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An Absorption-Adsorption Apparatus for Gases Purification from SO_2 in Power Plants

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Abstract

Different companies propose methods and apparatuses for waste gases purification from SO_2 using two-phase absorbent ($CaCO_3$ suspension).The basic problem of the carbonate absorbents is that its chemical reaction with SO_2 leads to CO_2 emission, which leads to similar ecological problems. The use of synthetic anionites as adsorbents for gas purification from SO_2 provides possibilities for adsorbent regeneration. In a proposed method, the waste gas purification is realized in two steps–physical absorption of SO_2 with water in a counter-current column and chemical adsorption of HSO_3^- from the water solution by synthetic anionite particles. The adsorbent regeneration is made with NH_4OH solution. The obtained $(NH_4)_2SO_3$ (NH_4HSO_3) is used (after reaction with HNO_3) for production of concentrated SO_2 (gas) and NH_4NO_3 (solution). The proposed method permits to be used the absorption columns, where are used the $CaCO_3$ suspensions. In this paper, the efficiency of the waste gases purification from SO_2 is increased in an absorption-adsorption apparatus, where the absorption is realized in co-current flows and the adsorption takes place in the flexible adsorbent. For this is proposed a new absorption-adsorption column apparatus with bubbling plates. A mathematical model of the absorption-adsorption process is presented, too.

Subject Areas

Chemical Engineering & Technology, Industrial Engineering

Keywords

Absorption, Adsorption, Gas Purification, SO_2, Power Plant, Convection- Diffusion Model, Average Concentration Model

1. Introduction

Different companies (Babcock & Wilcox Power Generation Group, Inc., Alstom

Power Italy, Idreco-Insigma-Consortium) propose methods and apparatuses for waste gases purification from SO_2 using two-phase absorbent ($CaCO_3$ suspension) [1]. The basic problem of the carbonate absorbents is that its chemical reaction with SO_2 lead to CO_2 emission (every molecule of SO_2 absorbed from the air is equivalent to a molecule of CO_2 emitted in the air), because the ecological problems (greenhouse effects) of SO_2 and CO_2 are similar. The large quantity of by-products is a problem, too. Another drawback of these methods is the impossibility for regeneration of the absorbents.

The theoretical analysis [2]-[8] of the method and apparatus for waste gases purification from SO_2 using two-phase absorbent ($CaCO_3$ suspension) shows the possibility to use an absorption-adsorption method. The use of synthetic anionites (basic anion-exchange resins-R-OH form of Amberlite, Duolite, Kastel, Varion, Wofatit) as adsorbents [9] [10] [11] for gas purification from SO_2 provides possibilities for adsorbent regeneration. In the proposed method [12] [13] the waste gas purification is realized in two steps–physical absorption of SO_2 with water and chemical adsorption of HSO_3^- from the water solution by synthetic anionite particles in a fixed bed adsorber. The adsorbent regeneration is made with NH_4OH solution. The obtained $(NH_4)_2SO_3$ (NH_4HSO_3) is used (after reaction with HNO_3) for production of concentrated SO_2 (gas) and NH_4NO_3 (solution). The proposed patent [12] makes it possible to create a waste-free technology for waste gases purification from sulfur dioxide by means of regenerable absorbent and adsorbent. The proposed method [13] permits to be used for the absorption columns, where are used the $CaCO_3$ suspensions.

The efficiency of the waste gases purification from SO_2 can be increased if the absorption is realized in co-current flows and the adsorption takes place in the flexible adsorbent. For this is proposed a new absorption-adsorption column apparatus with bubbling plates.

2. An Absorption-Adsorption Apparatus

In the proposed absorption-adsorption method for waste gases purification from sulfur dioxide [12] [13], the absorption is realized in the counter-current absorber (where practical gas velocity does not exceed 5 $m \cdot s^{-1}$) and the adsorption is carried out in a fixed bed adsorber. The efficiency of the process can be increased if the absorption is realized in co-current flows and the adsorption takes place in the flexible adsorbent. For this it can use a new absorption- adsorption apparatus with bubbling plates. The bubbling of the gas at a plate through a layer of aqueous suspension of synthetic anionite allows an increasing of the gas velocity (reduction of the diameter of the absorption column), elimination of the adsorption column and carrying out the adsorption in a flexible adsorbent. The movement of the gas between the plates leads to mixing in the gas phase, which increases the absorption rate because the absorption of sulfur dioxide in water is limited by mass transfer in the gas phase.

The new absorption-adsorption apparatus with bubble plates [14] is shown in Figure 1. The gas enters tangentially into the column 1 through the inlet 2,

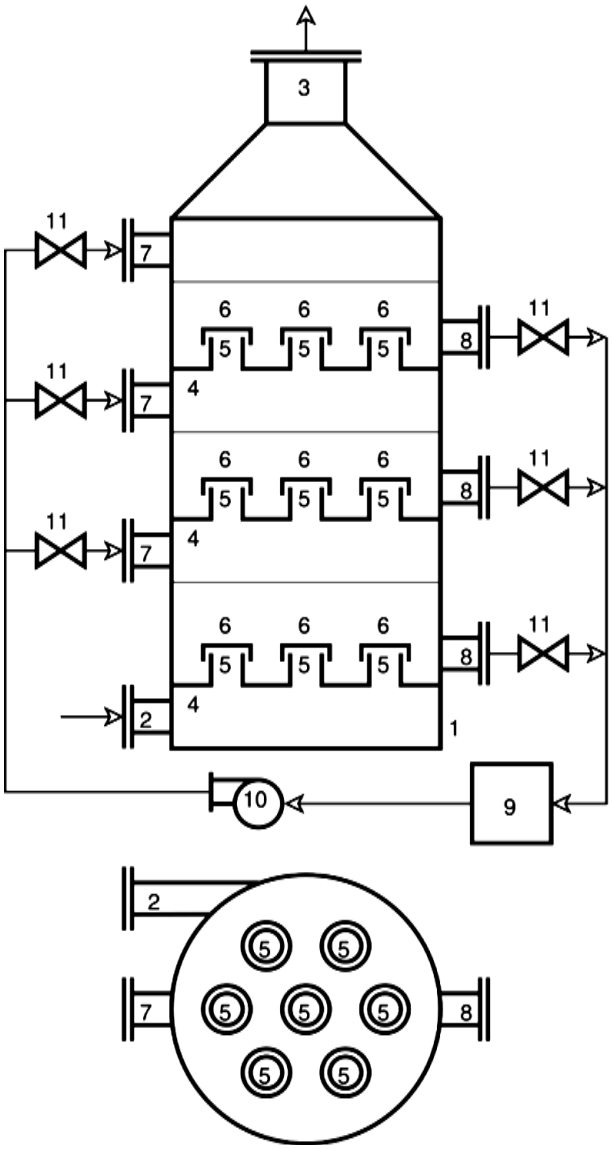

Figure 1. Absorption-adsorption apparatus.

passes through the distribution pipes 5, concentric bubble caps 6 of the plates 4 and exits the column through the outlet 3. The aqueous suspension of synthetic anionite enters the column 1 via the valve 11 and the pipes 7 and creates of plates 4 layer with a certain thickness. After saturation of the adsorbent with sulfur dioxide, the aqueous suspension is output from the column through the pipes 8 and valves 11 and enters in the system 9 for the regeneration of the adsorbent. The suspension of the regenerated adsorbent is removed from 9 by pump 10 is returned to the plates 4 in the column 1.

The operation of the absorption-adsorption apparatus is the following cyclic scheme:

1. Supply all the plates with the necessary amount (volume) of aqueous suspension of synthetic anionite.
2. Start the absorption-adsorption process and monitor the SO_2 concentration of

the exit 3.

3. When the increasing the SO_2 concentration at the outlet of the gas 3 exceeds permissible limits, the suspension of the first (bottom) plate is transferred in the regeneration system 9 and the plate is loaded with a new (regenerated) suspension.
4. When the increasing the SO_2 concentration at the outlet of the gas 3 exceeds permissible limits, the suspension of the second (next) plate is transferred in the regeneration system 9 and the plate is loaded with a new (regenerated) suspension.
5. The procedures are repeated until reaching the top plate, then starts again from the first plate.

3. Absorption-Adsorption Process Modeling

Tangentially supplying of the gas in the column [15] reduces the radial non-uniformity of the velocity at the column cross-sectional area and the gas velocity is a constant $\left(u_1 = const.\right)$, practically. Under these conditions, the concentration of SO_2 in the gas phase is changed only in the height of the column ($c_{11} = c_{11}\left(t, x\right)$), which increases the rate of mass transfer rate. On the other hand the gas bubbling creates an ideal mixing regime in the liquid phase and the concentration of SO_2 in the liquid phase is $c_{12} = c_{12}\left(t\right)$. The concentration of SO_2 in the solid phase (capillaries) is c_{13} and the concentration of active sites in the adsorbent is c_{23}.

The interphase mass transfer rate of SO_2 from the gas to the liquid is $k_0\left(c_{11} - \chi c_{12}\right)$, while the liquid to the adsorbent is $k_1\left(c_{12} - c_{13}\right)$. The chemical reaction rate of the SO_2 with the adsorbent is $k\, c_{13} c_{23}$.

The modeling of a non-stationary (as a result of the adsorbent saturation) absorption-adsorption process on the plate number $n\left(n = 1, \cdots, N\right)$, for gas purification from SO_2 [14], uses a combination of the physical absorption and chemical adsorption models [1]:

$$\begin{aligned}
&\frac{\partial c_{11}^{(n)}}{\partial t} + u_1 \frac{\partial c_{11}^{(n)}}{\partial z} = D_1 \frac{\partial^2 c_{11}^{(n)}}{\partial z^2} - k_0\left(c_{11}^{(n)} - \chi c_{12}^{(n)}\right);\\
&\frac{\mathrm{d}c_{13}^{(n)}}{\mathrm{d}t} = k_1\left(c_{12}^{(n)} - c_{13}^{(n)}\right) - k c_{13}^{(n)} c_{23}^{(n)}; \quad \frac{\mathrm{d}c_{23}^{(n)}}{\mathrm{d}t} = -k c_{13}^{(n)} c_{23}^{(n)};\\
&t = 0, \quad c_{11}^{(n)} \equiv c_{11}^{0}, \quad c_{13}^{(n)} \equiv 0, \quad c_{23}^{(n)} \equiv c_{23}^{0};\\
&z = 0, \quad c_{11}^{(n)}\left(0\right) \equiv c_{11}^{(n-1)}\left(l\right), \quad 0 \equiv \left(\frac{\partial c_{11}^{(n)}}{\partial z}\right)_{z=0}, \quad c_{11}^{(1)}\left(0\right) \equiv c_{11}^{0}; \quad n = 1, \cdots, N.
\end{aligned} \tag{1}$$

In (1) $z = 0$ is the entrance to the gas input of the plate number $n\left(n = 1, \cdots, N\right)$, l is the distance between the plates, D_1 is the diffusivity of SO_2 in the gas phase.

The concentration of SO_2 in the water of each plate is determined by the amount of absorbed SO_2 (W_1) and its distribution between liquid phase (W_2), the solid phase (capillaries) (W_3^1) and adsorbed on the surface of the capillaries is (W_3^2):

$$W_1 = V_1\left[c_{11}^{(n)}(0) - c_{11}^{(n)}(l)\right],\ W_2 = V_2 c_{12}^{(n)},\ W_3^1 = V_3 c_{13}^{(n)},\ W_3^2 = V_3\left(c_{23}^0 - c_{23}^{(n)}\right), \quad (2)$$

i.e.

$$c_{12}^{(n)} = \frac{V_1\left[c_{11}^{(n)}(0) - c_{11}^{(n)}(l)\right] - V_3 c_{13}^{(n)} - V_3\left(c_{23}^0 - c_{23}^{(n)}\right)}{V_2}, \quad (3)$$

where V_1, V_2, V_3 are the gas, liquid and solid phase volumes on the plates.

4. Generalized Analysis

The use of dimensionless (generalized) variables [16] allows to make a qualitative analysis of the models (1), (3), where as characteristic scales are used the inlet and initial concentrations, the characteristic time t_0 (saturation time of the adsorbent) and the distance between the plates l:

$$T = \frac{t}{t_0},\quad Z = \frac{z}{l},\quad C_{11} = \frac{c_{11}}{c_{11}^0},\quad C_{12} = \frac{c_{12}\chi}{c_{11}^0},\quad C_{13} = \frac{c_{13}\chi}{c_{11}^0},\quad C_{23} = \frac{c_{23}}{c_{23}^0}. \quad (4)$$

When (4) is put into (1), (3), the model in generalized variables takes the form:

$$\begin{aligned}
&\gamma\frac{\partial C_{11}^{(n)}}{\partial T} + \frac{\partial C_{11}^{(n)}}{\partial Z} = \mathrm{Pe}^{-1}\frac{\mathrm{d}^2 C_{11}^{(n)}}{\mathrm{d}Z^2} - K_0\left(C_{11}^{(n)} - C_{12}^{(n)}\right);\\
&\frac{\mathrm{d}C_{13}^{(n)}}{\mathrm{d}T} = K_1\left(C_{12}^{(n)} - C_{13}^{(n)}\right) - KC_{13}^{(n)}C_{23}^{(n)};\quad \frac{\mathrm{d}C_{23}^{(n)}}{\mathrm{d}T} = -K\alpha^{-1}C_{13}^{(n)}C_{23}^{(n)};\\
&T = 0,\quad C_{11}^{(n)} \equiv 1,\quad C_{13}^{(n)} \equiv 0,\quad C_{23}^{(n)} \equiv 1.\\
&Z = 0,\quad C_{11}^{(n)}(0) \equiv C_{11}^{(n-1)}(1),\quad 0 \equiv \left(\frac{\mathrm{d}C_{11}^{(n)}}{\mathrm{d}Z}\right)_{Z=0},\quad C_{11}^{(1)}(0) \equiv 1;\quad n = 1,\cdots,N.
\end{aligned} \quad (5)$$

$$C_{12}^{(n)} = \frac{\chi V_1\left[C_{11}^{(n)}(0) - C_{11}^{(n)}(l)\right] - V_3 C_{13}^{(n)} - \alpha V_3\left(1 - C_{23}^{(n)}\right)}{V_2},\quad n = 1,\cdots,N. \quad (6)$$

The following parameters are used in (5), (6):

$$\mathrm{Pe} = \frac{u_1 l}{D_1},\quad K = kt_0 c_{23}^0,\quad K_0 = \frac{k_0 l}{u_1},\quad K_1 = k_1 t_0,\quad \alpha = \frac{\chi c_{23}^0}{c_{11}^0},\quad \gamma = \frac{l}{t_0 u_1}. \quad (7)$$

Practically $0 = \gamma < 10^{-2}, 0 = \mathrm{Pe}^{-1} < 10^{-2}$ and as a result

$$\begin{aligned}
&\frac{\mathrm{d}C_{11}^{(n)}}{\mathrm{d}Z} = -K_0\left(C_{11}^{(n)} - C_{12}^{(n)}\right);\\
&Z = 0,\quad C_{11}^{(n)}(0) \equiv C_{11}^{(n-1)}(1),\quad C_{11}^{(1)}(0) \equiv 1;\quad n = 1,\cdots,N.
\end{aligned} \quad (8)$$

$$\begin{aligned}
&\frac{\mathrm{d}C_{13}^{(n)}}{\mathrm{d}T} = K_1\left(C_{12}^{(n)} - C_{13}^{(n)}\right) - KC_{13}^{(n)}C_{23}^{(n)};\quad \frac{\mathrm{d}C_{23}^{(n)}}{\mathrm{d}T} = -K\alpha C_{13}^{(n)}C_{23}^{(n)};\\
&T = 0,\quad C_{13}^{(n)} \equiv 0,\quad C_{23}^{(n)} \equiv 1.
\end{aligned} \quad (9)$$

The solution of the equations of the model (6), (8), (9) uses a two-stage algorithm. In the first stage must be solved the equations for $n = 1$. In the second stage must be applied consistently for every plate the algorithm for $n = 1$.

Algorithm of the solution

1. Put $n = 1$.
2. Put $C_{12}^{(1)} = X_i = 0.1i, i = 1,\cdots,10.$

3. The solution of (8) leads to
$C_{11}^{(1i)} = C_{11}^{(1)}\left(Z, X_i\right), C_{11}^{(1)}\left(0, X_i\right), C_{11}^{(1)}\left(1, X_i\right), i = 1, \cdots, 10.$
4. The solution of (9) leads to $C_{13}^{(1i)} = C_{13}^{(1)}\left(T, X_i\right), C_{23}^{(1i)} = C_{23}^{(1)}\left(T, X_i\right), i = 1, \cdots, 10.$
5. The solutions in 3 and 4 must be introduced in (6) and as a result is obtained
$C_{12}^{(1i)} = C_{12}^{(1)}\left(T, X_i\right), i = 1, \cdots, 10.$
6. Put $T = T_i = 0.1j,\ j = 1, \cdots, 10$ in $C_{12}^{(1i)} = C_{12}^{(1)}\left(T, X_i\right), i = 1, \cdots, 10$ an as a result is obtained $\overline{C}_{12}^{(1i)} = C_{12}^{(1)}\left(T_j, X_i\right), i = 1, \cdots, 10,\ j = 1, \cdots, 10.$
7. A polynomial approximation $P_{12}^{(1j)}\left(T_j, X\right), j = 1, \cdots, 10$ of $\overline{C}_{12}^{(1i)} = C_{12}^{(1)}\left(T_j, X_i\right),\ \ i = 1, \cdots, 10,\ j = 1, \cdots, 10$ with respect to X must be obtained.
8. The solutions of the equations $X = P_{12}^{(1j)}\left(T_j, X\right), j = 1, \cdots, 10$ with respect to X permit to be obtained $X_j,\ j = 1, \cdots, 10$ and the obtained solutions must be denoted as $X_j = C_{12}^{(1)}\left(T_j\right), j = 1, \cdots, 10.$
9. A polynomial approximation of $C_{12}^{(1)}\left(T_j\right), j = 1, \cdots, 10$ with respect to T permit to be obtained $C_{12}^{(1)}\left(T\right) = C_{12}^{(1)}\left(T_j\right), j = 1, \cdots, 10.$
10. The introducing of $C_{12}^{(1)}\left(T\right)$ in (8) and (9) permits to be obtained its solutions for $n = 1$.
11. The obtained solution of (8) $C_{11}^{(1)} = C_{11}^{(1)}\left(T, Z\right)$ in 10 permits to be obtained $C_{11}^{(1)} = C_{11}^{(1)}\left(T, 1\right)$ and as a result to be used the algorithm 1-11 for consistent solutions of the equations set (6), (8) for $n = 2, \cdots, N.$

5. Parameters Identification

The parameters in the model (6), (8), (9), which are subject to experimental determination are K, K_0, K_1. They may be obtained from experimental data of the SO_2 concentration at the gas outlet from the first plate $C_{11\text{exp}}^{(1j)} = C_{11}^{(1)}\left(T_j, 1\right), T_i = 0.1j,\ j = 1, \cdots, 10$, where $T = 1$ is the time for the fully saturation of the adsorbent on the first plate. For this purpose must be minimized the function of the least squares with respect to K, K_0, K_1:

$$F\left(K, K_0, K_1\right) = \sum_{j=1}^{10}\left[C_{11}^{(1)}\left(T_j, 1\right) - \overline{C}_{11\text{exp}}^{(1j)}\right]^2. \tag{10}$$

6. Conclusion

The proposed utility model [14] uses an absorption-adsorption column and gives the possibility to create a waste-free technology for waste gases purification from sulfur dioxide by means of regenerable adsorbent, where the absorbent regeneration system is similar to the regeneration system in the patent [12] [13]. The efficiency of the processes is increased in an absorption-adsorption apparatus, where the absorption is realized in co-current flows and the adsorption takes place in the flexible adsorbent. That's why a new absorption-adsorption column apparatus with bubbling plates is offered. A mathematical model of the absorption-adsorption process in a plate column is presented too.

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