MODELING OF THERMO- AND MASS TRANSFER PROCESSES AT SUBLIMATION OF MOLECULAR CRYSTALS OF CVD PRECURSORS

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A substantial factor hindering wide use of the volatile metal β-diketonates in real technological CVD processes is insufficient understanding of the mechanism of transport processes in the systems under consideration. The most efficient method to investigate the transport processes during the sublimation of molecular crystals in the flow of inert carrier gas is mathematical modeling because purely experimental investigation connects with substantial methodic and technical difficulties. In the present work we propose a non-stationary mathematical model for the sublimation of molecular crystals in the inert gas flow.

Non-stationary mathematical model includes the equations for convective heat and mass transfer in the channel and the thermal conductivity equations in the compounds and in the holder. These equations are supplemented by the equations for the quasi-stationary profile of gas rate and the equations of heat and mass balance on the sublimation surface.

The reliability of the model and of the numerical algorithm was tested when modeling the sublimation processes of chromium(III) acetylacetonate in argon flow at different temperatures. It follows from figure 1 that the calculated data of the mass rate of sublimation are in good agreement with the results of the experiment.

Two-component model for the sublimation process of copper(II) ketoiminate and chromium(III) acetylacetonate plates in argon flow was used. The calculated results are presented in figures 2 and 3. The calculated quasi-stationary rates of sublimation flows for copper(II) and chromium(III) β-diketonate derivatives differ from those determined experimentally by no more than 10%.

Thus, a two-dimensional non-stationary model of heat and mass transfer is developed for the sublimation of a binary system of metal β-diketonate derivatives in non-uniform flow of inert gas. The results of calculations demonstrated satisfactory agreement with experimental data, which proves the physical adequacy of the proposed model.

The proposed mathematical model can be used for preliminary modeling of the sublimation flows of different metal β-diketonates, for the purpose of choosing temperature regimes, linear dimensions, and properties of the inert gas involved, in order to obtain the desirable concentration relations in technological reactors for the CVD processes.

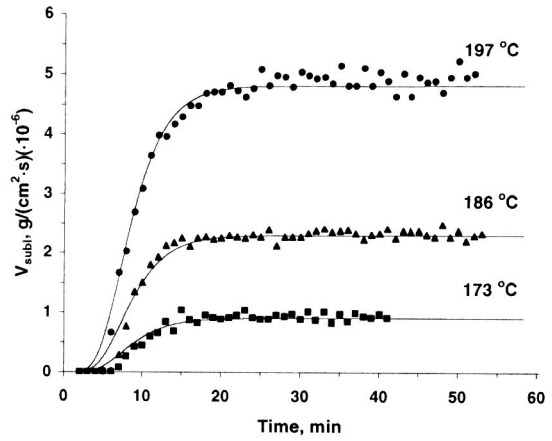

Figure 1. Calculated (continuous lines) and experimental (markers) values of change of the mass sublimation rate of chromium(III) acetylacetonate at different reactor temperature (argon flow rate is 2 l/h).

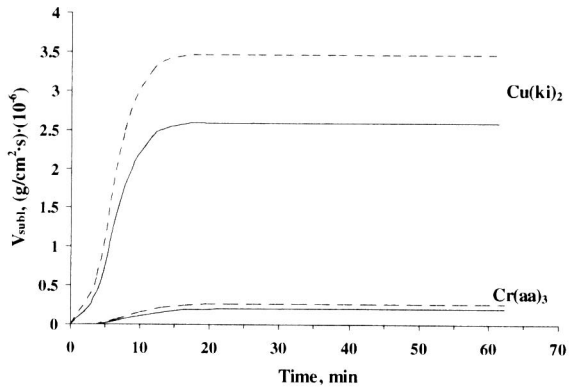

Figure 2. Dependence of mass rate of sublimation on time for copper(II) ketoiminate ($Cu(ki)_2$) and chromium(III) acetylacetonate ($Cr(aa)_3$) at the gas flow temperature of 148°C and gas flow rate of 5 l/h (continuous lines) and 10 l/h (dash lines).

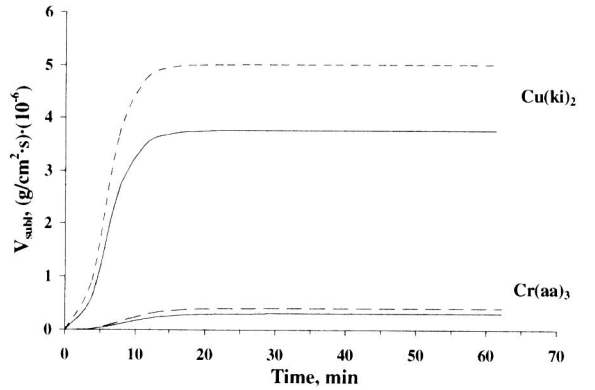

Figure 3. Dependence of mass rate of sublimation on time for copper(II) ketoiminate ($Cu(ki)_2$) and chromium(III) acetylacetonate ($Cr(aa)_3$) at the gas flow temperature of 158°C and gas flow rate of 5 l/h (continuous lines) and 10 l/h (dash lines).

ACKNOWLEDGEMENTS

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