50X1-HUM

INFORMATION REPORT INFORMATION REPORT

CENTRAL INTELLIGENCE AGENCY

C-O-N-F-I-D-E-N-T-I-A-L

COUNTRY USSR

REPORT

50X1-HUM

SUBJECT Expert Appraisal of Soviet Paper Entitled "Acoustic Coagulation of Vapor Containing Fluorine Compounds", by M L Varlamov and Collaborators (WHICH EXPERIMENTS WITH ULTRASONICS)

DATE DISTR. 3 Jan 63

NO. PAGES 2

REFERENCES

50X1-HUM

DATE OF INFO.

50X1-HUM

PLACE & DATE ACQ.

THIS IS UNEVALUATED INFORMATION

50X1-HUM

a translation of an

X unpublished article in Russian entitled, in English, "Acoustic Coagulation of Vapor Containing Fluorine Compounds" by M L Varlamov, Ye L Krichevskaya, A A Ennan, L M Kozakova and G A Manakin, Chair of Technology and Automation of Chemical Processes of the Odessa Polytechnical Institute

50X1-HUM

2. This work deals with laboratory scale experiments for the precipitation of fluorine effluents from the exhaust gases of a superphosphate granulator. The fluorine effluents were present both as aerosol and vapors. The flow rate treated in a standing waves tube averaged one CFM. The sound source was a modified Hartman whistle previously developed by the authors (see Use of Ultrasound in the Investigation of Matter, Proceed of OPI, 1960) for sulfuric acid coagulation with ultrasonics. The emission frequency was 16.5 KC and an average intensity level of 153-155 decibels was reached in the agglomeration pipe despite the use of a diaphragm in front of the generator. It was found that sound waves increase the precipitation yield from between 42-69 percent (without irradiation) up to 80-95 percent. The fluorine content oscillated between 0.6 and 4.5 gr/m^3 values which in general correspond to a very good load for sonic processing. As expected collection efficiency went up with increasing particles concentration and humidity (15 to 120 g/m^3). The best contact time averaged 3-4 seconds as expected from literature data.

3. Direct experimentation conducted with the exhaust gases of a superphosphate granulator demonstrated that with a three second contact time the precipitation yield reaches 87 percent when dealing with an inlet fluorine content as low as 0.18 gr/m3. In other words, the exhaust gases contained only 0.025 gr/m^3 which is a safe exit concentration according to Public Health Standards. The authors will continue this work with the idea to later develop an industrial gas cleaner.

50X1-HUM

C-O-N-F-I-D-E-N-T-I-A-L

GROUP 1 Excluded from automatic downgrading and declassification

50X1-HUM

STATE		ARMY		NAVY		AIR		FBI		AEC			

INFORMATION REPORT INFORMATION REPORT

CONTROLLED NO DISSEM ABROAD NO FOREIGN DISSEM

4. [redacted] the weak point of this investigation lies in the power requirements of the sound sources (which are not mentioned). Feasibility of the technique has been clearly established, but the bottleneck for future extrapolation will be the development of a high power economical sound source.

50X1-HUM

-end-

Acoustic Coagulation of Vapor Containing Fluorine Compounds*

By

M. L. Varlamov, Ye. L. Krichevskaya,
A. A. Ennan, L. M. Kozakova and
G. A. Manakin

Chair of Technology and Automation of Chemical Processes of the Odessa Polytechnical Institute

In the production of simple and granulated superphosphate there is a precipitation of fluorine compounds which are not completely trapped by the usual absorption devices. The exhaust gases of these processes contain 0.1 to 2.0 g/m^3 of fluorine /1-3/ , which is considerably in excess of the permissible health safety norms. Therefore, a need arises to devise new methods of fine purification of gases from fluorine compounds.

During the production of simple superphosphate, fluorine precitpitates from the reaction chambers and mixers, mostly in the form of SiF_4; further interaction with water causes the formation of H_2SiF_6 and silica gel /4/. The for-

*) First Report.

mation of a film of silica gel on the surface of droplets of the absorbing liquid impedes the further absorption of fluorine compounds /5/.

In simple superphosphate the fluorine is in the form of fluosilicic acid and salts containing fluorine. The equilibrium of pressure of vapors on fluorine compounds depends not only on the temperature and concentration of H_2SiF_6, but also on the content of free phosphorous acid in the liquid phase of superphosphate /6/.

During granulation of superphosphate precipitation of fluorine takes place mainly in the drying process, with 10 to 20 percent of the fluorine introduced with the simple superphosphate being transformed into the gaseous phase /1/.

According to the data of one of the investigations conducted at a superphosphate plant by us, no more than 1/3 of the fluorine is trapped in the absoprtion turret and gas pipes out of this amount; the discharge of fluorine into the atmosphere constitutes tens of kilograms per hour.

In the drying drums precipitation of fluorine takes place at increased temperatures. Subsequent decrease of temperature causes water vapor to condense, accompanied by the formation of a stable and slowly settling vapor. According to bibliographical data /2, 5, 6/, and our own observations, it contains the major portion of fluorine

compounds which are present in exhaust gases. The pressure of vapor of fluorine compounds on diluted solutions of H_2SiF_6 is insignificant, therefore on reaching a state of equilibrium, most of the fluorine is transformed into the liquid phase of the aerosol. Thus, at temperatures between 50 and 70°C, gas in a state of equilibrium contains only 0.02 to 0.04 g/m^3 fluorine /7/. The remaining quantity should be in the form of vapor. The transition into the liquid phase takes place fairly rapidly on account of the large specific surface of the aerosol.

Thus, the task of removal of fluorine is reduced to devising methods of fine purification from exhaust gases and vapor containing H_2SiF_6 and other fluorine compounds.

A number of investigations were dedicated to problems of absorption of fluorine compounds by water or by weak fluosilicic acid in nozzle and bubbling apparatus /8-13/. The use of Venturi tubes and other high-intensity apparatus makes possible a considerable increase of the rate of absorption of fluorine compounds; however, it is difficult to solve the problem of complete purification of gases by these methods, because fluorine is mostly present in the form of vapor which is poorly retained in absorption apparatus.

Fine purification of gases from vapor containing fluorine compounds can be accomplished either by electro-

filters [1, 14], or by the method of acoustic coagulation. A marked increase of collisions of particles of the aerosol takes place as a result of the effect of a powerful sonic field. The vapor rapidly consolidates, and can then be easily removed by regular dust extractors or other mechanical precipitators [15-21]. Acoustic coagulation successfully occurs at frequencies of 1 to 25 cps and an intensity of 0.1 to 1.0 W/cm^2.

Powerful sonic fields in gaseous media are produced by gas-jet generators: static and dynamic sirens. The method of acoustic coagulation is particularly important in these instances, where it is necessary to accomplish a fine purification of gases from finely-dispersed aerosols.

The purpose of this work is to investigate the acoustic coagulation of vapor containing fluorine compounds, and to study the possibility of using this method of purifying the exhaust gases of superphosphate production from fluorine.

Experimental Part

Method of investigation. The first stage of this work was to investigate acoustic coagulation under laboratory conditions. The initial aerosol was obtained by mixing water vapor with tetrafluoride of hydrogen, which precipitates under the action of concentrated sulfuric acid into a mixture of equal volume of silica sand and

commercial fluosilicate of natrium. Before the experiment the gas was diluted with air in a gas meter to a concentration of the order of 0.1 to 1.0 g/m^3, which is close to manufacturing conditions. Mineral oil of low viscosity was used as sealing liquid. Water vapor was obtained by mixing hot air saturated with steam with cooler air. Temperature stability in the gas mixture at the input of the sonic tube was provided by a thermoregulator. For a sonic generator we used an improved gas-jet generator GS-2 of the Hartman whistle type, employed by us in investigating the acoustic coagulation of sulfuric acid vapor [22].

The second stage was the investigation of acoustic coagulation of fluorine-containing vapor directly from the exhaust gases of the granulated superphosphate department.

A diagram of the apparatus is shown in Fig. 1. The investigated gas was pumped from the exhaust pipe 1 by vacuum pump 2 and transferred to sonic treatment tube 3 which has a diameter of 45 mm and a length of 960 mm. The front end of the tube, tightened with a rubber diaphragm, joined the sleeve of the gas-jet generator 4. The amount of gas passing through the sonic treatment tube was measured by rheometer 5. Part of the gas was removed for analysis by means of suction apparatus 6 and 7, both before, and after sonic treatment. The fluorine was absorbed with distilled water in a system of four Drexel jars 8 and 9.

Part of the gas was withdrawn into a continuous ultramicroscope 10, be means of which we calculated the number of particles in a unit of volume before and after acoustic coagulation. The condensate which accumulated in the sonic treatment tube in droplets was removed through tap 11. The temperature was measured by thermometers 12. Air was pumped into the sonic generator under pressure of 2 kg/cm^2 by compressor 13 through oil separator 14 and receptacle 15. The frequency of sound was measured by means of oscillograph E0-7 to which tension was transferred from a titanate-barium probe, which served as receiver of sound, and generator of electric oscillations ZG-10. The parity of frequencies was derived from Lissajous figures observed on the screen of the oscillograph. Sound intensity was determined with the help of a titanate-barium probe, connected to the amplifier and cathode voltameter.

The selection of method of analysis of gases for fluorine content is of essence /23-25/. In the control of production of superphospahte and in scientific research the most widespread method is to absorb fluorine in water with subsequent titration of the absorbing liquid with alkali according to two indicators. The alkalimetric method was used by us in the first series of laboratory tests in working with pure gases. This method is not suitable for analysis of plant exhaust gases, since along with fluorine

compounds, water absorbs superphosphate dust containing phosphorus acid and SO_2 and CO_2 from combustion gases. Therefore, analysis by the alkalimetric method gives results which are several times overstated.

In our investigation we used the colorimetric method based on the alizarin-zirconium reaction [26-28]. It is based on a weak pink coloring of the alizarin-zirconium complex in the presence of small quantities of fluorine. Colorimetric analysis was conducted with chemically pure solution of NaF by the standard scale method. Control analyses were conducted in the central laboratory of the Odessa Superphosphate Plant and in the analytical laboratory of the NIUIF by the method of low-temperature removal of fluorine with subsequent titration by a solution of thorium nitrate.

The humidity content of the gas was determined by means of absorption with calcium chloride and phosphorous anhydride. During the laboratory experiments splashes and large particles of vapor were deposited in traps. Under manufacturing conditions the content of splashes and vapor was determined by trapping them with absorbent cotton.

The control of the process of acoustic coagulation was also conducted by means of microphotography.

Fig. 1. Diagram of a device for the investigation of acoustic coagulation of vapor containing fluorine compounds from exhaust gases of the superphosphate granulation department.

1 - exhaust pipe; 2 - vacuum pump; 3 - sonic treatment tube; sonic generator; 5 - rheometer; 6, 7 - suction apparatus; 8, 9 - Drexel jars ; 10 - continuous ultramicroscope; 11 - condensate tap; 12 - thermometers; 13 - compressor; 14 - oil separator; 15 - receptacle; 16 - wires to titanate-barium probe.

Fig. 2. Sonic pressure along the axis of the sonic treatment tube.

Fig. 3. Effect of duration of sonic treatment on degree of purification of gases from fluorine.

Fig. 4. Effect of initial concentration of fluorine on degree of purification.

Results of Experiments

In order to determine the nature of the sonic field we took a number of measurements of the intensity of sound along the axis of the sonic treatment tube. The results are shown in Fig. 2. Due to the echo from the rear end part of the tube, a system of vertical waves is established in the tube. In the front part (up to 60 mm) the sonic field is distorted by conditions at the intake and by the presence of a nozzle for removal of the gas. The maximum sonic pressure is observed at a distance of 190 to 235 mm from the diaphragm; further damping of oscillations along the tube is insignificant. The mean sonic pressure along the axis of the tube corresponds to the level of intensity of 155 decibels.

We determined the effect on acoustic coagulation under laboratory conditions of the following: duration of sonic treatment, humidity content and initial concentration of fluorine at temperatures of 50, 55 and 60°C. The humidity content was controlled by changing the ratio between the expenditure of hot and cold humidified air.

In Fig. 3, we cite data of the relation between the degree of purification of gases from fluorine and the duration of sonic treatment, with humidity being constant (at a temperature of 55°C and the ratio between cold and

hot air $\frac{V_c}{V_h}$ = 10. The initial concentration of fluorine was 0.6 - 1.7 g/m^3. As can be seen from Fig. 3, the duration of the order of 4-5 seconds is sufficient for acoustic coagulation of vapor, ensuring a degree of purification from fluorine between 75 and 78 percent without additional mechanical precipitators. Further exposure of vapor to the sonic field provides an insignificant increase of the degree of purification. Analogous curves were obtained by us earlier for vapor of sulfuric acid [2].

Shown in Fig. 4 are data on the relation between the degree of purification and the initial concentration of fluorine. Experiments were made without sonic treatment and with sonic treatment of 3 to 4 seconds duration. In the area under investigation (0.6 to 4.5 g/m^3) purification is more complete with an increase of the concentration of fluorine. Without sonic treatment the precipitation of fluorine is 42 to 69 percent; with sonic treatment the degree of purification increased to 80-95 percent, depending on the humidity of the gas.

The degree of purification substantially changes with an increase of the weight concentration of vapor and steam. In Fig. 5 data are shown on the relation between the degree of purification and general humidity content of the gas. An increase of the latter from 15 to 120 g/m^3 causes an increase of the concentration of vapor and an increase

of the separation of fluorine from 73 to 95 percent.

Fig. 5. Effect of the general humidity content on the degree of purification from fluorine.
A - degree of purification (%), B - humidity content (g/m^3).

Fig. 6. Effect of duration of sonic treatment on the degree of purification from fluorine of the exhaust gases of the superphosphate granulation department.

A - degree of purification (%), B - duration of sonic treatment (sec).

Thus, laboratory experiments have shown that it is possible to use the method of accustic coagulation to purify industrial gases from fluorine compounds.

Expenditure of gas (l/min)	Duration of sonic treatment (sec)	Concentration of fluorine in gas (g/m^3) Initial	Final	Degree of purification (%)
Without Sonic Treatment				
		0.040	0.036	10.00
Following sonic treatment at a frequency of 16.5 cps				
Mean				

The results of the second stage of investigations of acoustic coagulation of fluorine compounds, conducted with exhaust gases of the superphosphate granulation department are shown in the Table. It follows from the data of the Table that for practical purposes the duration of sonic treatment of the order of 3 seconds is sufficient; this causes a precipitation of 87 percent of the fluorine contained in the gas. At a mean concentration of fluorine of 0.18 g/m^3 following sonic treatment of 3 seconds shows a remainder of 0.023 g/m^3 which is below the recommended health norm (0.03 g/m^3). The relation between the degree of purification of cases and duration of sonic treatment is shown in Fig. 6.

It should be noted that at the industrial department of superphosphate granulation acoustic coagulation occurred more completely than with gases of laboratory equipment. This can apparently be explained by the higher humidity content of the gas, as well as by the presence of center of condensation (specks of dust) which facilitate the formation of vapor.

In some experiments, along with the determination of degree of purification, we also made analyses of the cendensate from the sonic treatment tube. The fluorine content in the condensate varied from 0.7 to 8.5 g/l, depending on the initial concentration of fluorine in

the gas.

Determination of the number of particles in a unit of volume and their distribution according to size was made by means of a continuous ultramicroscope and microphotography.

Conclusions

1. In laboratory investigation of acoustic coagulation of vapor containing fluorine compounds (at the level of the intensity of sound of the order of 153 to 155 decibels and frequency of 16.5 cps) it was found that the action of sound increases the degree of purification from between 42 and 69 percent (without sound treatment) to between 80 and 95 percent.

2. The completeness of purification depends in large measure on the concentration of vapor in the gas. An increase of the total humidity content from 15 to 120 g/m^3 causes an increase of the degree of purification from 73 to 95 percent. With a constant humidity content the degree of purification increases with the increase of the initial concentration of fluorine in the gas and duration of sonic treatment.

3. Investigations were conducted of acoustic coagulation of vapor containing fluorine compounds from exhaust gases of the superphosphate granulation department. It

was found that with sonic treatment of 3 seconds duration there is a precipitation of 87 percent of fluorine contained in exhaust gases. With an initial concentration of fluorine of 0.18 g/m^3, its content, following acoustic coagulation, is reduced to a magnitude of the order of 0.025 g/m^3, i.e., below the recommended health norm.

4. The cited results were obtained in the sonic treatment tube alone, without additional mechanical precipitators. If the latter are used, the degree of precipitation of fluorine compounds will be even higher.

BIBLIOGRAPHY

/1/ I. M. Boguslavskiy and L. D. Kiosse, Soobshcheniya o na chno-tekhnicheskikh rabotakh NIUIF (Reports of Scientific-Engineering Works of the Scientific Research Institute of Fertilizers and Insectifuges), 2, 54 (1957).

/2/ H. Sh. Safiullin, Khim. prom. (Chemical Industry) 6, 77 (1959).

/3/ G. K. Shilin, Sbornik annotatsiy nauchno-issledovatel'skikh rabot NIIOGAZ za 1956-1957 gg. (Symposium of Annotations of Scientific Research Works of the Scientific Research Institute of Gas Purification for Industry and Sanitation for the Years 1956/1957).

/4/ K. E. Lunde, Ind.Eng.Chem. 50, 3, 293 (1958).

/5/ A. L. Whynes, Inst. Chem. Eng. Graduates & Students Section, Feb. 19 (1954).

/6/ A. I. Shereshevskiy, L. S. Goritskiy and L. N. Potapova, Soobshcheniya o nauchno-tekhnicheskikh rabotakh NIUIF, 2, 26 (1957).

/7/ V. V. Illarionov, T. I. Sokolova, S. Z. Kulagina

and I. G. Knyazeva, Soobshcheniya o nauchno-tekhnicheskikh rabotakh NIUIF, 2, 48 (1957).

/8/ I. M. Boguslavskiy, V. M. Ramm and V. I. Matrozov, Soobshcheniya o nauchno-issledovatel'skikh rabotakh NIUIF, 6-7, 10 (1958).

/9/ I. M. Boguslavskiy and V. M. Ramm, ibid., 41,
/10/ Ya. V. Shvartsshteyn and V. M. Ramm, ibid., 55.

/11/ V. I. Matrozov and V. M. Ramm, ibid., 102.

/12/ V. I. Matrozov, V. M. Ramm and G. M. Yusova, ibid. 108.

/13/ I. M. Boguslavskiy and V. M. Ramm, Soobshcheniye o nauchno-tekhnicheskikh rabotakh NIUIF, 2, 49 (1957).

/14/ I. Zaring and G. Popova, ZhPKH (Zhurnal Prikladnoy Khimii - Journal of Applied Chemistry), 14,866 (1936).

/15/ L. Bergman Ul'trazvuk (Ultrasonics) 1956.

/16/ A. E. Crawford, Ul'trazvukovaya tekhnika (Ultrasonic Engineering) 1956.

/17/ R. T. Hueter and R. H. Bolt, Sonic Techniques for Use of Sound and Ultrasound in Engineering and Science, N.Y. (1955).

/18/ Ye. A. Mednikov, Primeneniye ul'traakustiki k issledovaniyu veshchestva (Use of Ultrasound in Investigation of Matter) OPI, 3 (1956).

/19/ B. F. Podoshevnikov, Vestn.tekhn. i ekon.inform. (Herald of Technical and Economic Information), 1, 26 (1957); 4, 4 (1958); 4, 31 (1959).

/20/ R. M. G. Boucher, Genie Chimique,77, 6, 163 (1957); 78, 1, 14 (1957).

/21/ M. L. Varlamov, Ye. L. Krichevskaya, G. A. Manakin, L. M. Kozakova and A. N. Gospodinov ZhPKh XXXIII, 1, 14 (1960).

/22/ M. L. Varlamov, G. A. Manakin, A. N. Gospodinov, and Ye. L. Krichevskaya, Primeneniye ul'traakustiki k issledovaniyu veshchestva. Sb. trudov OPI (Use of Ultra-

sound in the Investigation of Matter. Symposium of works of OPI, 1960).

/23/ Ftor i yego soyedineniya (Fluorine and its Compounds) vol.2 (1956).

/24/ F. E. MacKenna, Nucleonics, 8, 6 (1951); 9, 1, 2 (1951).

/25/ M. M. Vinnik, Dissert. kand. (Candidate's Dissertation), NIUIF (1958).

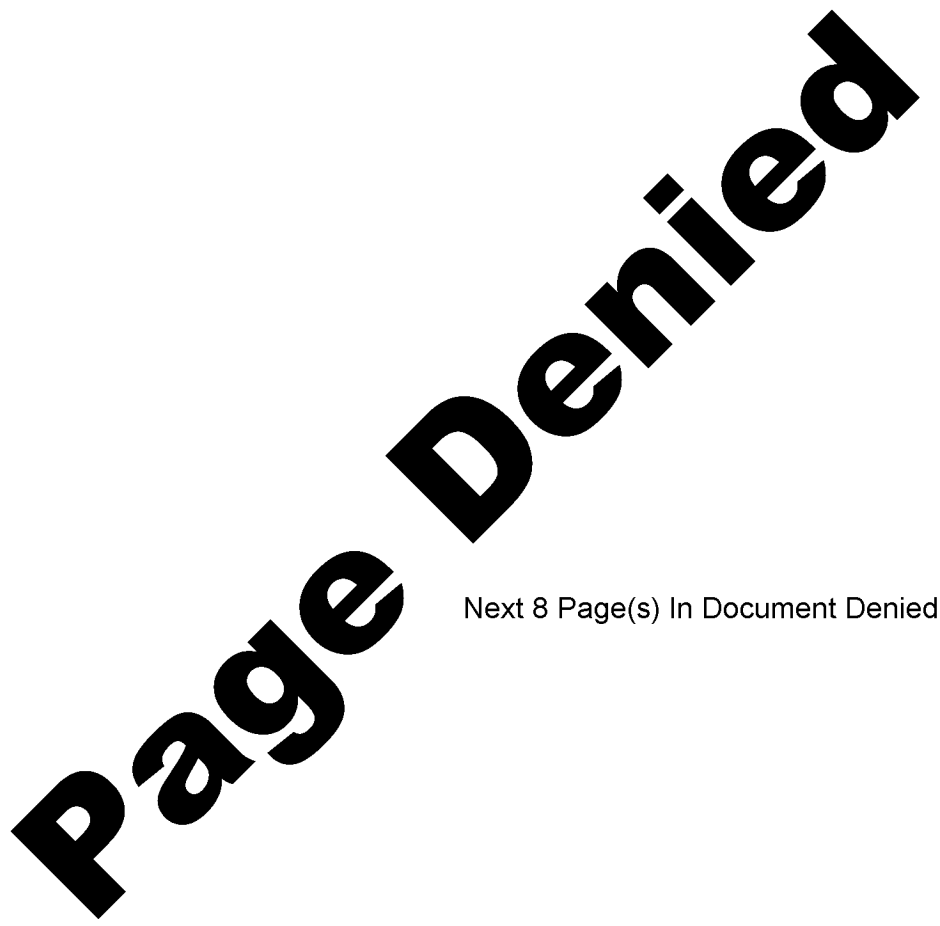
/26/ H. E. Bumsted and G. E. Wells, J.Analyt.Chem., 24, 1595 (1952).

/27/ R. Vigier, Bl.Soc.Chim.France, 2, 160 (1957).

/28/ K. B. Khait and K. V. Lyashcheva, Sb. "Voprosy ozdorovleniya superfosfatnogo proizvodstva" (Problems of making Superphosphate Production healthier), 1960.

50X1-HUM

1169



Next 8 Page(s) In Document Denied