Experimental Study and Mathematical Modeling of SOx Removal from Flue Gas in Coal Combustion using Alkaline Scrubbing System

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ABSTRACT

In this paper, absorption of SO2 emitted from coal combustion into water is taken as a reference. The relation between concentration of alkali slurries and removal of SO2 is presented with graphs. The SO2 removal efficiency for the slurry of sodium carbonate is greater than lime /limestone. It is observed from the experiments that sodium alkali sorbents have high reactivity relative to the lime and limestone. From the experimental data of n+1 data points, an n degree polynomial using Lagrange interpolation method is formulated and verified the experimental results and also found the maximum value of the removal of Sox with an optimal flow rate.

Keywords: *Coal lumps, slurry of limestone, lime and sodium carbonate, Lagrange interpolation, optimization*

1. INTRODUCTION

Coal is a complex mixture of organic and inorganic matters which has been formed over years from continual layers of fallen vegetation. Coals are ranked from lignite to anthracite with respect to their gradual variation in the natural metamorphosis [1, 2]. Anthracite, bituminous, sub bituminous and lignite are the major ranks of coal. Coal rank is determined on the basis of heating value, fixed carbon, volatile matter, moisture and oxygen content. Coal rank cannot be defined by a single parameter instead it depends upon the number of constraints. Coal rank rises with the increase of fixed carbon and decrease in the volatile matter and moisture content. Carbon, hydrogen and oxygen are the major components of coal with small amounts of nitrogen, sulfur, moisture and minerals[3].

Small amounts of coal are used for transportation, space heating, firing of ceramic products, etc. Combustible gases and chemical intermediates are also produced by the gasification of coal whereas different carbon products are produced by various heat treatments. The fraction of coal is used in miscellaneous applications such as fillers, pigments, foundry material and water filtration [4-6]. Coal has long been used to provide power, heat and light. It is used to generate energy, warm homes and besides being used as a fuel for cooking and found as the cheapest fuel in railway engines. In many industrialized countries, coal has been replaced by natural gas and other clean fuels as a source to produce liquid fuels and generate power.

2. EMISSIONS

Emissions from coal combustion depend on the rank and composition of the fuel, the type and size of the boiler, firing conditions, load, type of control technologies, and the level of equipment maintenance.

The major pollutants of concern from coal combustion are particulate matter (PM), sulfur oxides (SOx), and nitrogen oxides (NOx). Some unburned combustibles including carbon monoxide (CO) and numerous organic compounds are generally emitted even under proper boiler operating conditions and the same idea was used by [1] in different context as well as in studies [7, 8].

In addition to SOx and NOx emissions, combustion of coal also results in the emissions of chlorine and fluorine primarily in the form of hydrogen chloride (HCl) and hydrogen fluoride (HF)[9-12]. Lesser amounts of chlorine gas and fluorine gas are also emitted. A portion of the chlorine and fluorine in the fuel may be absorbed into fly ash or bottom ash. Both HC1 and HF are water soluble and are readily controlled by acid gas scrubbing systems.

2.1 Removal of Sulphur Dioxide

Wet scrubbing/FGD can be a physical absorption of SO2 or a chemical reaction. The absorbent should have a large capacity for absorbing SO2 at a fast rate so as to reduce the size of the equipment. Besides, it should be possible to regenerate the absorbent to make the process viable in practice, SO2 is only slightly soluble in water so its use as an absorbent is ruled out. Even then a process has been patented in Japan where the concentration of SO2 in the exit gases can be reduced to about 100 ppm by scrubbing with water (1.9 liter of water/m3 of gases)[13-16]. Low-pressure steam, normally available in the plant can be employed for stripping of SO2. In actual practice, a chemical reaction is incorporated since mass transfer with chemical reaction enhances the rate of absorption.

Sulphurdioxide being acidic in nature is readily absorbed by alkaline solutions[17, 18]. There are a number of such scrubbing solutions which are;

I. Lime/limestone solution/slurries
II. MgO slurries/Mg(OH)2 solution
III. NaOH solution
IV. Na2SO3-NaHSO3 solution
V. Na2CO3 solution
VI. NH3-liquor solution
VII. Dimethylaniline solvent
VIII. Xylidine-water system
IX. Ammonium Sulphite solution

2.2 Environmental Impact of Sox Emissions

Sulfur dioxide belongs to the family of SOx gases. These gases are formed when fuel-containing sulfur (mainly coal and oil) is burned at power plants and during metal smelting and other industrial processes. Fuel combustion largely from electricity generation accounts for most of the total SO2 emissions. These gases dissolve easily in water. Sulfur is prevalent in all raw materials including crude oil, coal and ore that contain common metals like aluminum, copper, zinc, lead, and iron. SOx gases are formed when fuel containing sulfur, such as coal and oil is burned and when gasoline is extracted from oil or metals is extracted from ore. SO2 dissolves in water vapor to form acid and interacts with other gases and particles in the air to form sulfates and other products that can be harmful to people and their environment[19, 20].

In 1990, the consumption of coal by various sectors such as utilities, industrial, commercial and residential were about 860 million tons per year [21]. The amounts of SO2 discharged to the atmosphere are over 65% from utilities only using coal as fuel. Other sources of SO2 include industrial services such as petroleum refineries, metal processing industry and cement plants which consume coal or oil to yield process heat. Similarly, engines, large ships and diesel equipment using high sulfur fuels give SO2 emissions in the large amount to the air [18, 19].

3. EXPERIMENTAL

Lakhra coal is used for the combustion to study the removal of SO2 emissions. Proximate and ultimate analysis is given in table 1.

Table 1: Analysis of Lakhra Coal

Coal	Proximate analysis			Elemental analysis				Heating Value
	VM	FC	Ash	C	H	N	S	HHV (MJ/kg)
(%, db)	46.10	39.40	14.5	69.50	3.50	1.02	6.20	24.94

First of all coal lumps are crushed in the jaw crusher and then ground into fine product in laboratory Ball Mill. Similarly the limestone, lime and sodium carbonate are ground in same Mill. After grinding, these samples are screened with different sieves using sieve shaker and obtained fine powder passing through mesh no.200. Slurry of limestone, lime and sodium carbonate in different concentrations are prepared. Figure 1 shows the schematic diagram of experimental system.

In a fixed bed furnace, coal is combusted at a constant flow rate and air is provided at a flow rate of 92 cubic feet per min. Due to high sulphur in coal, the produced flue gases are rich in SO2. Flue gases are passed through a heat exchanger in order to reduce its temperature. Absorption of SO2 with water is considered as reference.

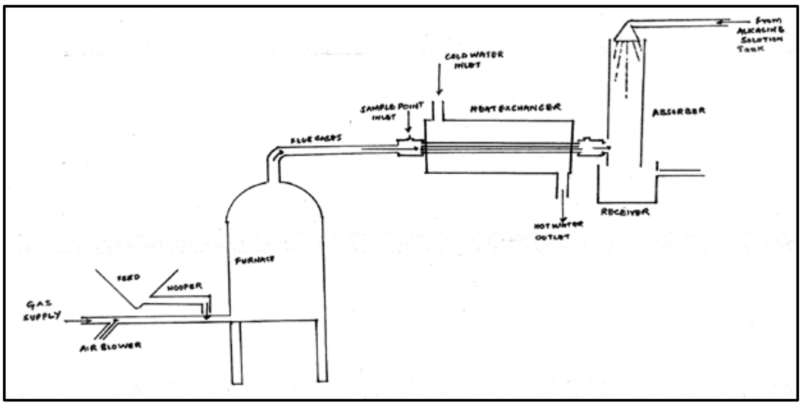

Fig 1: Schematic of Experimental System

After cooling, the flue gas is passed from the bottom of a scrubber and the slurry is showered from the top of scrubber. The SO2is absorbed into the scrubbing solution which is collected into the receiver while clean gas is vented to the atmosphere. The SO2 emitted from coal combustion is continuously monitored at the inlet and outlet of the scrubber by Flue Gas Analyzer. This SO2 removal method is based on the reaction of SO2 with different absorbents (CaCO3,Ca(OH)2, Na2CO3) to form calcium sulfite, CaSO3 which is then oxidized with excess oxygen both in the flue gas and water streams to form the final product CaSO4 as;

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$CaCO_3 + SO_2 + 1/2\ O_2 \rightarrow CaSO_4 + CO_2$
$Ca(OH)_2\ (aq) + SO_2\ (g) + ½\ O_2 \rightarrow CaSO_4 + H_2O$
$Na_2CO_3 + SO_2 + ½\ O_2 \rightarrow Na_2SO_4 + CO_2$

Temperature in the furnace is maintained at 750 – 850°C. Coal feed size and absorbent particle size is 200 mesh whereas the amount of oxygen in flue gases is maintained at 12%.

4. RESULTS AND DISCUSSIONS

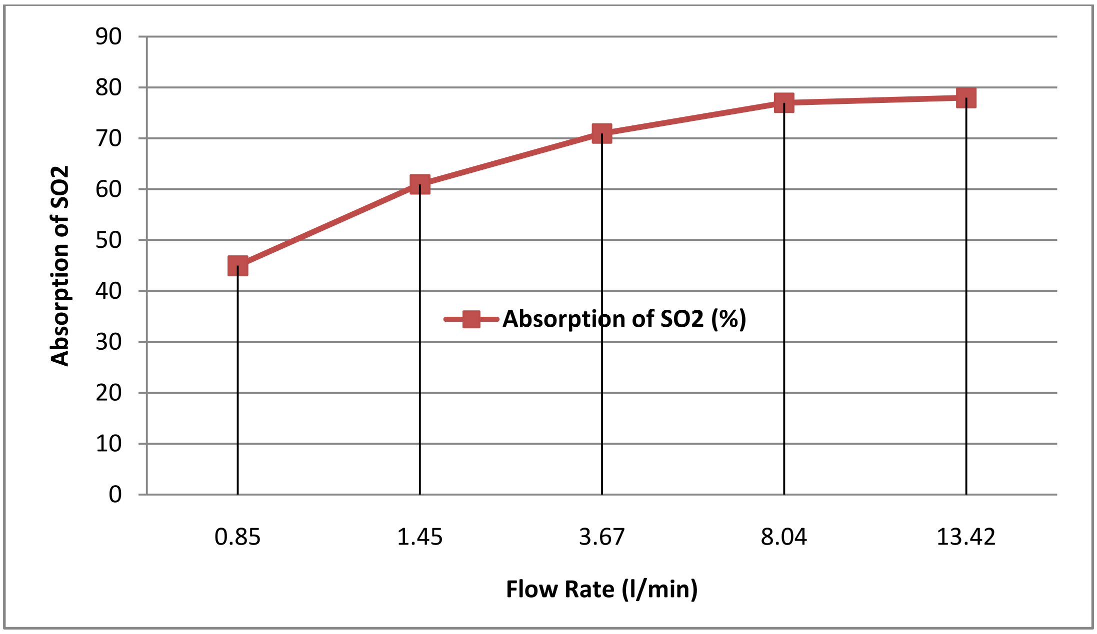

Fig 2: %age absorption of SO2 in water at different flow rates

Table 2: Comparative study of removal of SO2 with different slurries

Sr. #	Dose (%) (x_i)	Removal of SO_2 Using		
		CaCO3 Slurry (%) (y_i) (I)	CaO Slurry (%) (y_i) (II)	Na2CO3 Slurry (%) (y_i) (III)
1	2	42	48	55
2	4	57	62	70
3	6	72	76	82
4	8	81	84	89
5	10	83	86	92
6	12	84	88	93

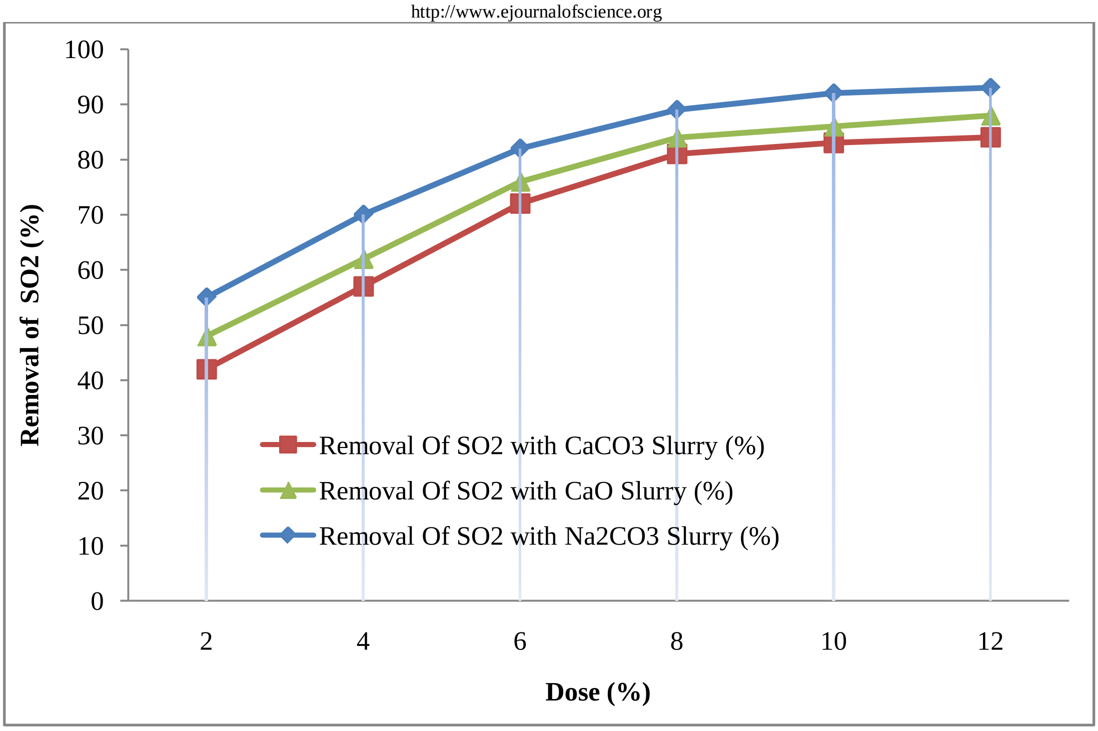

Fig 3: Removal of SO2 with different absorbents at various doses

5. MATHEMATICAL MODELINGUSING LAGRANGE INTERPOLATION POLYNOMIAL TECHNIQUE

In the mathematical field of Numerical analysis, interpolation is a method of constructing new data points from a discrete set of known data points. In engineering and science, one often has a number of data points as obtained by sampling or some experiment and tries to construct a function which closely fits those data points. This is called "Curve Fitting". Interpolation is a specific case of curve fitting in which the function must go exactly through the data points. A different problem which is closely related to interpolation is the approximation of a complicated function by a simple/polynomial function. There are many methods to interpolate/ extrapolate the data sets and here we use Lagrange Interpolating Polynomials" our experimental data is unequal spaced.

Lagrange Interpolating Polynomial is the polynomial $P(x)$of degree $n-1$ that passes through the $n$points $(x_1,y_1),(x_2,y_2),(x_3,y_3),\cdots,(x_{n-1},y_{n-1}),(x_n,y_n)$ is given by; $P(x)=\sum_{j=1}^{n=6} P_j(x)y_j$,

where

$$P_1(x)=\frac{(x-x_2)(x-x_3)\cdots(x-x_{n-1})(x-x_n)}{(x_1-x_2)(x_1-x_3)\cdots(x_1-x_{n-1})(x_1-x_n)}$$

$$P_2(x)=\frac{(x-x_1)(x-x_3)\cdots(x-x_{n-1})(x-x_n)}{(x_2-x_1)(x_2-x_3)\cdots(x_2-x_{n-1})(x_2-x_n)}$$

$$P_3(x)=\frac{(x-x_1)(x-x_2)\cdots(x-x_{n-1})(x-x_n)}{(x_3-x_1)(x_3-x_2)\cdots(x_3-x_{n-1})(x_3-x_n)}$$

$\ldots$

$$P_n(x)=\frac{(x-x_1)(x-x_2)\cdots(x-x_{n-2})(x-x_{n-1})}{(x_n-x_1)(x_n-x_2)\cdots(x_n-x_{n-2})(x_n-x_{n-1})}$$

The formula was first published by Warring (1779), rediscovered by Euler in 1783 and published by Lagrange in 1795. Now, we apply this Lagrange interpolation to our results obtained by experiments.

5.1 Numerical Results

Consider the data sets in table-2 with six data points and for these data points we have five degree polynomial using Lagrange interpolation polynomials.

Let x represents Dose (%) and y represents removal of SOx (%) then using data set-I for removing SO2 with CaCO3 Slurry (%) we get,

$$P(x)=0.00052x^5-0.0026x^4-0.2084x^3+2.38542x^2-0.925x+36.0 \quad (1)$$

using data set-II for removing SO2 with CaO Slurry (%) we get,

$$P(x)=-0.00130x^5+0.0572917x^4-0.9322917x^3+6.39583x^2-11.5x+52 \quad (2)$$

using data set-III for removing SO2 with Na2CO3 Slurry (%) we get,

$$P(x)=-0.000521\,x^5+0.0234\,x^4-0.375\,x^3+2.1563\,x^2+2.5083\,x+44 \quad (3)$$

For determining its optimum or maximum value, we follow the first and second derivative test.

In order to find the critical points, we put first derivative of the five degree polynomial equal to zero and find the real valued critical points and then at these critical points if second derivative is positive then that critical point gives maximum value. The results of these three data sets are given in the following table 3.

Table 3: Comparative study of removal of SO2 with different slurries

Assumed Dose (%) (x_i)	Removal of SO_2 Using Lagrange Interpolation Polynomial		
	CaCO3 Slurry (%)	CaO Slurry (%)	Na2CO3 Slurry (%)
1	37.25	46.019531	48.3125
3	48.98437	54.214844	62.57813
5	64.96875	69.597656	76.59375
7	77.45313	80.792969	86.10938
9	82.6875	85.980456	90.875
9.830487	83.00826	86.930481	91.850237
11	82.92188	87.720919	92.64063
12	84	88.1296	93.000001
12.55036	85.84543	88.223829	93.112983
13	88.40625	88.168681	93.156250
13.12104	89.29485	88.124303	93.15852
13.5	92.72241	87.875812	93.133057

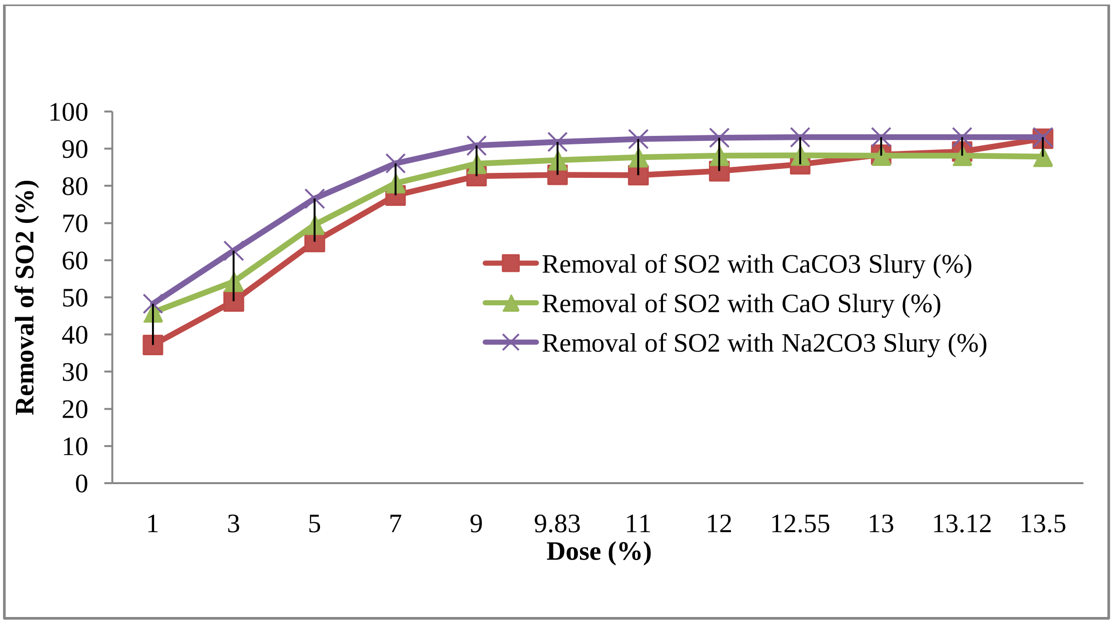

Fig 4: Removal of SO2 using Mathematical Model

Table 4: Optimum value of removal of SO2 using Lagrange Interpolation Polynomial

Critical Value for Doze (%) (x_i)		Optimum Value of Removal of SO2 Using Lagrange Interpolation Polynomial (y_i)
CaCO3 Slurry (%)	9.8304868	83.00826
CaO Slurry (%)	12.550355	88.223829
Na2CO3 Slurry (%)	13.12104	93.15852

6. CONCLUSION

Following conclusions are extracted from the experimental and mathematical results:

a) Absorption of SO_2 emitted from coal combustion into water is taken as a reference. The trend of graph shows that absorption of SO_2 increases with increase of flow rate of water and becomes almost constant at a point.
b) Absorption of SO_2 into water cannot be used for the removal of SO_2 because it yields sulfurous acid (H_2SO_3) which is a COD source; on oxidation it gives sulfuric acid (H_2SO_4) which is another pollutant.
c) The relation between concentration of alkali slurries and removal of SO_2 is developed as shown in the graphs.
d) At a constant L/G ratio, SO_2 removal efficiency is greater for sodium carbonate slurry than lime and limestone slurries.
e) Sodium alkali sorbents have high reactivity relative to lime and limestone sorbents. The scrubbing liquid is a solution than slurry because of high solubility of sodium salts. That's why the SO_2 removal efficiency is higher for sodium solution. The sodium scrubbers are generally limited to smaller sources because of high reagent cost.
f) Limestone scrubber is most suitable for the removal of SO_2 because of the availability and cost of the reagent.
g) The SO_2 removal efficiency also depends upon the concentration of the slurries, the particle size of the sorbents. The final particles have better sulfur retention as compared to the coarse particles.
h) This system is simple in construction and requires less initial cost as compared to the other conventional systems.
i) Mathematical modeling using Lagrange Interpolation method verifies the experimental results and give the optimum value of removal of SOx at a given flow rate.
j) The final waste from the system is harmless slurry of calcium sulphate and can be discharged without the risk of secondary pollution instead it is a fertilizer.

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