and SF_4^+ 8%. Though SF complexes are observed, their concentrations are significantly low (less than 1%) except SF_5, which is 3.8%. Also present in the gas mixture, though to a very low extent is SF_3NF [8], which was not observed in presence of cycloaliphatic resin spacer or for that matter in SF_6 and N_2SF_6 gas mixture.

8. BY-PRODUCTS WITH POLYPROPYLENE

Even in case of PP, SF has a relative abundance of 3%, SF_4: 2.7%, SF_2: 3.12%, SF_3: 19%, SF_4^{++}: 7.75%. Though several complexes involving SF_2, SF_3, SO_2F_{10}, SO_2+SO_2F, $S_2O_3F_6$, SF_5+SF_5 etc. are formed, their concentration is insignificant. Thus PP appears to be quite stable and major by-products formed are almost identical.

9. BY-PRODUCTS WITH PMMA

With PMMA material after 6 hours of sparking in 80:20 N_2/SF_6 gas mixtures, the relative abundance of SF in this case is 2.7%, and SF complex is 2.45, SF_4 is 2.42, SF_2^+ is 2.77, SF_3 is 17 % and SF_4^+ is 7%. Species like $S_2O_3F_6$ and SF_4+SOF_2 though observed are in very low concentration.

The sparking was continued for 10 hours and SF content is observed to increase to 3.88, SF_4 to 3.55, SF_2 to 3.5. Thus on an average SF, SF_4 and SF_2 have a relative abundance of 3.7%. Even SF_3 content increases to 20% while SF_4 increases to 8%. SF_5 complexes also increase to 4.5%, which was not observed earlier.

With further increase in the sparking duration to 16 hours, SF concentration goes up further to 4.35%, SF_4 to 3.9% and SF_2 to 3.8%. Thus all the species show increase in relative abundance as compared to the results of sparking for 8 hours. In case of SF_3, increase in relative abundance (%) is from 20 to 21.65% when sparking duration is increased from 10 to 16 hours and SF_4 remains almost constant at 8%. Even concentration of SF_5 complex is almost constant at 4.3%.

10. BY-PRODUCTS WITH PTFE

SF_6 - N_2 (20:80) gas mixture was sparked in presence of PTFE disc at 30 kV initially for a period of two hours. Because of the popularity of this material for many applications, it was decided to monitor closely the by-products formed from the initial stages. In this case, SF has a relative abundance to 2.6%, SF_4 0.9 %, SF_2 2.8 %, SF_3 15.64 %, and SF_4^+ 6.5 %. In addition HN_3O_3, C_4NO_3, $C_4H_2NO_4$, $C_3H_2N_2O_4$, and SF_5NF_3 are also formed. Some of these fragments are peculiar to PTFE. Thus SF_3 is a major constituent, in addition to SF_2, SF, and SF_4. Similar results are also obtained after 6 hours of sparking. After 8 hours of sparking, species like HN_3O_3, C_4NO_3, $C_4H_2NO_4$ and $C_3H_2N_2O_4$ are absent but SF_5NF_3 is present in a very low concentration.

With the sparking being continued for 10 hours, the relative abundance of SF was 4.37%, SF_4: 3.88%, SF_2: 2.7 % and SF_4: 7.58%.

11. CONCLUSIONS

The following are the important conclusions of this study:

(1) In pure SF_6 gas, SF percentage increases gradually with sparking from 4 hours to 6 hours, but from 6 hours to 10 hours, there is an indication of recombination and its percentage goes down. On the other hand even SF_4 seems to show the same trend and its percentage increases initially but tends to decrease with continuous sparking. SF_3 is always formed with the highest relative abundance and a decrease in its concentration is evident as the duration of sparking is increased. Similar results are also seen with SF_6– N_2 gas mixture.

(2) With the CAR, SF_3 is a major constituent of the spark by-products followed by SF_4. With sparking of longer duration, the abundance of SF_5 increases.

(3) With PE, SF_3 is again a major constituent of the by-products followed by SF_4. The presence of SF_4+ SOF_2 points towards formation of more such complexes or fragments with further increase in duration of sparking.

(4) With the increase in duration of sparking, N_2 has a tendency to inhibit the formation of certain by-products, seen in SF_6 and SF_6 - N_2 gas mixtures.

12. ACKNOWLEDGEMENTS

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13. REFERENCES

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