Environmentally Safe Microwave Heating of Non-Polar Materials

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Abstract

The current research presents results for the theoretical basis of heat transfer and experimental studies of a microwave method of heating uranium hexafluoride for safe evaporation from containers, including defective, and low- and medium-temperature heating of oil fractions. The process is environmentally safe because heat generation occurs in the bulk of the heated medium and is easily controlled. The proposed method is based on uranium hexafluoride evaporation at the expense of heat energy transferred from substances absorbing microwave energy. The conducted investigation on a laboratory bench with a direct heating regime of samples by microwave field of a travelling wave of lowest mode H_{10} and in the process studied, samples were heated both individually and in combination with UF_6. The results show that it was determined that uranium oxides are the most preferable for use as heat absorbing agents for the microwave heating process. A demonstration on a pilot scale was designed for testing the feasibility of this process. The dielectric characteristics of two components mixing were studied. The penetration depth of microwave into the mixture and heat-absorbing agent amount for process implementation has been determined. Several experimental tests on the environmentally safe technology at pilot scale installation were conducted at various continuous modes. The results revealed from the pilot unit showed that the microwave method of evacuation of DUF_6 from steel containers allows them to be emptied safely with practically cold ($T<100^0C$) walls, with high efficiency of using microwave energy ($\eta\approx97\%$) while maintaining the level of pressure in the containers.

Paper type: Research paper

Keywords: microwave energy, heat-absorbing agent, heat transfer; non-polar materials, depleted uranium hexafluoride.

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Introduction

Depleted uranium (DU) is a by-product of the enrichment operation of natural or reprocessed uranium. Enrichment plants have been operating since the 1940s and a large amount of depleted uranium has been accumulated as a result of more than half a century's use of nuclear power. Containers with depleted uranium hexafluoride are stored for a long time in open areas and are subject to corrosion. World stocks of depleted uranium totalled about 1.2 million metric tons of elemental uranium and continue to increase (IAEA BULLETIN, (1985), Depleted Uranium Hexafluoride Management Program (2001), Management of Depleted Uranium, (2001)). About 80% of the world's inventories are held in the USA and the Russian Federation (IAEA BULLETIN, (1985), Depleted Uranium Hexafluoride Management Program (2001), Management of Depleted Uranium, (2001), Nikitin *et al*. (2020)). In Russia, the principal amount of DU hexafluoride is stored in 2.5 m³ vertical steel containers, and outside Russia in 4.0 m³ containers (Nikitin *et al*. (2020)). The lifetime of some containers exceeds 50 years. There are defects (deformation, joint leakage, etc.) in some old containers. The evacuation of uranium hexafluoride from such containers as well as the storage may result in accidents and pollution of border territory.

When DU hexafluoride is exhausted into the atmosphere, it reacts with the moisture of the air and forms easily soluble uranyl fluoride (UO_2F_2) and volatile hydrogen fluoride (HF). By accident, about 10% of the total uranium hexafluoride is in gaseous form uranium hexafluoride and the aerosol of the submicron particles of uranyl fluoride. The estimated area of eventual pollution in hypothetic cases of one container (2.5 m^3) destruction at full uranium hexafluoride hydrolysis will be 3.21km^2. The pollution by hydrogen fluoride was calculated for the secondary cloud. After the evacuation and conversion of depleted uranium hexafluoride (DUF_6), depleted metallic uranium can be used in various industries and, first of all, for the manufacture of metal (uranium-steel) containers and heavy concrete for radioactive waste storage (Gotovchikov *et al*. (2005); Gotovchikov *et al*. (2006); Gotovchikov *et al*. (2005)). Currently, uranium hexafluoride is evacuated from containers by heating with an outer source supporting pressure in the container sufficient for UF6 sublimation (Nuclear fuel cycle science and engineering, (2012)). During the process of outer heating, the temperature of the container walls can exceed the melting point of UF6. The density of UF6 decreases upon melting by ~1.4 times (from 5.06g/cm^3 up to less than 3.63g/cm^3), which can result in hydraulic shocks even during sublimation/evaporation from the solid phase with local overheating, for example, with heating of a container bottom. The relevant characteristics, from the point of view of process safety, for UF6 evaporation, are the duration of evaporation from a container, lag of UF6 consumption control, and calorific intensity of the container walls. These characteristics are particularly significant for the process of depleted UF6 (DUF6) evaporation from long-term storage containers. Obvious and latent defects are possible in such containers. Therefore, for safety, the evaporations are conducted at very low rates. The main reason for the poor efficiency is the poor heat transfer from the walls of the container to solid DUF6. In a rather short period (some hours), a gap appears between the wall of the container and solid DUF6, which increases with time. The pressure in a container during sublimation is supported at a low level that does not provide a high rate of evaporation. The unexpected failure of "ex-melting" pieces of solid DUF6 is possible during evaporation that is fraught with accidents and excursions of the technological mode. It is necessary to add to the above as a deficiency, the low efficiency of heating installations of UF6 containers, as heating through a wall is accompanied by large heat losses to the environment due to heat radiation.

An analysis of all factors demonstrates that external heating of containers during UF6 evaporation does not guarantee the safety of the process and it reaches a limiting output. The temperature rise in the container wall by induction heating can reduce the draining cycle by up to 5 days, although at the same power input, according to thermodynamic calculations, the draining cycle can be reduced by up to one day. It is possible to reach such productivity and to secure the process safety during evaporation of DUF6 from long-term storage containers, even defective ones, only by using an internal heat source in a container. This is possible if a microwave electromagnetic field is used as an energy carrier. The optimal version is direct heating of solid DUF_6 by microwave energy without a heat transfer through the wall. However, according to literature data on UF_6 electrophysical properties, it does not absorb electromagnetic field energy in the microwave range to conduct evaporation or sublimation processes. Therefore, this process can be realized by placing a material that absorbs microwave energy on the UF6 surface or by imbuing the UF6 with an applicable liquid so that the heating of solid UF6 will be ensured at the expense of its direct contact with the heated substance (Shatalov (1999); Saranchin (2005); Seyrankaya *et al*. (2006); Grossin *et al*. (2006); Fatykhov *et al*. (2005); Imenokhoyev *et al*. (2013)). An analysis of UF6 electromagnetic properties allows the following to be taken into consideration:

1. Direct heating of solid and liquid UF_6 by microwave energy is most likely ineffective or unfeasible owing to the low values of both dielectric and magnetic permeability and loss factors.
2. Indirect heating of solid UF_6 is made possible by locating solids on its surface or by impregnating it with liquids which absorb microwave energy well.

1 Materials and Methods
1.1 Laboratory study

To realize the proposed method, materials capable of heating with microwave energy and inert in uranium hexafluoride medium must be chosen. According to these limitations, tablets of uranium and iron oxides (ferrites) from solid substances, i.e., heat-absorbing agents, and a series of liquid ethane halocarbons (freons) of the marks 112, 113, 122A can be recommended for the tests. Uranium oxides (UO_2 and U_3O_8) are the most preferred due to their conglomerate form, they have a high thermal capacity and a high density of $\approx$10 g/cm^3 (approximately twice that of UF_6), which should reduce the effect of a «gas cushion» upon contact of the heated tablets with subliming UF6. Being good dielectrics, ferrites, i.e., sintered iron oxides (with special additions), have high magnetic susceptibility and magnetic permeability. This makes it possible to heat them effectively with the magnetic component of a microwave field. The magnetic properties of ferrites, including in the microwave range, are well known but investigations on their application as heat-absorbing agents have not been carried out. Liquid heat-absorbing agents, i.e., halocarbons 112, 113, and 122A, give steady solutions with UF_6 up to 100^0C and permeate well the solid UF6. The solution properties are close to theoretical, which simplifies subsequent separation of the halocarbon from UF_6, if necessary, and recycles it for reuse. Isomers of halocarbon molecules can have dipole moments. It is possible that UF6 solutions in halocarbons can have particular dielectric properties contributing to their heating in a microwave field. There are no data in the literature about such research and they must be obtained

experimentally. A laboratory bench to study the dielectric properties of the above mention materials was developed. The laboratory bench consists of two independent blocks (microwave and technological). The power (microwave) block of the laboratory bench (**Figure 1**) is adjacent to the exhaust hood. The technological block of the laboratory bench (**Figure 2**) is mounted in a fume hood. The laboratory bench is intended for research on the absorption capacity of microwave energy by different heat-absorbing agents (oxides of uranium, ferrites and freons) separately, and in a contact with DUF_6 at heating by microwave known field intensity. Also, it was proposed to use a laboratory bench as a base for the manufacture of demonstrating installation and testing of developed technology at conditions close to real ones. Investigation on the laboratory bench is conducted at the direct heating regime of samples (solid and liquid) by microwave field of a travelling wave of lowest mode H_{10} and in the process studied, samples were heated both individually and in combination with UF_6.
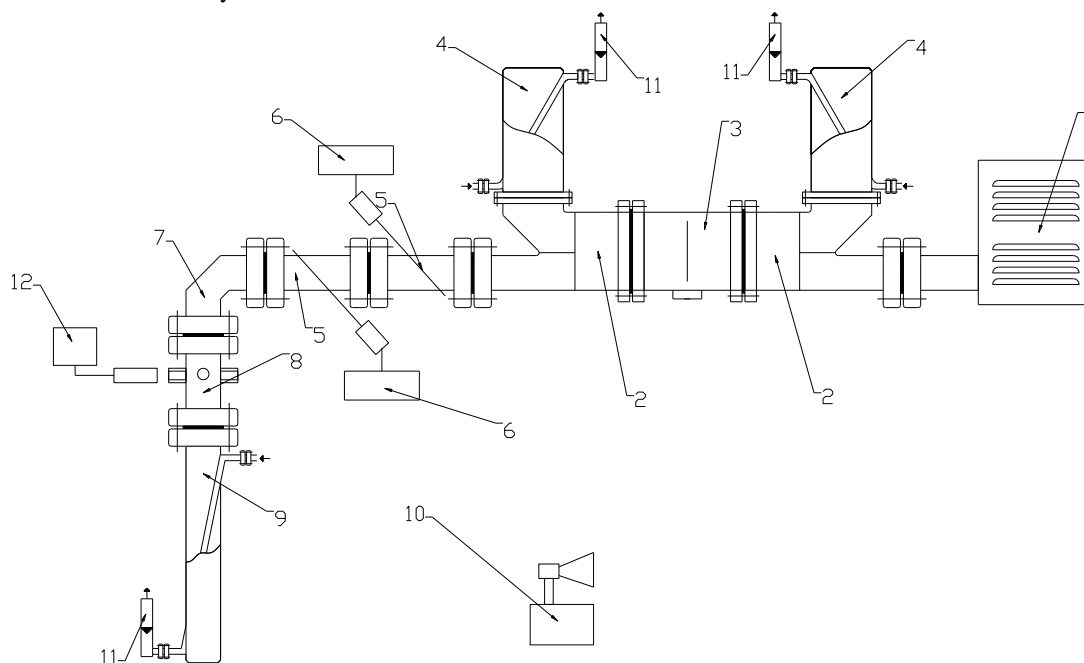

Fig.1 Scheme of the power block. 1- Generator on the base of magnetron with air cooling, 2–slot wave-guide bridges, 3–phase shifter; 4–matched water load; 5–unit for dropping and reflected microwave power meter; 6-wattmeters for dropping and reflected power measurement after attenuator 2; 7-waveguide turn (90^0); 8– working (measuring) unit where a tube with the tested sample is located; 9-terminal load of the waveguide line; 10-a device for microwave radiation measurement over workplaces; 11- power meter; 12– pyrometer.

The laboratory bench was made taking into account the physicochemical properties of uranium hexafluoride and the requirements of the rules for handling it, excluding the release of UF_6 vapours into the atmosphere. The technological unit is carried out taking into account the working conditions with uranium hexafluoride. The test sample, both separately and in a mixture with DUF_6, is placed in a quartz tube. The relative orientation of the electric component of the microwave field and the tube with the studied heat-absorbing agent is shown in **Figure 3**.
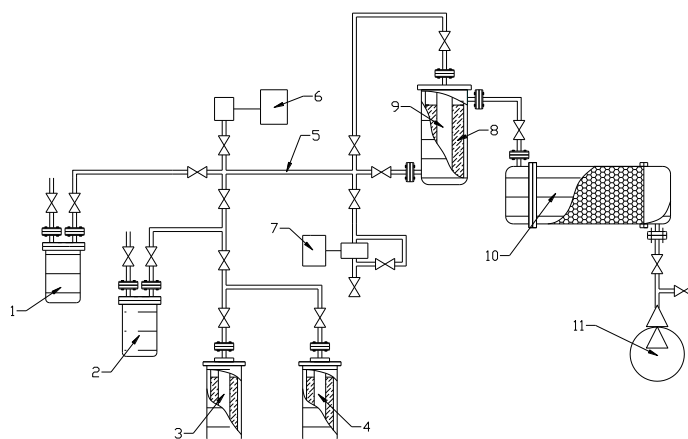

Fig. 2 Scheme of the technological block of the laboratory bench, 1-container with DUF6; 2- receiver; 3–re-condensation tube; 4- working tube; 5-vacuum line; 6-thermocouple vacuum gauge; 7–differential manometer; 8-nitrogen trap; 9-sampler; 10–absorption column; 11-vacuum pump.

1.2 Method for losses factor evaluation on laboratory bench

The heat exchange process in this system is complex and can be divided into heating by the microwave field of heat absorbing agents (Sychev, (1991), Landay *et al*., (1991); Lykov, (1978); Philippov, (1986)) and their heat exchange with the material (DUF$_6$). The heating process of materials with a given heat capacity c and density ρ by microwave energy with intensity **E** is described by the heat transfer equation (Landay *et al*., (1991); Lykov, (1978)):

$$\rho c \frac{\partial T}{\partial t} = \mathbf{0.5} \varepsilon_0 \varepsilon'' \omega \langle \mathbf{E}^2 \rangle \tag{1}$$

When materials are subjected briefly to a microwave field, differential equation (1) can be replaced by the finite difference one:

$$\Delta T = \frac{\mathbf{0.5} \varepsilon_0 \varepsilon'' \omega \langle \mathbf{E}^2 \rangle}{\rho c V} \Delta t \tag{2}$$
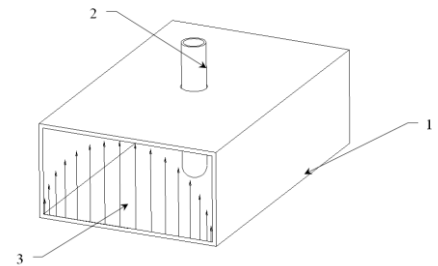

Fig. 3 Scheme of working unit of laboratory bench 1. Section of the rectangular single-mode waveguide; 2. Quartz tube with heated material; 3. Electric field pattern for mode H10.

If the interaction zone of the microwave field and the tested material is organized so that the average intensity E-squared for the period will be given, the dielectric losses ε'' of the material can be calculated from equation (2). For microwave energy acting on the tube with the sample (with a given volume, density and specific heat) (as measured with a wattmeter or calorimetric method) and the given electric field intensity E during a fixed time Δt, the dielectric losses are calculated from equation (2):

$$\varepsilon'' = \frac{c\rho V}{0.5 \varepsilon_0 \omega \frac{E_m^{\,2}}{2}} \cdot \frac{\Delta T}{\Delta t} \tag{3}$$

Electric field intensity *E* was determined as follows. The method is based on measuring the absorption capacity (dielectric loss factor) of the samples of heat absorbing agents (uranium oxides, ferrites, freons) when they are directly heated by a microwave field, the characteristics of which can be measured, and the field structure is set and slightly distorted when a sample is placed in it. The design of the bench allows the sample to be heated in an almost uniform electric field when a quartz tube with a sample is placed in the centre of the waveguide in the middle of a wide wall. The field distribution of a travelling electromagnetic wave in the waveguide is determined by solving uniform wave equations that may be derived from the system of fundamental Maxwell's equations (Landay *et al*., (1991); Sychev, (1991))**:**

$$\frac{\partial^2 \mathbf{E}}{\partial t^2} + \nabla^2 \mathbf{E} = 0 \,; \frac{\partial^2 \mathbf{H}}{\partial t^2} + \nabla^2 \mathbf{H} = 0 \tag{4}$$

The solution of this equation in a rectangular waveguide with cross-sectional dimensions (a·b) and a medium filling this cavity that does not contain free and bound relaxing charges (i.e., for an ideal dielectric with relative permittivity ε = 1) for the lowest type of oscillations H$_{10}$ is given by the following formulas (Landay *et al*., (1991); Sychev V.V., (1991)):

$$\boldsymbol{E_x = 0;}\ E_y = AZ_B \sin\left(\frac{\pi x}{a}\right); \boldsymbol{E_z = 0;}\ H_x = -A\sin\left(\frac{\pi x}{a}\right); \boldsymbol{H_y = 0;}\ H_z = -jA\frac{\lambda_b}{2a}\cos\frac{\pi x}{a} \tag{5}$$

Where x, y, and z are the coordinates, respectively, along the wide wall of the waveguide cross-section, along the narrow wall and perpendicular to the cross-section, i.e. along the waveguide. The distribution of the amplitude of the electric field along the wide wall of the waveguide is given by the component E_y, i.e. half of the period of the sine wave with a wide maximum in the centre of the waveguide. In this place of the waveguide, a test tube with the tested material is placed. In this case, the electromagnetic field lines E_y are parallel to the axis and walls of the tube and, consequently, the maximum values of the electric field intensity in an empty waveguide and in a dielectric are practically the same (Okress, (1968); Slettery, (1978); Brandt, (1963); Emme, (1967)). The maximum amplitude of the electric field intensity is proportional to the microwave energy feeding the waveguide line. The following equation determines the amplitude of the electric field intensity of a travelling wave of type H$_{10}$ at the middle of a broad wall of the waveguide with channel dimensions a and b (Harvey A.F., (1963), Ocress E.C., (1968), Temam R., (2005), Slettery J.S., (1978)):

$$P_0 = \frac{E^2 ab}{4Z_B} \tag{6}$$

Where P_0 is the power passing on the waveguide without a dielectric; Z_B–wave impedance, Ω. For the traveling wave of type H_{10} the wave resistance is represented by (Harvey, (1963); Ocress, (1968); Temam, (2005); Slettery, (1978)):

$$Z_B = \frac{120\pi}{\sqrt{1-\left(\frac{\lambda_0}{2a}\right)^2}} \tag{7}$$

Where λ_0, is the wavelength in free space (it is 12.2cm for frequency 2450MHz). During the determination loss factor of heat absorbing agent the energy absorbed by the sample is determined by energy balance:

$$P_{abs} = P_0 - P_{refl} - P_{pass} \tag{8}$$

Where: P_0, is the microwave energy travelling on the sample; P_{pass}, microwave energy passed out of the sample and absorbed by the matched load; P_{refl}, microwave energy reflected by the sample. Dielectric losses in the material (studied sample) or the energy dissipated by the dielectric is determined by the equation (Landay et al., (1991); Lykov, (1978); Ocress, (1968)):

$$P_{diss} = 0.5\varepsilon_0\varepsilon''\omega E^2 \cdot V \tag{9}$$

The square of electric field intensity is determined from equations (6) and (7):

$$E^2 = \frac{4P_0}{ab} \cdot \frac{120\pi}{\sqrt{1-\left(\frac{\lambda_0}{2a}\right)^2}} \tag{10}$$

The formula for calculation of the dielectric losses factor is derived from (9) and (10):

$$\varepsilon'' = \frac{P_{abs}}{0.5 \cdot \varepsilon_0\omega} \cdot \frac{a \cdot b\sqrt{1-\left(\frac{\lambda_0}{2a}\right)^2}}{4P_0 \cdot 120\pi} \cdot \frac{4}{V} \tag{11}$$

It must be emphasized that calculations based on such measurements can give dielectric loss factors with significant errors. However, this accuracy is sufficient for comparison and substantiated determination of a heat-absorbing agent. The required orientation of the microwave field electric intensity and the tube with the studied materials is arranged in the working unit where the conditions are appropriate for measuring the temperature of samples and the indirect electric component of the microwave field intensity (bypassing power measurement).

1.3 Pilot scale installation

For testing the feasibility of this method was developed demonstration installation on a pilot scale. A schematic diagram of the demonstration installation for testing the microwave heating technology for DUF_6 evacuation from the container is presented in **Figure 4**. Antenna input of microwave energy (coaxial emitter of microwave energy) is a basic element of microwave unit of demonstrating installation shown in **Figure 5**. It is intended for microwave energy supply into the evaporative container with UF_6. The same type of stainless steel cylinders with a volume of 24 litres each are used as evaporative and receiving tanks. The cylinders are equipped with a coaxial antenna input in the centre of the neck and two nozzles with shut-off valves for connection to the process line. After DUF_6 condensation is completed, the cylinders are swapped and the cycle repeats. The receiving tank is placed in a dewar and cooled with liquid nitrogen. The evaporative tank stands free. During the operation of the evaporative tank, temperature control is carried out at 3 points: in the bottom part, directly above the tablet layer and in the neck at the gas outlet from the tank. The process line allows the system to be evacuated to a residual pressure of 1.0-1.5mm Hg and to measure the pressure before and after the critical diaphragm with a hole diameter of 1.6 mm to determine the gas flow rate during condensation (G_{max}=11.45 g/min). Information from temperature and pressure sensors is sent to the computer.

1.4 Determination of heat-absorbing agent amount for the process implementation

The heat-exchange efficiency between a heat-absorbing agent and a solid UF_6 can be evaluated as follows (Volchonok, (1991); Dulnev, (1991)). The electrophysical properties of the solid UF_6 heat absorbing agent mixtures determine the penetration depth of the microwave field and the heat release. The technological process of UF_6 evaporation from containers includes loading a heat-absorbing agent into a container and irradiation with microwave energy. Heat release occurs in the heat-absorbing agents which are distributed in a certain surface layer of the solid uranium hexafluoride.
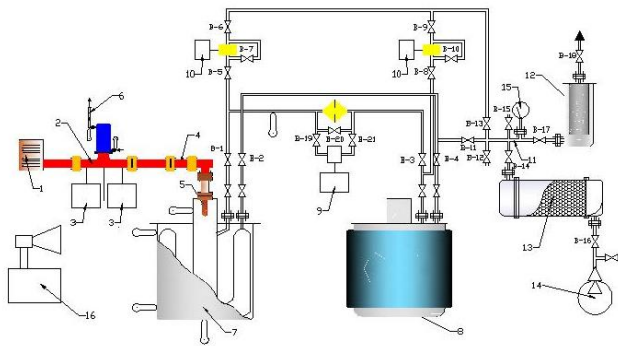
Fig. 4 Diagram of the demonstration installation, 1-microwave generator; 2-Ferrite circulator; 3-wattmeters of absorbed power; 4-coaxial waveguide junction; 5-antenna; 6-rotameter; 7-evaporation container; 8-receiving container; 9-flowmeter UF6; 10-differential pressure gages; 11-vacuum rake; 12-metal-ceramic filter; 13-absorption column; 14-vacuum pump; 15-vacuum meter; 16-device for microwave radiation measurement.
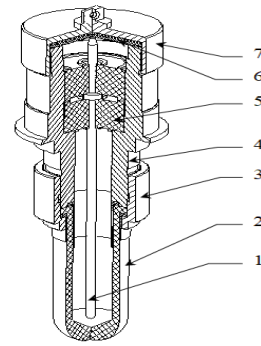
Fig. 5 Coaxial microwave emitter inserted into the neck of the container with DUF6, 1-antenna rod; 2-radio transparent antenna shell; 3-antenna casing mounting clip; 4-external antenna coaxial electrode; 5-radio transparent centering washers; 6-gasket disc; 7-antenna connector plug.

A heat-balance equation is used to determine the total microwave energy for uranium hexafluoride sublimation:

$$wVdt = L_{UF_6}Gdt + c_p\rho_{uh}V_{uh}\Delta Tdt + c_{p\,haa}\rho_{haa}V_{haa}\Delta Tdt \tag{12}$$

Here: w is the absorbed microwave energy, wt/cm^3; ρ, density of the heat-absorbing agent, g/cm^3; c_{haa}, the specific heat capacity of the heat-absorbing agent, J/g°C; L_{uh}, the specific sublimation heat of uranium hexafluoride, J/g; c_p, the specific heat capacity of gaseous uranium hexafluoride, J/g °C; V, the volume in which the microwave energy is absorbed, cm^3; V_{uh}. the amount of evaporated uranium hexafluoride, cm^3; and V_{haa}, the amount of heat-absorbing agent loaded on solid uranium hexafluoride, cm^3.
The absorbed microwave energy per unit volume is written:

$$w = 0.5\varepsilon_0\varepsilon''\omega E^2 \tag{13}$$

Total absorbed energy:
$$W_{micr} = wV \tag{14}$$
The absorbing volume V is equal to the product of solid uranium hexafluoride surface area S in the container and the penetration depth h of the microwave field into the uranium hexafluoride and the heat-absorbing agent mixture:

$$W_{micr} = wSh \tag{15}$$
The quantity W is determined by the cross-section surface of the container where the uranium hexafluoride is stored and the penetration depth, by the electrophysical properties of uranium hexafluoride and the heat-absorbing agent mixture.

1.5 Calculation of effective dielectric permittivity of two-component mix

A row of formulas may be applied for the calculation of dielectric characteristics of a two-component mix. Hereinafter, a row of basic formulas for calculating the permittivity of two-component mixtures is considered. For a matrix mix of solid uranium hexafluoride with lonely inclusions, the dependence of dielectric permittivity inclusions ε_1 and dielectric permittivity of continuous medium ε_2 with a volume ratio of inclusion amount and total medium amount η is determined by the following formula:

$$\frac{\varepsilon_a - \varepsilon_{a2}}{\varepsilon_a + 2\varepsilon_{a2}} = \eta\frac{\varepsilon_{a1} - \varepsilon_{a2}}{\varepsilon_{a1} + 2\varepsilon_{a2}} \tag{16}$$

Where ε_a dielectric permittivity of the mix. It is applied for a small amount of inclusions. It may be transformed to the next:

$$\varepsilon_a(\eta) = -\varepsilon_2\frac{-\varepsilon_{a1} - 2\varepsilon_{a2} - 2\eta\varepsilon_{a1} + 2\eta\varepsilon_{a2}}{\varepsilon_{a1} + 2\varepsilon_{a2} - \eta\varepsilon_{a1} + \eta\varepsilon_{a2}} \tag{17}$$

Matrix mix is described by the following formula:

$$\frac{\varepsilon_a - \varepsilon_{a2}}{3\varepsilon_{a2}} = \eta \frac{\varepsilon_{a1} - \varepsilon_{a2}}{\varepsilon_{a1} + 2\varepsilon_{a2}} \tag{18}$$

The solution to this equation is:

$$\varepsilon_a(\eta) = \frac{-\left(-\varepsilon_{a1} - 2\varepsilon_{a2} - 3\eta\varepsilon_{a1} + 3\eta\varepsilon_{a2}\right)}{\varepsilon_{a1} + 2\varepsilon_{a2}} \tag{19}$$

The statistical mix contains a number of inclusions more than matrix one. It requires using another formula. The statistical mix is described by the next formula:

$$\eta \frac{\varepsilon_{a1} - \varepsilon_a}{\varepsilon_{a1} + 2\varepsilon_a} + (1-\eta)\frac{\varepsilon_{a2} - \varepsilon_a}{\varepsilon_{a2} + 2\varepsilon_a} = 0 \tag{20}$$

The solution to this equation is:

$$\varepsilon_a(\eta) = \frac{3}{4}\eta\varepsilon_{a1} - \frac{3}{4}\eta\varepsilon_{a2} + \frac{1}{2}\varepsilon_{a2} - \frac{1}{4}\varepsilon_{a1} + \frac{1}{4}(9\eta^2\varepsilon_{a1}^2 - 18\eta^2\varepsilon_{a1}\varepsilon_{a2}) +$$
$$\frac{1}{4}\left(+18\eta\varepsilon_{a1}\varepsilon_{a2} - 6\eta\varepsilon_{a1}^2 + 9\eta^2\varepsilon_{a2}^2 - 12\eta\varepsilon_{a2}^2 + 4\varepsilon_{a2}^2 + 4\varepsilon_{a1}\varepsilon_{a2} + \varepsilon_{a1}^2\right)^{\frac{1}{2}} \tag{21}$$

In the range of volume parts η from 0.01 to 0.1 differences the above formulas keep within a 20% interval.

1.6 Calculation of two-component mix loss factor

For the determination of the dielectric loss factor of the mix, the formula for the tangent of the dielectric loss factor is applied:

$$tg\delta_{mix} = \sum_i \eta_i tg\delta_i \tag{22}$$

At a small concentration of inclusions, the tangent of the dielectric loss factor is linearly increased with concentration the same as for dielectric permittivity dependence. From the formula, for a two-component mix, i.e. for one form of inclusions into a continuous medium, the next formula may be obtained

$$\frac{\varepsilon_{mix}''}{\varepsilon_{amix}} = \eta \frac{\varepsilon_{haa}''}{\varepsilon_{ahaa}} + (1-\eta)\frac{\varepsilon_{uh}''}{\varepsilon_{auh}} \tag{23}$$

To reduce the loss factor of the mix, the analytical dependence will be in the form:

$$\varepsilon_{mix}'' = \eta \frac{\varepsilon_{haa}'' \cdot \varepsilon_{mix}}{\varepsilon_{ahaa}} + (1-\eta)\frac{\varepsilon_{uh}'' \cdot \varepsilon_{mix}}{\varepsilon_{auh}} \tag{24}$$

1.7 Calculation of penetration depth and heat-absorbing agent amount for microwave process implementation

Penetration depth into the mix and heat-absorbing agent amounts for process implementation is determined on the base of the effective dielectric permittivity of the mix. The following dependence is applied for the determination of penetration depth of microwave into the mix of uranium hexafluoride and heat-absorbing agent (Harvey, (1963); Ocress, (1968); Temam, (2005); Slettery, (1978); Emme, (1967)):

$$h = \frac{\lambda_0}{\pi\sqrt{\varepsilon_a}\frac{\varepsilon''}{\varepsilon_a}} = \frac{\lambda_0\sqrt{\varepsilon_a}}{\pi\varepsilon''} \tag{25}$$

where ε_a and ε'' are given values for mix, λ_0 wavelength in free space.

Penetration depth is less than uranium dioxide pellet dimensions at volume parts above 0.06. Therefore, the heat-absorbing agent must be placed on the uranium hexafluoride surface at volume parts about 0.06-0.07. The total mass of the heat absorbing agent is the product of the volume in which the electromagnetic field absorbed $V=hS$ and the volume parts of the heat-absorbing agent η and its density ρ:

$$M = hS\eta\rho \tag{26}$$

2 Results and Discussion
2.1 Laboratory study

Once put into operation works were completed, calibration of dropped and reflective power meters were realized and the graduation curve of the adjustable attenuator was constructed. The next stage of work was to test the technique for the determination of the absorption coefficient (tg δ) of liquid heat-absorbing agents on a sample with well-known characteristics (conductive and distilled water). Then thermograms of freons heating were obtained in the range from 20 to 95⁰C and tg δ was determined as well. It was found that the general view of the temperature dependence of absorption coefficient of water and its value at frequency 2.45MHz fit well to ones represented in reference books (Puschner, 1966). Produced results were used for applying the correction for calibration characteristics of power meters that did not take into account heat losses in the environment during experiments. Before proceedings to direct measurements of absorption coefficients of 112 ($C_2F_2Cl_4$), 113 ($C_2F_3Cl_3$), and 122a, ($C_2F_2HCl_3$) freons, were there conducted investigations allowing to determine appropriation for its use as heat-absorbing agents of microwave energy. With this object of view, thermograms of the above freons heating (up to boiling points) were obtained at the same level of supplied energy. Then analogous investigations were conducted with samples of freons containing 10% (by volume) uranium hexafluoride. It was found that with the addition of uranium hexafluoride in the amount of 10% to freons, the ability of mixes to absorb microwave energy is kept constant and boiling points alone were changed. **Figure 6** presents a typical heating diagram for mixtures of halocarbons with uranium hexafluoride and distilled water for comparison.

The short-circuit method was used to evaluate the dielectric properties of the most promising eat-absorbing agents (UO_2 and U_3O_8). The measuring procedure and calculation of dielectric characteristics are discussed in detail in the literature (Harvey, (1963); Ocress, (1968); Temam, (2005); Slettery, (1978), Emme, (1967); Puschner, 1966). **Figure 7** presents the dielectric properties of the heat-absorbing agents (UO_2 и U_3O_8) in the frequency range of 2-3MHz and the temperature range of 25-95 C. It was found that the dielectric properties of the uranium oxides varied within large limits. The comparison of the above heat-absorbing agents by the dielectric loss factor at the frequency of 2.24MHz and in the temperature range of 25-95 C is shown in **Figure** 8.
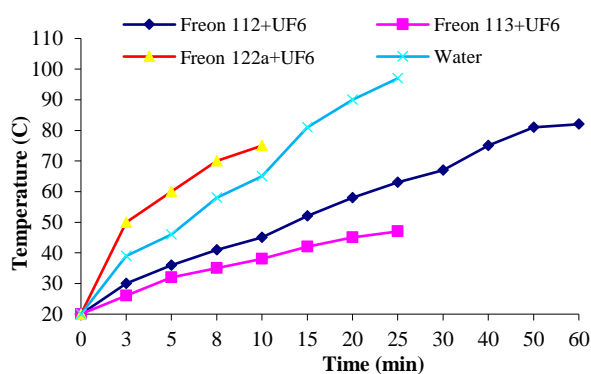

Fig. 6 Typical heating diagram.

2.2 Microwave method test on pilot-scale installation

At first, there were determined penetration depth and the total mass of heat-absorbing agents for experimental tests of microwave technology on pilot scale installation. The experimental dependence of the penetration depth of microwave energy into a mixture of uranium hexafluoride and uranium dioxide pellets and the volume fraction of pellets is shown in **Figure 9**.

Penetration depth is less than uranium dioxide pellet dimensions at volume parts above 0.06. Therefore, a heat-absorbing agent must be placed on the uranium hexafluoride surface at volume parts about 0.06-0.07. The total mass of the heat-absorbing agent is the product of the volume in which the electromagnetic field absorbed $V=hS$ and the volume parts of the heat-absorbing agent η and its density.

Fig.7 Loss factors and frequency dependence.

It should be noted that calculated penetration depth for a given heat-absorbing agent amount (vol.) and heat-absorbing mass is minimum for process realization. Matching microwave generators with a container filled with solid uranium hexafluoride and the heat-absorbing agent is not considered.

The experimental tests of the environmentally safe technology on pilot scale installation were conducted as follows. Depleted uranium hexafluoride (8-9 kg) was charged into an evaporative container. Then a heat-absorbing agent (about 600g) was located on the surface of the solid uranium hexafluoride. The experimental study was conducted under various conditions of microwave energy supply. After the experiments, the containers were weighed to determine the average rate of evaporation. During the process the following parameters were controlled:
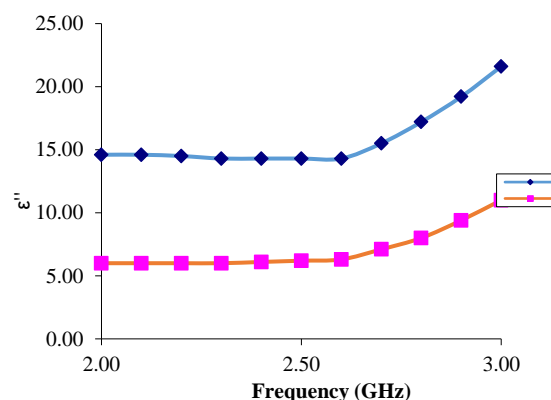
- Temperature before the metal-ceramic filter

- Temperature of the evaporation container bottom
- The temperature in the gas outlet from the container
- Temperature over the heat-absorbing agent
- Pressure before the flow-rate disk
- Pressure before the metal-ceramic filter
- Pressure behind the flow-rate disk

The controlled parameters to decrease the supplied energy or to switch off the generator were:

- the temperature of the heat-absorbing agent above 100^0C;
- equilibrium pressure of UF_6 in the container greater than the vapor pressure at a given temperature;
- the pressure increase in the receiving container breaking the critical gas flow rate through the disk;
- alert conditions: vacuum violation, microwave escape, etc.

Below are presented results of experiments where the microwave energy was supplied in continuous mode. **Figure 10** shows the dependence of the pressure in the evaporation container during depleted uranium hexafluoride sublimation. This value is proportional to the sublimation rate. **Figure 11** shows diagrams of temperatures at control points. Microwave energy was supplied into the container after a pressure reduction of about 20mm mercury. During the tests, with a continuous supply of microwave energy before entering the stationary mode, the generator power was 50watts. Then the supplied power was reduced to 35watts to avoid overheating the walls of the tank. The average rate of sublimation in this experiment was 10.2g/sec.

Experiments with cyclic microwave energy supply are presented in **Figures 12** and **13**. Microwave energy supply was increased up to 100watts. The sublimation process between intervals of microwave supply was conducted due to heat accumulated by a heat-absorbing agent. Diagrams of temperatures in control points are shown in Fig. 13.
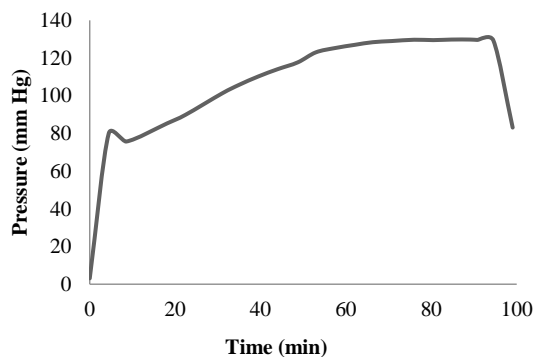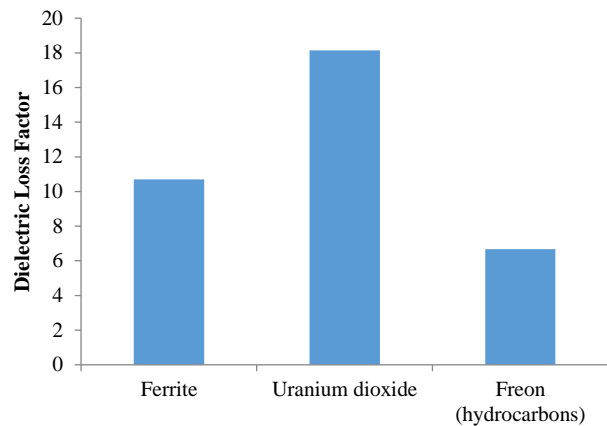

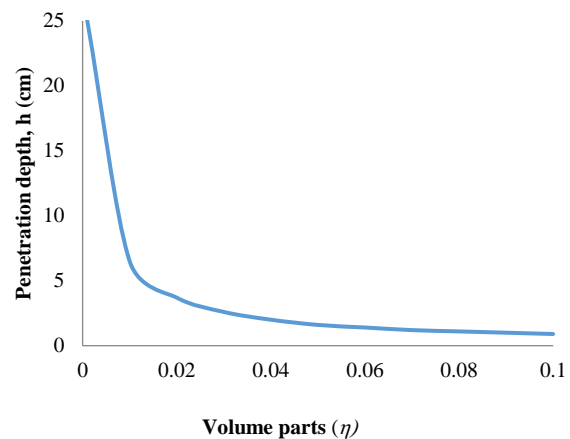**Fig. 8** Comparison diagram of the dielectric loss factor of the studied materials.

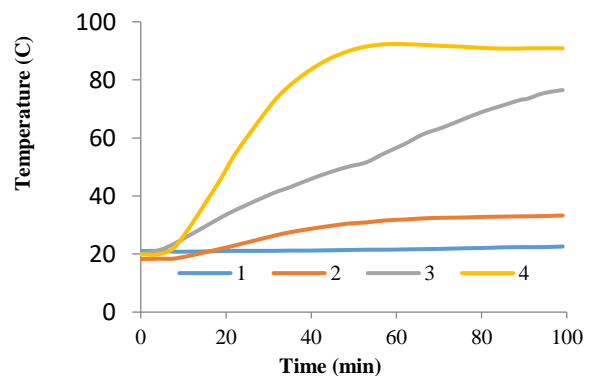**Fig. 9** Dependence of penetration depth of microwave into mixture and volume fraction of pellets.

Fig.10 Dependence of pressure in the container during depleted uranium hexafluoride sublimation vs. duration.

Fig. 11 Temperature dependence at monitoring points:1–temperature before metal-ceramic filter, 2-temperature of evaporation container bottom, 3-temperature in gas outlet from the container, 4-temperature over heat-absorbing agent.

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Fig. 12 Dependence of pressure in the container during depleted uranium hexafluoride sublimation vs. duration.

Fig. 13 Temperature dependence at monitoring points: 1–temperature before metal-ceramic filter, 2-temperature of evaporation container bottom, 3-temperature in gas outlet from the container, 4-temperature over heat-absorbing agent.
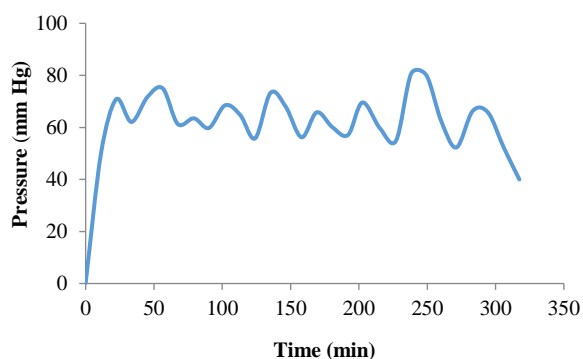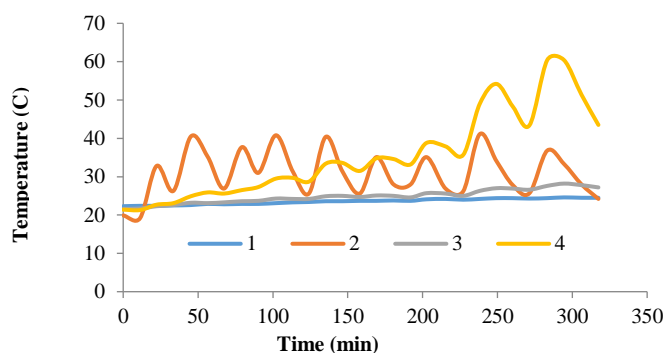
Switching on and off microwave energy supply is easily determined with inflection on pressure and temperature diagrams. The average duration of one cycle ("heating–cooling") was about 32min. including "microwave energy supply on"–10min. and "microwave energy supply off"–22min. The average rate of sublimation in this experiment was 12.92g/sec. Diagrams of pressure and temperatures for the cycle "heating–cooling" are presented in **Figures 14** and **15**.
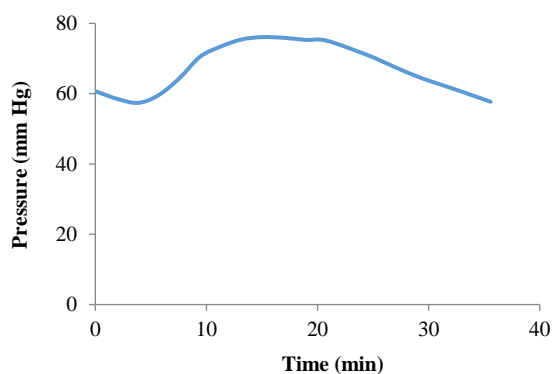

Fig. 14 Dependence of pressure in the container during depleted uranium hexafluoride sublimation vs. duration.
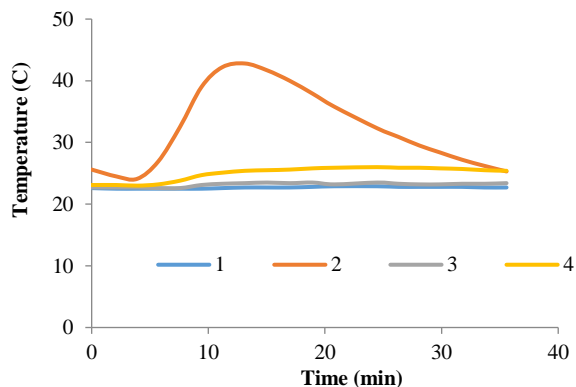

Fig. 15 Temperature dependence at monitoring points: 1–temperature before metal-ceramic filter, 2-temperature of evaporation container bottom, 3-temperature in gas outlet from the container, 4-temperature over heat-absorbing agent.

As the result of conducted experiments on a laboratory bench, there were determined characteristics of heat absorbing agents. It was found:

1   As evident from the thermogram, the ability of 122a freon to absorb microwave energy is similar to water whereas 112 and 113 freons have negligibly small absorption ability to microwaves. An application of 122a freon as a heat-absorbing agent is limited by the specific application. It has a row of advantages uniform ingress into solid uranium hexafluoride and high microwave penetration depth, and disadvantages – its losses in evaporation process and low losses factor as compared to solid heat-absorbing agents.

2   Uranium dioxide has the most absorptive characteristics to microwave and affinity with uranium hexafluoride. Ferrites are effective enough to absorb microwave but it is inferior to uranium dioxide in this respect.

3   Uranium dioxide and ferrites as solid heat-absorbing agents are proposed for tests at demonstrating installation.

4   Considering uranium dioxide as a promising heat-absorbing agent for uranium hexafluoride in industry, it is appropriate to conduct a study of its dielectric characteristics (losses factor and dielectric permeability) for the determination of the optimum amount of heat-absorbing agent.

5   Since part of depleted uranium stock is stored as uranium mixed oxides, a similar study is appropriate to conduct with

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uranium mixed oxides.

Since part of depleted uranium stock is stored as uranium mixed oxides, a similar study is appropriate to conduct with uranium mixed oxides. The experiments conducted on the demonstration installation showed that the microwave method of sublimation of DUF_6 from steel containers allows them to be emptied safely with practically cold ($T<100^0C$) walls, with high efficiency of using microwave energy ($\eta\approx97\%$) and with maintaining a given level of pressure in the containers and process performance during evaporation. Tests of two modes of conducting the process (with the continuous and periodic supply of microwave energy) have shown that the periodic mode is preferable: it allows to reach a given performance faster (according to experimental data by six times) and more economical (according to experimental data by 13%). As a result of conducted experiments on demonstration installation it was found:

- feasibility to evaporate uranium hexafluoride from containers using microwave energy;
- microwave method of depleted uranium hexafluoride sublimation is a safe technique for maintaining «cold" walls and given pressure in the container and flow rate of gas;
- the proposed technique allows to exclude the damage or deterioration of hermetic containers due to local overheating;
- easy to control;
- this technique applies to any kind of container;
- possibility to control temperatures and conditions of container walls;
- this method is recommended for pilot and industrial tests. It is easy to realize and there is a technical base for its application.

In the presented experiments, sintered pellets of ferrites were used as a heat-absorbing agent. Contamination of DUF_6 during its evacuation from the containers, as well as deformation of ferrites, was not detected. The number of experiments in which ferrites were used was relatively small. It is possible that with their longer-term use, deformations of pellets may be observed. No special studies have been conducted on this issue.

When heating materials are liquids, for example, low- and medium-temperature heating of oil fractions and similar flows by microwave energy at technological installations of oil refining, oil and coke chemistry and other industries (Yudina, *et al*, (2019), Morozov, (2010); Salomatov, *et al*, (2018); Salomatov, *et al*, (2018); Arutyunyan, (2021); Anfinogentov, *et al*, (2014)), it is advisable to use ferrites (thermal conductivity, 3.5wt/m·K; specific heat, 0.7 kJ/kgK; density, 3-5kg/m^3; specific resistance, 10^{7}Ohm.cm) as heat-absorbing agents. The process of microwave heating of ferrites is similar to the above. Currently, the aforementioned liquid flows are heated by fire tube heat exchangers. Combustion products in such heat exchangers are released into the atmosphere and pollute the environment. A low-efficiency conductive heat-exchange process is carried out through the outside surface of pipe-heated furnace gases. Microwave heating avoids this problem because heat is released in the bulk of the material and sharply increases heat exchange. The heat exchange of a mix of liquid and ferrites can be considered heat exchange in a fluidized bed. This research was suspended at the stage of the statement of work due to a lack of funding. Experimental studies were not conducted.

Conclusions

Based on conducted experiments the following recommendations for industrial technology application using microwave energy may be made:

- Proposed frequency of generator 915MHz for power 25-50kwat taking into account container dimensions;
- The facility must include water loading for adjustment of microwave system and energy excess balance;
- The facility must include measuring unit of standing wave coefficient at the automatic regime for power and disaster protection control;
- Junction for reflective energy (such as microwave isolator in demonstrating installation) is to be included for system safety;
- The antenna input of the microwave must be located in a separate block of the magnetron.
- The power supply and control block should be remote from the generator;
- The technological unit must support the long-time operation of installation;
- Control and measuring instruments and automatic equipment must provide parameters control: vacuum, pressure, temperatures at control points and evaporation flow rate.

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Leading engineers A. N. Evdokimov and V. I. Dushechkin have made a great contribution to the implementation of the project (experimental studies, design, manufacture, and commissioning of installations). We thank Dr Dennis W. Wester, Pacific Northwest National Laboratory, for collaboration on ISTC project #2285.

Nomenclature

a, b, h	=geometric dimensions	[m]
c	=heat capacity	[kJ/K]
c_p	=specific heat capacity	[kJ/kgK]
E	=electric field intensity	[V/m]
L_{uh},	=specific sublimation heat of uranium hexafluoride	[J/g]
P_0	=microwave energy travelling on the sample	[kJ]
P_{abs}	=microwave energy absorbed by the matched load	[kJ]
P_{pass}	=microwave energy passed out of the sample	[kJ]
P_{refl}	=microwave energy reflected by the sample	[kJ]
S	=surface area	[m^2]
T	=process time	[min]
T	=temperature	[K]
V	=volume	[m^3]
E	=relative electric permittivity of the material	[-]
ε'	=eal part of electric permittivity	[-]
ε''	=imaginary part of electric permittivity (loss factor)	[-]
ε_0	=electric permittivity of the vacuum	[F/m]
$\varepsilon_a, \varepsilon_{0,} \varepsilon$	=absolute electric permittivity	[F/m]
η	=volume parts	[-]
μ	=relative magnetic permittivity	[-]
μ_0	=magnetic permittivity of the vacuum	[H/m]
ρ	=density	[kg/m^3]
ω	=frequency	[1/s]
λ_0	=wavelength in free space	[1/m]

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