CONF-940671--1

Effect of a solid insulator on the spark yield of S_2F_{10} in SF_6.

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Abstract — Because of its toxicity S_2F_{10} production in SF_6 discharges has been the focus of intensive study in recent years. In previous work we have examined the yield of S_2F_{10} for corona, spark and arc discharges and, in sparks, the effects of water and oxygen have been studied systematically. Here we report the influence of an insulating solid spacer on the production of S_2F_{10} in SF_6 when subjected to spark discharges in the energy range 1.6-43 J/spark at a gas pressure of 1 atm. Analyses of the sparked gases were performed using a cryogenic enrichment gas chromatography technique with a sensitivity of less than 10 ppb (parts-per-billion) or 1 in 10^8. With this sensitivity S_2F_{10} can be detected after one or just a few spark(s), minimizing secondary effects of gas contamination, electrode erosion and insulator damage that can influence S_2F_{10} production and make quantitative yield determinations difficult to establish. For these studies Teflon was used as the spacer sandwiched between two stainless steel electrodes. Energy measurements were made after recording the voltage and current waveforms. The amount of S_2F_{10} produced per unit energy deposited into the discharge was found to be greater in the presence of the spacer than for a purely gas gap. Factors which influence S_2F_{10} stability such as surfaces and water will also be discussed.

INTRODUCTION

Formation of S_2F_{10} by electrical discharges in SF_6 has been the subject of extensive study over the past few years. This highly toxic SF_6 byproduct has been detected in corona, spark and arc discharges [1,2]. In recent work we have examined the influence of oxygen and water on the S_2F_{10} yield in spark discharges [3]. In those studies it was found that either oxygen or water addition to SF_6 results in decreased S_2F_{10} production. This is attributed to competitive reactions of O from O_2 or H_2O, or OH from H_2O, with the SF_5 radical, a precursor to S_2F_{10} formation. Reactions 1-3 are listed to illustrate some of the likely pathways for SF_5, some of which leads to S_2F_{10} and S_2OF_{10} production.

$$SF_5 + SF_5 \longrightarrow S_2F_{10} \quad (1)$$
$$SF_5 + O \longrightarrow SF_5O \quad (2)$$
$$SF_5O + SF_5 \longrightarrow S_2OF_{10} \quad (3)$$

Other factors believed to influence S_2OF_{10} production include electrode condition and surface-adsorbed species such as moisture. Electrode conditioning may in fact be mainly a problem of desorption of adsorbed contaminants. In an effort to explore further the influence of various factors affecting S_2F_{10} production, and approach conditions occurring in practice, we have examined the effect of an insulating spacer on the spark yield of S_2F_{10}.

EXPERIMENT

Spark discharges were produced in a 1.1-ℓ stainless steel chamber housing a pair of stainless steel electrodes, one fixed and one moveable. Two electrode geometries were used, sphere-plane and plane-plane. In either case a Teflon disk was positioned between the electrodes, with the electrodes in contact with the Teflon. For comparison spark experiments were made without the Teflon disk but at the same interelectrode gap. Because of the numerous factors that influence byproduct formation, it was desirable to compare the byproduct yield with and without the insulator, without opening the chamber or altering the conditions of the gas or the chamber. This was accomplished in one series of experiments by positioning the chamber, so that the electrode axis was horizontal, and moving the adjustable electrode to permit the insulator to fall. Sparks made with the gas gap were then compared to the insulated gap. Fig. 1 shows the spark chamber and energy measurement system, which has been previously described [4]. Spark energies were in the range 1.6-43 J per spark for experiments with the Teflon insulator and 7-80 J per spark for gas gap breakdown experiments. Spark energy was varied by varying the gap spacing, which in turn changes the breakdown voltage. For gas gap breakdown

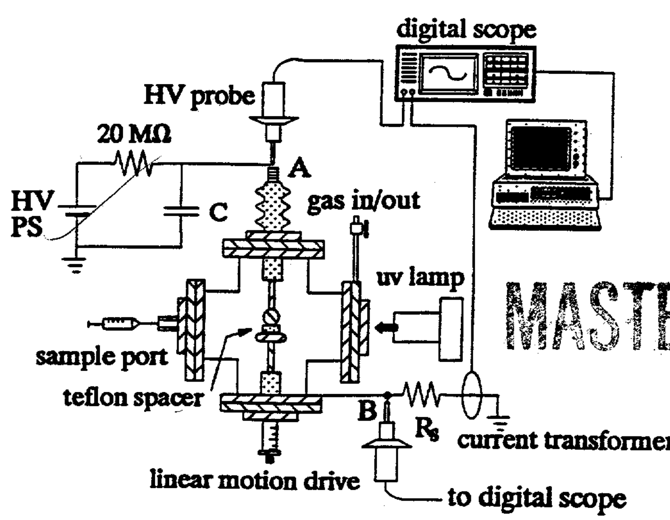

Fig. 1 Schematic of spark cell and measurement system.

DISCLAIMER

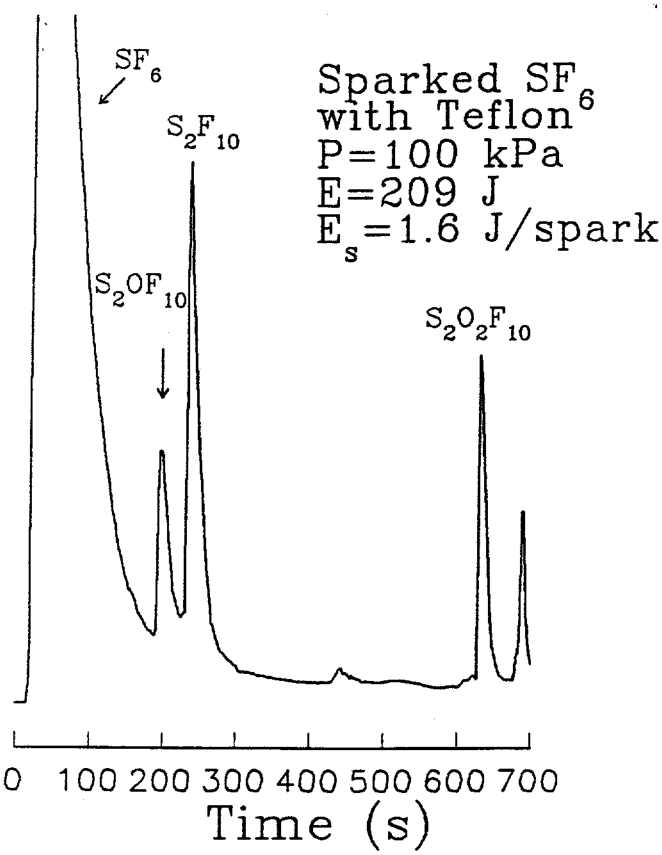

Figure 2. Typical chromatogram of sparked SF_6.

the S_2F_{10} yield was found to be independent of the energy per spark above 7 J per spark [3].

After sparking, the gas was sampled by injecting the needle of a syringe through a port containing a septum. The gas sample, typically 2-ml in volume at the sparked gas pressure of 1 atm, was then injected into a cryogenic enrichment gas chromatography system which has also been described previously [3-5]. A typical chromatogram is shown in Fig. 2, for sparked SF_6, indicating the formation of S_2F_{10}, S_2OF_{10}, and $S_2O_2F_{10}$. This system, permitted the detection of S_2F_{10} down to concentrations below 10 ppb (parts per billion) in SF_6. The range of concentrations reported in these measurements were 10-600 ppb.

Results and Discussion

The increase in concentration of S_2F_{10} (in ppb) for 3.6 J sparks in SF_6 is shown in Fig. 3 over the range 0-250 sparks. After about 50 sparks the S_2F_{10} concentration increases more rapidly with number of sparks. The yield increases from 1.2 x 10^{-11}mol/J for less than 50 sparks to 3 x 10^{-11}mol/J for greater than 50 sparks. In Fig. 4 is shown the increase in S_2F_{10} production (in ppb) with number of sparks (up to 30 sparks) for four different spark energies, 3.6, 4.4, 14.9, and 43 J per spark. The 4.4 J data was made with sphere-plane geometry while the other spark energies in Fig. 4 were made with plane-plane geometry. From linear fits to the data, the production of S_2F_{10} (in ppb per spark) is plotted in Fig. 5 as a function of spark energy, showing a linear increase with spark energy over this range of energies. The yield (in mol per joule) is therefore relatively constant in this energy range.

A comparison of S_2F_{10} production with and without the Teflon spacer is shown in Fig. 6 for an experiment made at 14.9 J per spark where the Teflon spacer is dropped as described in the experimental section. Decreased production is indicated in the gas gap as compared to the insulated gap. In another series of experiments at 3.6 J per spark, the gas was analyzed after 75 sparks were made in two separate experiments, one with and one without the Teflon spacer. Again there was a factor of two increase in S_2F_{10} yield when the insulator was present. Comparison of S_2F_{10} production with and without the spacer is shown in

Table 1. S_2F_{10} production with and without Teflon spacer.

Energy per spark (J)	Sparks	With spacer (ppb)	Without spacer (ppb)	Method
3.6	75	264	120	sm
14.9	30	178	118	dm

sm: separate measurements; dm: spacer intentionally dropped during experiment.

Table 2. Spark yield of S_2F_{10} with and without Teflon spacer at various spark energies.

Spark Energy (J)	Yield (10^{-11}mol/J)	
	With Spacer	Without Spacer[b]
1.6	57	--
2.4	14.6	--
3.6	4.4	--
4.4	3.2	--
7.0	--[a]	1.2
14.9	1.9	--
20	--	1.0
30	--	2.1
43	2.0	--
80	--	1.7

[a] Yield not determined at this energy; [b] Yields from Reference 3.

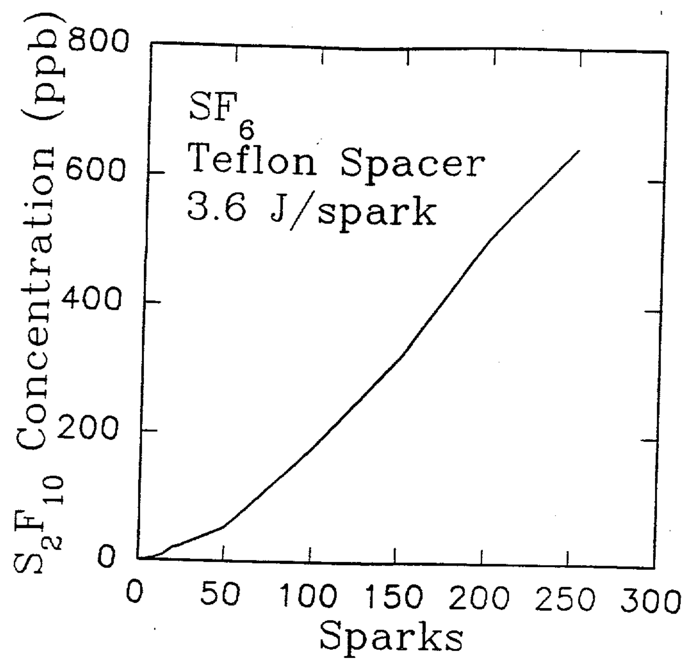

Figure 3. S_2F_{10} concentration (in ppb) as a function of number of sparks for sparked SF_6 at 3.6 J across an insulating spacer.

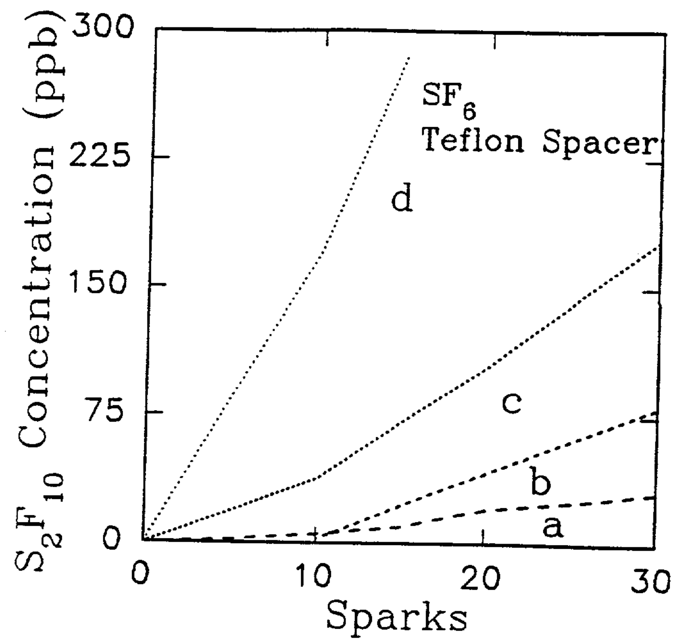

Figure 4. Production of S_2F_{10} (in ppb) as a function of number of sparks in sparked SF_6 across an insulating spacer for (a) 3.6 J, (b) 4.4 J, (c) 14.9 J, and (d) 43 J.

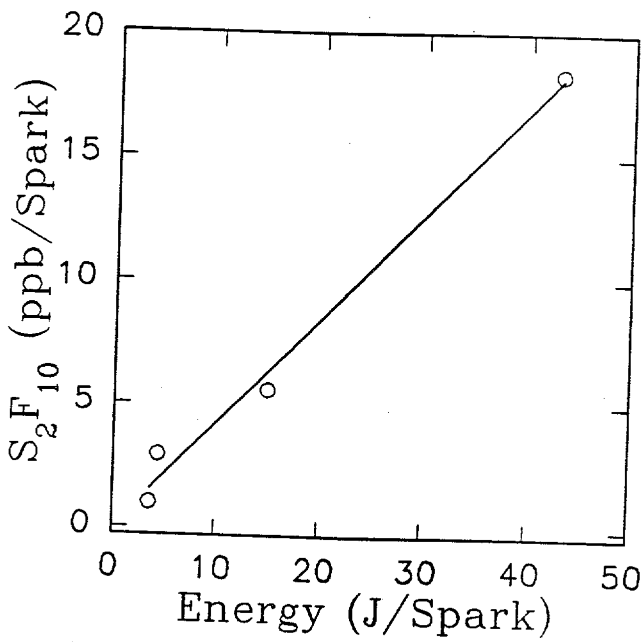

Figure 5. Production of S_2F_{10} (in ppb per spark) as a function of spark energy.

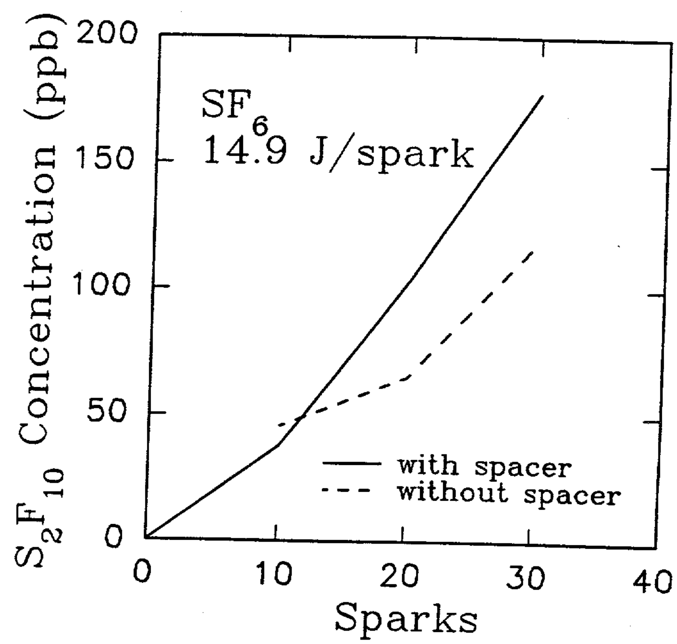

Figure 6. Comparison of S_2F_{10} production (in ppb) with and without the Teflon spacer. Teflon was removed from the interelectrode gap during the experimental run (see text).

Table 1.

In Fig. 7 we show the S_2F_{10} yield as a function of spark energy for spark energies in the range 1.6-43 J per spark, showing increased production at low energies. At 1.6 J the S_2F_{10} yield reaches the highest value observed, 57×10^{-11} mol/J. S_2F_{10} yields at various energies are summarized in Table 2 for spark discharges across a Teflon spacer. Also shown in the table are previous results of gas gap breakdown at various energies. It is speculated that as the energy per spark decreases the discharge becomes cooler, resulting in increased S_2F_{10} production, due to the increased S_2F_{10} thermal stability at lower temperatures [6,7].

SUMMARY

S_2F_{10} yield as a function of spark energy has been measured for spark breakdown of SF_6 across an insulating spacer, in this case Teflon. The yield with the spacer was found to be higher by about a factor of two over the gas gap at 3.6 and 14.9 J spark energy. The yield was

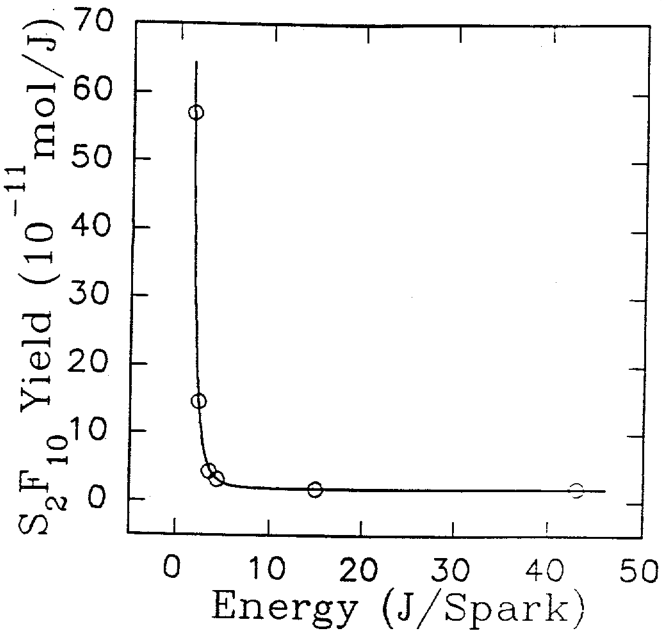

Figure 7. Spark yield (in 10^{-11}mol/J) of S_2F_{10} as a function of spark energy (in J/spark) with Teflon spacer.

relatively independent of spark energy for spark energies above 5 J per spark, having a value of about 2 x 10^{-11}mol/J. At lower energies the yield was found to increase substantially with decreasing spark energy, to 57 x 10^{-11}mol/J at 1.6 J.

ACKNOWLEDGEMENTS

Work was supported by a Cooperative Research and Development Agreement with the Bonneville Power Administration, the Tennessee Valley Authority, the Electric Power Research Institute, the Canadian Electric Association, Empire State Electric Energy Research Corporation, Ontario Hydro, Hydro Quebec, the National Institute of Standards and Technology, and the Department of Energy Office of Energy Management under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

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