Mathematical model desublimation conditions

Abstract—The developed and software-implemented simplified three-dimensional mathematical model of the unsteady-state process of HFS desublimation into immersible vertical tanks with vertical finning is described. The study of regularities in the HFS desublimation process is performed by numerical modeling.

Keywords: ammonium hexafluorsilicate, desublimation, mathematical modeling, heat transfer processes

Introduction

One of the acute issues of today is the development of advanced methods for obtaining highly purified silicon dioxide, with environmentally, health and economy-friendly methods by using wastes containing Si (sand, coal, rice husk) for solar panel applications. This is due to the fact that such a product acts as a raw material base for the production of mono- and polycrystalline silicon, on which the entire electronic industry is based. In addition, silicon dioxide is used to produce silicon carbide. Both for the synthesis of metallic silicon and for the production of silicon carbide, high-purity carbon is a necessary component. Therefore, its purity for the synthesis of these target products must also be given great attention [1].

To obtain silicon dioxide from sand or other raw materials, a laboratory unit was designed to study the process of oxide separation. The installation contains a rotating reactor and sublimator reactor in which the process of separation of oxides takes place [2].

Obtaining data on the sublimation and desublimation of HFSA, as the main link for obtaining high-purity silicon dioxide, is of interest not only in terms of obtaining scientific data to increase efficiency, but also in terms of finding technological parameters for industrial plants

The study of gas mixture flows based on the solution of the complete system of Navier-Stokes equations has a long tradition. A very limited number of cases are known in the literature that allow analytical integration of the Navier-Stokes equations [3]. The Navier-Stokes equations have a number of specific features that significantly affect their numerical solution, regardless of their form of writing. One of the essential features is the non-linearity and paraboloelliptic nature of these equations. Therefore, in order to correctly model the elliptic nature of the Navier-Stokes equations, it is necessary to use the elliptic Poisson equation for pressure.Therefore, progress in this area is possible only through the use of numerical methods. At present, for the numerical solution of the Navier-Stokes equations, several dozen varieties of difference schemes exist and are used.

At the present stage of the development of computational fluid mechanics, its further progress is associated with the improvement of flow models, mixing processes, and computer technologies for integrating the initial equations. The last of them are usually associated with the complication of computational algorithms through the use of approximation schemes of a higher order of accuracy and structured grids. However, this path is associated with the increasing complexity of computational algorithms and programs, which leads to great difficulties in their implementation, and therefore is available only to some professionals. This limits the possibility of their wide use in design and development activities. Therefore, the main goal of our work is to create and discuss a relatively simple numerical algorithm for solving the fundamental Navier-Stokes equations.

This work is devoted to mathematical modeling of the process of desublimation of gaseous HFS into vertical transport containers. The existing mathematical models of the HPS desublimation process [4] are, as a rule, stationary or quasi-stationary. Their common disadvantages are the lack of consideration of heat transfer by convection and friction of the gaseous HFS against the desublimate layer during its movement along the heat exchange walls; desublimation on the end elliptical walls of heat exchange tanks; nonstationarity of heat and mass transfer processes. In this case, The mathematical model was developed taking into account the following assumptions: all the heat released during the phase transition is removed by the refrigerant through the vessel wall and the desublimate layer; gaseous HFS does not contain impurities; thermal processes inside the system under consideration can be described within the frame work of the traditional theory of heat conduction; the gas flow before entering the container is considered to be isentropic, so the values of the total enthalpy and entropy are set on the cut.

The calculation of the hydrodynamic regime in the working volume of the apparatus was carried out on the basis of a system of complete stationary axisymmetric averaged differential equations of the Navier-Stokes elliptic type [5], written in variables of vorticity Ω, stream function Ψ. Eq. (1), (2) are written in a cylindrical coordinate system, where r is the radial and z is the axial coordinates:

$$u_r \frac{d\psi}{\partial r} + u_z \frac{d\psi}{\partial z} = \frac{1}{\mathrm{Re}} \frac{1}{r} \left[\frac{\partial}{\partial r}\left(r\nu_t \frac{\partial \psi}{\partial r} \right) + \frac{\partial}{\partial z}\left(r\nu_t \frac{\partial \psi}{\partial z} \right) \right] + F_j . \ (1)$$

$$\frac{\partial^2 \psi}{\partial r^2} + \frac{\partial^2 \psi}{\partial z^2} = \Omega r^2 + \frac{1}{r}\frac{\partial \psi}{\partial r}, \quad \Omega = \frac{1}{r}\left[\frac{\partial u_r}{\partial z} - \frac{\partial u_z}{\partial r} \right],$$

$$u_r = \frac{1}{r}\frac{\partial \psi}{\partial z}, \quad u_z = -\frac{1}{r}\frac{\partial \psi}{\partial r} . \ (2)$$

Left side of eq. (1) describes the convective transfer of a substance (gaseous HFS) due to the velocity of the medium (vapor-gas mixture). The first term on the right side of eq. (1) describes the diffusion transfer of a substance due to the viscosity νt of the medium along the coordinates r and z. The term F_j describes the additive source components due to the change in the volumetric flow rate of the medium over the cross section as a result of the loss in the HFS mass from the gas-vapor mixture during its desublimation. The Poisson equation (2) for determining the stream function Ψ was obtained from the continuity equation [6].

The formulation of the problem in terms of spatial coordinates is two-dimensional and is described by the system of non-stationary Navier-Stokes equations for the gas phase and the heat equation conductivity for the solid phase.

Using the above equations, we have developed a program for calculating the processes of cooling and desublimation of HFS ammonia from a vapor-gas mixture.

2.Thermal and hydrodynamic calculations.

When calculating the cooling and desublimation, the HPS is successively determined for the flows shown in Fig. 1: 1) integral physical and chemical characteristics of the vapor-gas mixture at the inlet and outlet of the desublimator; 2) heat transfer for an axisymmetric problem.

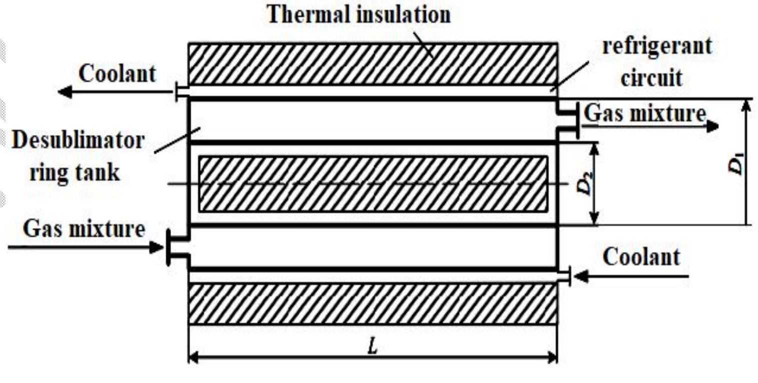

Fig.1. Scheme of flows entering and leaving the desublimator. L is the length of the annular container of the desublimator; D_1, D_2 - diameters of the outer and inner rings of the device

The average speed of movement of the gas-vapor mixture over the cross-section of the apparatus W, m/s is determined by the formula Q= W/S, where S is the cross-sectional area of the apparatus, m^2.

The main criteria for thermal and hydrodynamic similarity (Reynolds Re, Prandtl -Pr and Peclet -Pe) are [5]: $\mathrm{Re}=\frac{W\rho_{mix}d_e}{\mu_{mix}}$; $\mathrm{Pr}=\frac{c_{pmix}\mu_{mix}}{\lambda_{mix}}$; $Pe=\mathrm{Re}\,\mathrm{Pr}$. Where de is the equivalent diameter of the annular section of the apparatus, m.

The heat transfer equation for the case of axially metric laminar flow has the form [7]:

$$\frac{\partial T}{\partial \tau}+W\frac{\partial T}{\partial x}=a\left(\frac{\partial^2 T}{\partial x^2}+\frac{1}{r}\frac{\partial T}{\partial r}+\frac{\partial^2 T}{\partial \tau^2}\right)\ (3).$$

Then approximately, $\frac{\partial T}{\partial x}=\frac{T-T_0}{x}$, $\frac{\partial^2 T}{\partial x^2}=\frac{T-T_0}{x^2}$ and consider the relation

$$\left(W\frac{\partial T}{\partial x}\right)/\left(a\frac{\partial^2 T}{\partial x^2}\right)=\frac{Wx}{a}=\frac{Wd_e}{v}\frac{v}{d_e}=\mathrm{Re}\,\mathrm{Pr}\frac{x}{d_e}=Pe\frac{x}{d_e}>>1$$

Where $W\frac{\partial T}{\partial x}$ is the convection heat transfer, $a\frac{\partial^2 T}{\partial x^2}$ s the diffusion heat transfer, $a=\frac{\lambda_{mix}}{c_{mix}\rho_{mix}}$ coefficient of thermal conductivity.

The estimation carried out shows that the ratio of convective heat transfer to heat transfer by thermal conductivity >>1 in the entire computational domain. Therefore, the transfer of the heat flux along the longitudinal axis (x) by heat conduction can be neglected in comparison with the transfer by convection.

As a result, an approximate equation of the boundary layer was obtained. Taking into account the ratio between the values of the thermal conductivity (λ) and heat transfer coefficients (α), as well as the geometric dimensions of the region of the vapor–gas mixture flow in the annular desublimator, it can be assumed that the gas flow is cooled to a constant temperature equal to the HFS desublimation temperature, and the entire the heat flux released during desublimation is removed through the side surfaces (Fig. 2). The following designations are accepted: T_0, T_1, T_s, T_w are the temperatures of the gas-vapor mixture at the inlet to the cooled zone of the desublimator, the desublimation front, the desublimation surface and the cold wall of the desublimator; q_1, q_2 are the heat fluxes removed from the gas flow to the desublimation surface and through the desublimate layer to the apparatus wall.

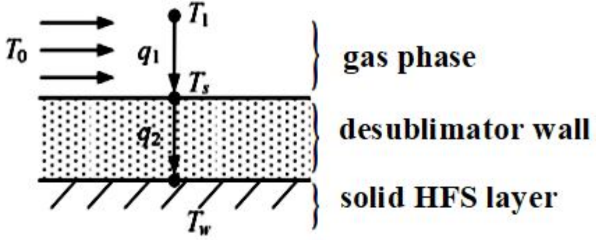

Fig. 2. Scheme of the process of cooling and desublimation of HFS from a vapor-gas mixture

From Newton's law for heat transfer, one can express the heat flux transferred by the gas to the desublimation surface: $q_2=-\lambda_s\frac{T_s-T_w}{\delta}=-\frac{\lambda_s}{\delta}(T_s-T_w)$ and from the Fourier law for heat transfer - the heat flux transmitted through the layer of solid HFS to the wall of the apparatus: $\alpha(T_s-T_w)=-\frac{\lambda_s}{\delta}(T_s-T_w)$. Based on the foregoing, we accept q_1=q_2, then (6) desublimation surface temperature, we have: $T_s=\left(\alpha+\frac{\lambda_s}{\delta}\right)\alpha T_1+\frac{\lambda_s}{\delta}T_w$.

$$T_s=\frac{\alpha T_1+\frac{\lambda_s}{\delta}T_w}{\alpha+\frac{\lambda_s}{\delta}}=\frac{T_w}{1+\frac{\alpha\delta}{\lambda_s}}+\frac{\frac{\alpha\delta}{\lambda_s}T_1}{1+\frac{\alpha\delta}{\lambda_s}}\ (4).$$

Since the Nusselt criterion is equal $Nu_s=\frac{\alpha\delta}{\lambda_s}$, you can get: $T_s=\frac{T_w}{1+Nu_s}+\frac{Nu_s T_1}{1+Nu_s}$ (5).

Numerical implementation of the mathematical model is carried out in the following order: 1) given by the initial data: the initial geometry of the computational domain (D_1, D_2, L); physical properties and consumption parameters of each of the components of the initial gas-vapor mixture (ρ, μ, λ, c_p, dm) HFS, N_2, crystalline HFS (ρ_s, λ_s, ΔH); boundary conditions for speed and temperature (Q_0, T_0, T_w), process time (τ_{con}); 2) a global calculation cycle is organized in time with a given step δ are found; 3) an internal calculation cycle is organized according to the condition ($x \le L$ and dm HFS >0), where x is the current coordinate in the calculation area, changing within $0 \le x \le L$; L is the length of the cooled part of the desublimator. Within this cycle, the physicochemical characteristics of the gas mixture, the similarity criteria for the flow of the mixture Re, Pr, Nu are calculated, and the heat transfer coefficient α is determined.

Given that in the initial section of the cooling of the gas-vapor mixture, the process proceeds in a non-stationary mode, the calculation of the desublimating HFS mass is found taking into account the proportion: $K=\frac{T_0-T_i}{T_0-T^*}$ where T_i is the current temperature, K; T* is the desublimation temperature, K. Correction K is taken into account until the moment when T_i >T*, further it is assumed that K=1 and the temperature of the gas mixture remains constant and equal to T* as long. Based on the found parameters, the mass transfer coefficient β is calculated taking into account K. The internal loop of the program continues the calculation, taking into account the growth of the HFS layer and the temperature change on the surface of this layer Ts (Fig. 2); 4) after the end of the outer cycle, graphs of the function δ=f(x) are plotted. As an example, in fig. Figures 3 and 4 show graphs of the change in the temperature of the gas-vapor mixture depending on the transverse (y) and longitudinal (x) coordinates in the annular space of the desublimator in the initial non-stationary cooling region for the case of supplying the initial mixture to the desublimator at a mass velocity MHFS wt. It can be seen from the graphs that the length of the non-stationary zone for cooling the vapor-gas mixture to the HFS desublimation temperature along the apparatus axis does not exceed 0.1L

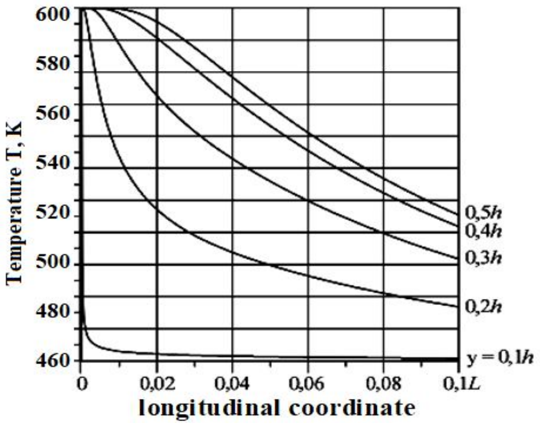

Fig. 3. Dependence of the temperature of the gas-vapor mixture on the transverse coordinate at different values of x

Due to the low gas velocity (w ≤ 0.06 m/s) and the laminar flow regime of the gas-vapor mixture in the annular section of the desublimator (Re ≤ 20), no mixing of gases occurs in the transverse direction (along the y-axis). Therefore, in the transverse direction, heat transfer in the gas flow occurs only due to thermal conductivity.

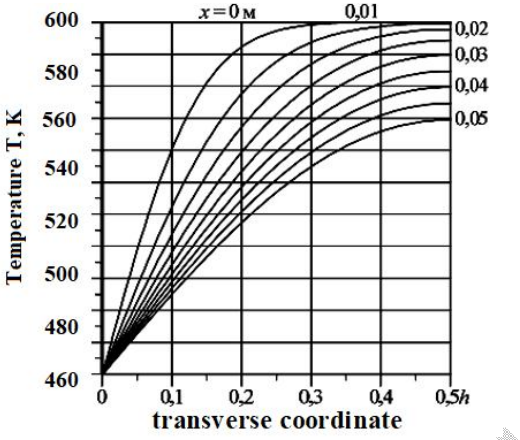

Fig. 4. Dependence of the temperature of the vapor-gas mixture on the longitudinal coordinate for various values of y

The length of the zone of the apparatus, in which the entire HPS is desublimated at a desublimator wall temperature of 243 K and a desublimator load of up to 0.5Gmax according to HPS, is 0.5L in the absence of a region of non-stationary cooling of the vapor-gas mixture (Fig.5) and 0.8L in its presence (Fig.6), and the total length of the desublimation part of the apparatus is L. In this case, the thickness of the HFS layer on both walls of the annular cavity was 0.5h and 0.4h, respectively

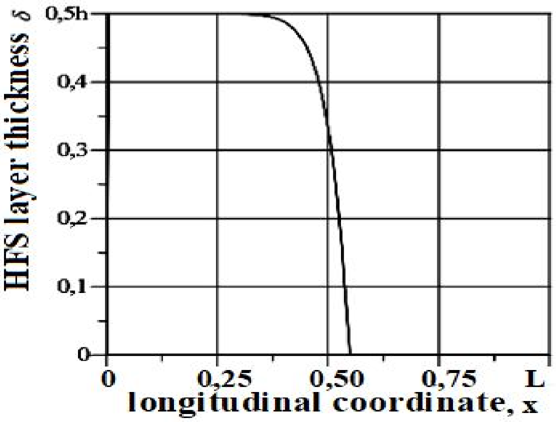

Fig. 5. Dependence of the layer thickness of the desublimated HPS on the longitudinal coordinate in the absence of a region of non-stationary cooling of the vapor-gas mixture at an apparatus wall temperature of 243 K

Graphs of desublimator zones in which the entire HPS is desublimated at apparatus wall temperatures of 243, 253, 263, and 273 K in the presence of a region of non-stationary cooling of the vapor-gas mixture mixtures are shown in fig. 6. From fig. 6 it can be seen that the length of the zone of the apparatus, in which the entire HFS is desublimated when the desublimator is loaded up to 0.5Gmax according to the HFS in the presence of a region of non-stationary cooling, at temperatures of the desublimator wall of 253, 263 and 273 K, is 0.83L, 0.88L and 0, 98L, respectively, with the total length of the desublimation part of the apparatus L. At such desublimation temperatures, the thickness of the HFS layer on both walls of the annular cavity is

0.77h, 0.70h, and 0.65h, respectively, with the width of the annular gap h.

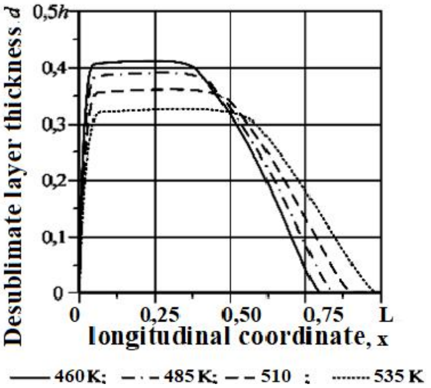

Fig. 6. Dependence of the thickness of the desublimated HFS layer on the longitudinal coordinate in the presence of a region of nonstationary cooling of the vapor–gas mixture at different temperatures of the apparatus wall

Conclusion

A mathematical model has been developed for the process of cooling and desublimation of HFSs from a vapor-gas mixture entering the desublimator, which makes it possible to qualitatively and quantitatively determine the effect of heat transfer, on the considered processes, thereby optimizing the technological parameters of the process and creating a system for automatic control of the desublimation process.

REFERENCES

1. Kamenskyh, D.S., Tkachenko, T.V., Yevdokymenko, V.O., Kashkovskiy, V.I. (2015). Explosive autohydrolysis of pentosan-containing raw material. Catalysis and Petrochemistry. 24. 90—95 [in Ukrainian].
2. Patent application for invention of Ukraine № a 201706242 Yevdokymenko V.O., Kamenskyh D.S., Kashkovskiy V.I., Vakhrin V.V. Method of obtaining of the amorphous silicon dioxide from rice husk [in Ukrainian].
3. Belozerov, B.P., Rusakov, I.Yu., Andreev, G.G., Zhganov, A.N., and Kobzar’, Yu.F., Analysis of a process and equipment for the desublimation of uranium hexafluoride and other volatile fluorides, Tsvetn. Met., 2012, no. 1, p. 58.
4. Raev, V.V., Saprygin, A.V., Tsedilkin, A.P., and Novokshonov, K.V., Determining the thermal conductivity of the solid phase of uranium hexafluoride, Fundam. Probl. Sovrem. Materialoved., 2011, vol. 8, no. 4, p. 125.
5. Vil’nina, A.V. and Liventsov, S.N., An adaptive algorithm for controlling a uranium hexafluoride desublimation unit, Izv. Tomsk. Politekh. Univ., 2008, vol. 312, no. 5, p. 133.
6. Gubanov, S.M., Krainov, A.Yu., and Mazur, R.L., Theoretical and experimental modeling of the cooling of uranium hexafluoride vapor desublimation tanks, Theor. Found. Chem. Eng., 2016, vol. 50, no. 5, p. 766.
7. Gubanov, S.M., Krainov, A.Yu., Mazur, R.L., and Durnovtsev, M.I., Theoretical modeling of cooling of containers for the desublimation of hydrogen fluoride vapors, Theor. Found. Chem. Eng., 2017, vol. 51, no. 3, p. 352.