### Mathematical Modeling of Mass-Transfer Processes in the Rotary Calciner, Moscow, Russia – 11491

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# ABSTRACT

The rotary calciner is a device of first stage included in the two-stage installation for liquid radioactive waste immobilization. This device can be regarded as a rotating heating pipe with a little inclination to horizontal. Liquid radioactive waste inputs inside the calciner. While it advancing inside the device, inputted matter is being heated and evaporating. During further motion the solid free-flowing matter remaining after evaporation is undergoing chemical changes. The main object for modeling of the calciner process is to determine main characteristics of calcined product in dependence on parameters of the device and parameters of inputted matter. The developed mathematical model, making it possible to determine main process characteristics influencing on output quality, is being considered in the present paper.

# **INTRODUCTION**

Radioactive waste preparation for long-term storage or disposal includes waste conversion into chemically, thermally, and radiation-stable matter forms which preserve its properties during transfer, storage, and disposal [1]. Generation of matter with pointed out above properties in a case of liquid radioactive waste (LRW) consist of water removal, decomposition of salts and inclusion of received solid matter into vitreous matrix. Such process is called as a process of liquid radioactive waste immobilization.

The perspective technological scheme of the process is two-stage processing sequence consisting of a rotary calciner (first-stage device) and an induction melter with "cold" crucible (second-stage device) [2]. The first-stage device realizes LRW evaporation and salts decomposition. The second-stage device realizes vitrification of received after first stage matter.

Stable and coordinated processing of any device in the installation is necessary for qualitative fixation of radioactive waste in solid matrix. Wrong processing regime selection for one of the devices in the installation can lead to worsening of received product quality or failing the installation. The necessity of analyzing processing regimes for predicting calcined product characteristics leads to a necessity for development of mathematical models. This paper describes the developed mathematical model of the first-stage device – the rotary calciner.

### THE ROTARY CALCINER

The rotary calciner can be regarded as a rotating cylindrical pipe with a small inclination angle of its axis to horizontal (Fig. 1). The pipe is placed into a heating device.



Fig. 1. Scheme of a rotary calciner.  $J_{out}$  is an output flow;  $J_{in}$  is an input flow;  $\alpha$  is the inclination angle of the retort axis to the horizontal.

LRW is inputted in the calciner. During advancement inside the device LRW is being heated and evaporating. Forming as a result of evaporation gaseous phase is being removed from the device. The matter remained after evaporation continues motion inside the calciner. During advancement inside a heated pipe it is undergoing chemical changes: salt decompositions with oxides formation (calcination) take place in the pipe. Temperature of a part of the device's tube is to be kept up rather high for providing the calcinations conditions. The chemically stable free-flowing matter is to be formed at the output of the calciner.

### **PROBLEM DEFINITION**

Mathematical model of the rotary calciner is required to estimate main characteristics of output flow (e.g., calcinations degree, moisture) depending on parameters of inputted matter and characteristics of the device.

The flow of LRW fed into the device undergoes some transformations as a result of contact with heated inner surface of the calciner's pipe. For describing LRW transformations during advancement inside the device let's mark out a certain volume of inputted matter. The marked out matter is being heated while it moving down the length of the calciner tube. When material's temperature reaches the boiling point, evaporation of liquid phase becomes important, and we can assume that the heat from the inner surface of the device is consumed by evaporation. This

process (evaporation) proceeds until all liquid has evaporated. Solid particles remaining after the evaporation is being subjected to calcination during further motion along the calciner. Let's suppose that the designated above volume of inputted matter is rather small, so we can assume the transformation processes listed above take place sequentially. This means that the length of the calciner can be divided into three parts corresponding to three main processes occurring in the calciner sequentially: heating, evaporation, and calcination (Fig. 2).



Fig. 2. Division of the calciner into zones.

Each of these three processes has its own features and should be considered separately from the rest.

The objective of the present work is to estimate dimensions of designated above zones and to find residence time of matter in each of them in dependence on parameters of calciner and parameters of input flow. Radius of the calciner's pipe, temperature of its inner surface and angle to horizontal are the parameters of the device that are used in the model, and feed rate and thermodynamic properties of feeding matter are the parameters of input flow. Thus, the developed mathematical model include three parts corresponding to three main processes occurred in the calciner. The objective of each part of the model is to estimate the length of the zone where the process proceeds and residence time of matter in the zone in dependence on parameters of calciner and parameters of feeding flow.

# MATTER BEHAVIOR FEATURES INSIDE THE ROTATING CYLINDER

In accordance with [3, 4] there are two extreme regimes of liquid motion in the rotating horizontal cylinder. The first one corresponds to the case when all matter is uniformly distributed on inner surface of retort. Second extreme regime corresponds to the case when all liquid is situated at the bottom of cylinder. Any regime realization depends on ratio of forces having an effect upon liquid. In the considered system the forces having an effect on liquid are next: gravity, viscous forces and centripetal force.

Second extreme regime appears when centripetal force is considerably smaller than gravity and viscous forces. Rotating velocities of the rotary calciner in real technological process are smaller than 1 revolution per second, and inner diameter of the retort is 30-40 centimeters.

An estimation of forces having an effect upon liquid moving in the device with above parameters is performed. There was found out that it is second extreme regime that is realized in the rotary calciner. It means, the matter inputted inside the calciner is situated at the bottom of the device.

For determination of average rate of liquid motion along the axis of cylinder, the described in [5] model of liquid layer motion with free surface on an inclined plane can be used. Liquid in the rotary calciner advances not on an inclined plane and on an inclined cylinder. However, using of the model [5] for describing liquid dynamic in the calciner is rightfully as matter fills only a little part of volume inside the cylinder in real technological regimes of LRW reprocessing.

Liquid layer motion rate V inside the inclined cylinder that is determined in [5] can be written as Equation 1.

$$V(y,z) = \frac{g\sin\alpha}{2\nu} z(h(y) - z) \qquad , \qquad (Eq. 1)$$

g is free fall acceleration; v is kinematic viscosity; other parameters are given at Fig. 3.



Fig. 3. A cut set of the pipe by perpendicular to its axis plane.

The equation 1 allows defining a quantity of liquid flowing through cross-section of a cylinder per time unit. To determine this value it is necessary to take integral of velocity (1) over the perpendicular to cylindrical axis section of liquid layer. As a result of calculation, maximum thickness of liquid layer (Eq. 2) and average rate of matter motion along the axis of a cylinder (Eq. 3) were determined.

$$h_m = 1.28 \left( \frac{Q}{\sqrt{R}} \frac{v}{g \sin \alpha} \right)^{2/7},$$
(Eq. 2)  
$$V_{av} = 0.49 \left( \frac{Q}{\sqrt{R}} \right)^{4/7} \left( \frac{g \sin \alpha}{v} \right)^{3/7} ,$$
(Eq. 3)

R is inner radius of the calciner's pipe.

#### **HEATING OF LIQUID**

Let's consider that a flow of LRW with temperature  $T_0$  is inputted into the calciner with feeding rate Q. Matter temperature of a selected volume of liquid is arising while matter contacting with hot pipe surface. Let's determine temperature of the volume in dependence on term of matter location in the device.

As it follows from a heat balance condition, alteration of internal energy of the marked out volume of liquid per time unit equals to heat flow through the contact surface between the liquid and the pipe. Heat flow through the contact surface is proportional to temperature gradient near the surface. As it was mentioned above, liquid layer thickness is much smaller than radius of retort in real technological conditions of LRW reprocessing, therefore, temperature gradient near the inner surface of the device can be replaced by temperature difference divided by characteristic length (Eq. 4).

$$q = -\theta \left. S \frac{\partial T}{\partial x} \right|_{S} \approx -\theta \left. S \frac{T - T_{sur}}{\delta} \right|_{S}$$
(Eq. 4)

S is the contact surface;  $\theta$  is heat conductivity of LRW matter; T is average temperature of the marked out liquid volume;  $T_{sur}$  is temperature of the inner surface of the device;  $\delta$  is characteristic thickness of the matter layer where temperature changes.

The solution of the heat balance equation in common with the equation 4 determines the next time dependence of average temperature in marked out matter volume.

$$T = T_{sur} - (T_{sur} - T_0) \exp\left(-\frac{2\chi}{h_m \delta}t\right); \quad \chi = \frac{\theta}{c\rho} \quad ,$$
 (Eq. 5)

where  $\chi$ , *c* and  $\rho$  are temperature conductivity, specific heat capacity and density of LRW matter correspondingly;  $h_m$  is maximum thickness of the matter layer determining from equation 2.

The only unknown value in right-hand part of the equation 5 is characteristic length  $\delta$  of temperature changing. Characteristic length  $\delta$  can be determined from accurate solution of heat-transfer equation for plane liquid layer with thickness  $h_m$  in common with given temperature at one bound. Determined in that way characteristic length is following:

$$\delta = 0.36h_m. \tag{Eq. 6}$$

The equation 5 defines the time dependence of matter temperature in the selected volume. The equation 5 allows to estimate the time  $\tau_h$  required for LRW matter heating to boiling-point  $T_I$ .

$$\tau_{h} = 0.3 \frac{1}{\chi} \left( \frac{Q}{\sqrt{R}} \frac{v}{g \sin \alpha} \right)^{4/7} \ln \frac{T_{sur} - T_{0}}{T_{sur} - T_{1}} , \qquad (Eq. 7)$$

Besides, by using the equation 3 for average rate of matter motion inside the calciner, it is possible to determine length  $l_h$  of the pipe where matter is heating to boiling point.

$$l_{h} = 0.15 \frac{1}{\chi} \left(\frac{Q}{\sqrt{R}}\right)^{8/7} \left(\frac{\nu}{g \sin \alpha}\right)^{1/7} \ln \frac{T_{sur} - T_{0}}{T_{sur} - T_{1}} \quad .$$
(Eq. 8)

Thus, the desired values, length of the heating zone and resident time in this one, are determined.

### LIQUID PHASE EVAPORATION

As in case of matter heating, marked out matter volume is looked at. The matter volume with temperature  $T_{sat}$  which equal to boiling point is advancing inside the pipe, whose surface is heated to temperature  $T_{sur}$ . If  $T_{sur} > T_{sat}$ , then advancing matter absorbs heat. The heat is spent to liquid phase evaporation.

Let's determine heat quantity absorbed by the matter during advancement inside the calciner. Normal regime of the rotary calciner processing is characterized by bubble liquid evaporation. Heat quantity absorbed by a matter volume in a time unit in this case is following:

$$\frac{dq}{dt} = \gamma S(T_{sur} - T_{sat}) \quad , \tag{Eq. 9}$$

q is absorbed heat, S is the surface area of contact between marked up matter volume and inner surface of the calciner;  $\gamma$  is heat transfer coefficient.

Let's estimate heat transfer coefficient by using the half-empirical relation [6]. This relation is used for water evaporation dynamic analysis.

$$\gamma = 4.34 j^{0.7} \left( p^{0.14} + 1.35 \times 10^{-2} p^2 \right) \quad , \tag{Eq. 10}$$

*j* is heat flow density (Wt/m<sup>2</sup>); *p* is pressure in liquid (MPa);  $\gamma$  is heat transfer coefficient (Wt/(m<sup>2</sup>K)).

The equation 9 taking into account the equation 10 is transformed to the next form

$$\frac{dq}{dt} \approx AS\Delta T^3; \quad \Delta T = T_{sur} - T_{sat}; \quad A \approx 9.6 \quad \text{Wt/(m}^2\text{K}^3) \qquad . \tag{Eq. 11}$$

All-liquid evaporation term  $\tau_{ev}$  can be determined taking into account that absorbed heat is used to perform liquid phase to gaseous phase:

$$\tau_{ev} = 0.96 \frac{r\rho}{A\Delta T^3} \left(\frac{Q}{\sqrt{R}}\right)^{2/7} \left(\frac{v}{g\sin\alpha}\right)^{2/7} \left(1 - \left(\frac{c_0}{\rho_s}\right)^{2/3}\right)$$
, (Eq. 12)

*r* is heat of vaporization;  $\rho$  is density of vaporizable liquid;  $\rho_s$  is density of solid matter in the marked out volume;  $c_0$  is concentration of solid matter in two-component system.

Length of the calciner tube section, where evaporation takes place, can be determined by using average rate (Eq. 4):

$$l_{ev} = \int_{0}^{\tau_{ev}} V_{av}(t) dt$$
 . (Eq. 13)

The average rate in the equation 13 depends on thickness of moving matter layer which is being decreased during the motion because of liquid evaporation.

Besides, during liquid phase evaporation, viscosity of moving matter is being changed.

Matter advancing inside the rotary calciner can be considered as a two-component system consisting of liquid and solid phases. For determination of matter viscosity, it is possible to estimate influence of solid particles contained in the matter on liquid phase motion. Such estimation has been performed in [5]. Dependence of viscosity on concentration of solid matter containing in the system is following:

$$\nu = \nu_{w} \left( 1 + \frac{\nu_{0} - \nu_{w}}{\nu_{w}} \frac{c}{c_{0}} \right)$$
(Eq. 14)

 $v_w$  is viscosity of water;  $v_0$  is initial viscosity of matter; *c* is concentration of solid matter in twocomponent system;  $c_0$  is concentration of solid matter in two-component system. Substitution the average rate (Eq. 3) and viscosity (Eq. 14) into the equation 13 lead to the next result for length of the calciner's zone where evaporation proceeds:

$$l_{ev} = 0.16 \frac{r\rho}{A\Delta T^3} \left(\frac{Q}{\sqrt{R}}\right)^{6/7} \left(\frac{g\sin\alpha}{v_w}\right)^{1/7} \frac{\rho r}{A\Delta T^3} f\left(\frac{\upsilon_0 - \upsilon_w}{\upsilon_w}, \frac{c_0}{\rho_1}\right) , \qquad (Eq. 15)$$

$$f(K, M) = 1 - 2K - M^2 + 2KM + 2K^2 \ln\frac{1+K}{M+K} .$$

The equation 15 in common with the equation 12 gives the solution for the part of the model which describes matter behavior in evaporation zone.

# SOLID MATTER MOTION

Solids remained after full evaporation of liquid phase continues motion inside the device. During LRW reprocessing chemical compounds (salts) decomposes exactly at the last section of the rotary calciner. Let's determine resident term of matter inside this zone. Length of this zone can be considered a definite, as lengths of other two zones are defined and length of the calciner is known.

Motion equations for solid particle were developed for describing matter movement along the last section of the device. These equations use cylindrical coordinate system and take into account gravity, friction and rotating. As variables radial  $v_{\tau}$  and longitudinal  $v_z$  particle velocities and polar angle  $\varphi$  were chosen. The system of equations described above has next form:

$$\begin{cases} \frac{dv_z}{dt} = g \sin \alpha - \mu \left( g \cos \alpha \cos \varphi + \frac{v_\tau^2}{R} \right) \frac{v_z}{\sqrt{v_z^2 + (v_\tau - \omega R)^2}} \\ \frac{dv_\tau}{dt} = -g \cos \alpha \sin \varphi - \mu \left( g \cos \alpha \cos \varphi + \frac{v_\tau^2}{R} \right) \frac{(v_\tau - \omega R)}{\sqrt{(v_\tau - \omega R)^2 + v_z^2}} , \qquad (Eq. 16) \\ \frac{d\varphi}{dt} = \frac{v_\tau}{R} \end{cases}$$

 $\mu$  is friction coefficient;  $\omega$  is rotating velocity of the calciner's pipe; *R* is radius of the pipe.

The received system of motion equations for solid particle inside the calciner is non-linear. Analyze of the system showed that the system has the only stable stationary solution (Eq. 17).

$$\begin{cases} v_{\varphi} = 0\\ \sin \varphi = \sqrt{\frac{\mu^2 - tg^2 \alpha}{\mu^2 + 1}}\\ v_z = \omega R tg \alpha \sqrt{\frac{\mu^2 + 1}{\mu^2 - tg^2 \alpha}} \end{cases}$$
(Eq. 17)

Condition of stationary solution existence is following:

$$\mu > tg\alpha \qquad . \tag{Eq. 18}$$

Let's determine the residence term  $\tau_s$  of particle inside the calciner in stationary regime. Its value equals to the length of the last section of the device divided by radial velocity of the particle (19):

$$\tau_s = \frac{L}{\omega R t g \alpha} \sqrt{\frac{\mu^2 - t g^2 \alpha}{\mu^2 + 1}} \qquad (Eq. 19)$$

L is length of the tube section along the cylinder's axis where solid particles advance.

The residence time of solids inside the calciner is defined by the equation 19. This parameter is very important in practical application of the model. So important characteristics of output flow as moisture and degree of calcination (or decomposition degree of contained in the output matter salts) depend on temperature of heated surface of the pipe and on indicated by the equation 19 time. Length of the calcination zone is estimated by subtraction lengths of other zones from length of calciner. Therefore, expressions included in the model of rotary calciner characterize matter behavior in the device and can be used for estimation of some important properties of output flow.

### CONCLUSION

The developed mathematical model of the rotary calciner describes main features of matter behavior in the device in dependence on inputted flow and calciner geometry. LRW is fed into the device. During moving, the matter is being heated and evaporating. Freeflowing substance received after evaporation is undergoing chemical changes while it contacting with heated surface of the calciner.

Three sequentially passing processes (heating, evaporation, decomposition-calcination) are described in the developed model of the rotary calciner. The length of the device can be divided into three sections corresponding to each of these process. Characteristic lengths of device's

sections and residence times of matter in these sections are estimated. Relations received in the model can be used for estimation so important parameters of output flow as moisture or calcination (decomposition) degree. The model can be applied to practical tasks appearing in the time of designing of new or optimization of existent calciners.

# REFERENCES

- 1. IAEA Safety Standards, No WS-R-2, "Predisposal Management of Radioactive Waste, Including Decommissioning", Vienna (2000)
- G.B. BORISOV, N.D. MUSATOV, P.P. POLUEKTOV, T.V. SMELOVA, L.P SUKHANOV, I.N. SHESTOPEROV, "Development and application outlook on compact installations for liquid high-level waste immobilization", X International Conference "Nuclear Technologies Safety – Radwaste and spent nuclear fuel Management", St. Peterburg (2007)
- 3. J.A. DEIBER, R.L. CERRO, "Viscous Flow with a Free Surface Inside a Horizontal Rotating Drum. 1. Hydrodynamics" Ind. Eng. Chem., Fundam., Vol 15, No 2 (1976)
- 4. YU. V. NAUMENKO. "Numerical Calculations of a Fluid Partially Filling a Horizontal Rotating Heat-Exchange Cylinder" Journal of Engineering Physics and Thermophysics, volume 74, number 3 (2001)
- 5. L.D. LANDAU, E.M. LIFSHITZ, "Fluid Mechanics". Vol. 6 (1rst ed.). Butterworth-Heinemann, ISBN 978-0-080-33933-7 (1987)
- 6. P.L. KIRILLOV, YU.S. YURIEV, V.P. BOBKOV, "Spravochnik po teplohydravlichesckim raschetam" Moscow (1984)