



## REMOVAL OF CARBON MONOXIDE FROM FLUE GAS

**Anjali Puranik**

Sheth J. N. Paliwala Commerce  
Science and Arts College  
Pali, Dist- Raigad  
Maharashtra, India

**Salil Puranik**

Sheth J. N. Paliwala Commerce  
Science and Arts College  
Pali, Dist- Raigad  
Maharashtra, India

### Abstract

Carbon monoxide gas is poisonous for human beings. It is emitted in atmosphere as one of the components of flue gas. The concentration of carbon monoxide in flue gas varies in the range of 10-10,000 ppm. The legislative limiting value for emission is 50 ppm. Its life in atmosphere is 0.3-0.7 years. It combines with other gases in atmosphere and generates more noxious gases. Like another flue gas components, it should be removed at source before releasing the furnace exhaust into atmosphere. The proposed work puts forth a post-combustion adsorption method by which carbon monoxide can be completely removed from flue gas. In a fixed bed adsorber, a dry sorbent is placed on mesh through which the flue gas diffuses and losses carbon monoxide. Both, adsorbent and the product are ecofriendly.

**Key words:** carbon monoxide, flue gas, adsorption, post combustion

### Introduction:

Carbon monoxide is an inevitable component of flue gas. It is left untreated in almost every flue gas treatment plant. Appearance of carbon monoxide in flue gas is claimed as the resultant of inefficiency of furnace and inadequate supply of oxygen. However, it is proved that exhaust of an efficient furnace contains a sizable amount of carbon monoxide<sup>1</sup> and even with supply of excess air/oxygen the concentration of carbon monoxide in flue gas is considerable<sup>2</sup>.

A flue gas treatment plant combines a variety of post combustion the removal methods<sup>3</sup> for SO<sub>x</sub>, NO<sub>x</sub> and CO<sub>2</sub>. From 1980's acid precipitation act<sup>4</sup>, the emission of acidic gases namely SO<sub>x</sub> and NO<sub>x</sub> has taken place substantially<sup>5</sup>. Carbon capture<sup>6</sup> (CC) and carbon capture and storage<sup>7</sup> (CCS) are getting adopted as policies by all nations in response to Copenhagen accord<sup>8</sup> and Parris agreement<sup>9</sup>. The agreement has set the target of reduction in emission called as Nationally defined contribution (NDC). In this scenario removal of carbon monoxide and a method for the same is rarely observed.

Gas separation methods pertaining to carbon monoxide are summarized further: Activated carbon adsorbs carbon monoxide. When 1000 ppm of CO was fed into the pure and impregnated sorbent, it was reduced to 40.2 and 10.4 ppm



respectively<sup>10</sup>. Impregnation is done by 34.7%  $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$  heated to produce AC-  $\text{SnO}_2$ . Another Tin- activated carbon<sup>11</sup> used for recovery of CO could recover 92.1 to 99% CO with purity 57-77%.

Govind Sethia et al<sup>12</sup> carried-out adsorption of CO on zeolite -X exchanged with magnesium, calcium strontium, and barium using volumetric gas adsorption method. Strontium exchanged zeolite showed maximum adsorption capacity of 28.4 molecules of CO per unit cell. The zeolite molecular sieve synthesized commercially<sup>13</sup> for the purpose have modified framework of  $\text{SiO}_2/\text{Al}_2\text{O}_3$  with 20-100 molar ratio and contain  $\text{Cu}^+$  ions to enhance the adsorptive capacity of zeolite for CO. The atomic ratio of Cu to Al is 0.49. A simulated blast furnace vent was composed as

CO: 27.5 v/V %,  $\text{CO}_2$ : 11.5 v/V%,  $\text{N}_2$ : 60 v/V %,  $\text{H}_2$ : 1 v/V %

Saturated with water vapor at 1atm and 50<sup>0</sup>C. Almost 100% CO was adsorbed by the zeolite.

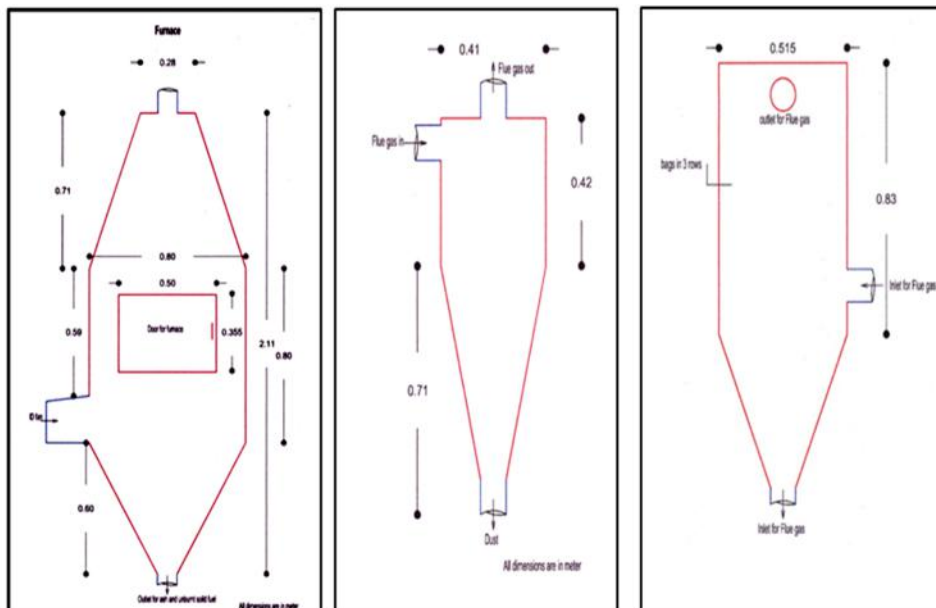
G. Spoto et al<sup>14</sup> doped H- ZMS by equivalently exchanging monovalent copper. The  $\text{Cu}^+$  ions are highly coordinately unsaturated and form  $\text{Cu}^+(\text{CO})_n$  complexes where (n = 1,2 or 3). Xie et al<sup>15</sup> treated a variety of zeolites including 5A, zeolite -X, zeolite-Y with cuprous chloride and bromide at different concentrations, temperatures, atmospheres and heating hours. They showed a removal capacity of 1.8 mmol to 3.2 mmol of CO per gm of zeolite. In all there are 48 adsorbents which include alumina and silica as a solid support for  $\text{Cu}^+$  ions. The adsorption of water on zeolite is very strong. Sircar and co-workers<sup>16</sup> reported that this limits the use of zeolites for removal of CO from flue gas since it contains moisture.

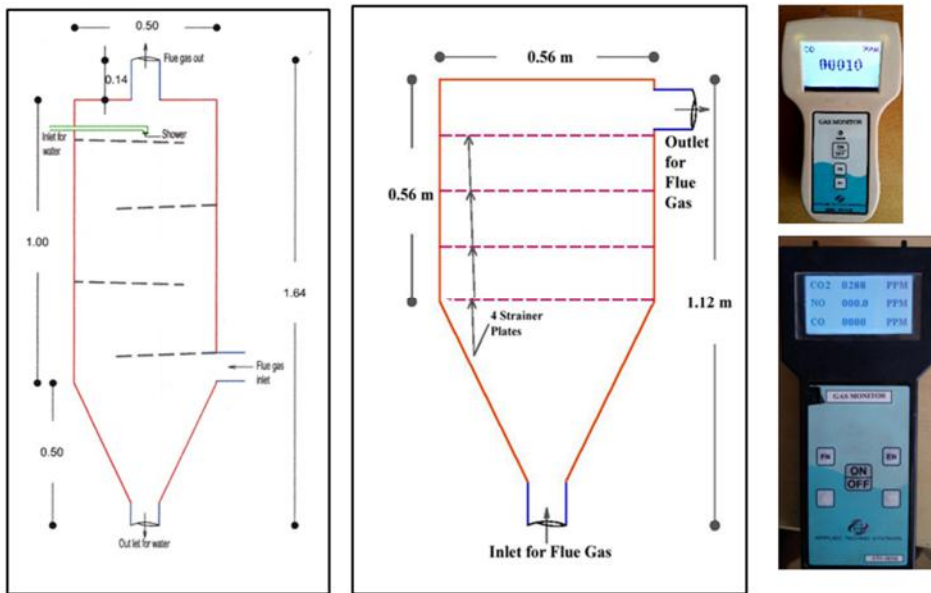
In older methods<sup>17</sup> CO was absorbed in acidic solution of CuCl or ammonical solution of  $\text{Cu}_2\text{CO}_3$  or Cu- formate. At room temperature and 200 atm pressure. Recovery of CO was done by releasing the pressure and heating the solution to 152<sup>0</sup>C. Willum group and Ilse<sup>18</sup> found a method for separation of water gas into carbon monoxide and hydrogen. After variety of trial with cuprous salts, organic acids and phenols they found cuprous ammonium lactate has most favorable properties of adsorbent. It is under trial in semi plant scale. Gardner C-Ray and Paul H Jonson<sup>19</sup> found an improved solvent for CuCl. It is 'Orthoanisidine' which is capable of dissolving cuprous salt up to 20% by its weight. It selectively adsorbs CO in presence of other gases except  $\text{SO}_2$  and  $\text{H}_2\text{S}$  at 10-37<sup>0</sup>C. It needs anti -freezing agent like MeOH. Desorption is done by VSA or TSA. Cho et al<sup>20</sup> prepared a cuprous salt with glucose and ammonia which showed removal capacity of 1.0 mmol per gram of sorbent. Alumina was studied for adsorption of CO by Stanislaus et al<sup>21</sup>. An adsorbent of CuCl with alumina and silica-aluminium carriers was prepared by Tsuji et al<sup>22</sup>. They showed removal capacity of 0.5 to 1 mmol per gram of sorbent. Koichi et al<sup>23</sup> attempted composition of Cu/Zn/ $\text{Al}_2\text{O}_3$  catalyst prepared by impregnation method for removal of CO from

water gas shift reaction. Hirai et al<sup>24</sup> reached adsorption capacity of 31.5 mmol per gram of sorbent even in presence of water. They used combination of Cu(I) halide plus Aluminium (III) halide with either polystyrene or activated carbon/graphite. They could remove 1-99% v/V CO in presence of 40,000 ppm water. G.D. Buckley and N.H. Ray<sup>25</sup> found that CO reacts with hydrazine under high pressure to give variety of products depending on conditions employed. A Commercial catalyst<sup>26</sup> is used in air purifier converts CO to CO<sub>2</sub>. It is composed of manganese and copper oxide plus salts of sodium, potassium and calcium. Anand Patwardhan and Mohan Sharma<sup>27</sup> compared the kinetics of absorption of CO in aqueous solution of NaOH and Ca(OH)<sub>2</sub> slurries. They suggested a mechanism of reaction in which hydroxyl ion attacks on carbon of CO molecule by lone pair of electrons and 'formate ion' is formed. Michal Bails and Frank S. Stone<sup>28</sup> observed the reaction of CO with O<sup>2-</sup> ions on CaO and MgO. They found an enthalpy of formation of CO<sub>3</sub><sup>2-</sup> is 56% of theoretical value of CaCO<sub>3</sub> and 47% of theoretical value of MgO. This decrease is due to formation of chemisorbed complexes which include highly conjugated (CO)<sub>n</sub><sup>2-</sup> anions which impart yellow colour (455 nm) to the oxide.

### Experimental:

**Designing of the system:** Pipeline: 6 mm pipe, 3 inches diameter. Steel plates of all device 6 mm





**1. Furnace:** A real flue gas is generated in a steel furnace using peat coal. The size of the furnace is as shown in the figure. Coal pieces are placed in lower middle part on a grid. FD (forced draft) is provided to supply ambient air for combustion. The flue gas is released from the top of the furnace.

**2. Cyclone separator:**

Flue gas moves with tangential velocity producing cyclone of flue gas inside. Particles strike on the walls and slide down into bottom.

**3. Baghouse filter:** Pulse jet type baghouse filter is designed. Twelve bags covering maximum volume of baghouse. Every bag of size 8×3 inches housed on metallic cage. Material is commercially available. It is a felted type biodegradable material.

**4. Water scrubber:**

Counter current flow type water scrubber is constructed. Three showers fitted at the top. Four inclined plates built a plane of water alternately. Flue gas entering from bottomside upwards, passes through planes and shower of water of water and leaves from the top.

**5. ID fan:** The velocity of gas is further increased by ID (Induced draft) fan.



6. **The adsorber:** In a cubical box tapered at the bottom, four steel nets are fitted. A cotton cloth is placed on each strainer. Dry sorbent is spread on each cloth manually. Flue gas enters from the bottom and leaves from the top.
7. **Sampling points:** To the pipe at entrance and exit of adsorber three identical thin pipes are welded. These pipes are connected to gas detector.
8. **The control panel:** A panelcontrolling speed of ID and FD fans, water pump to scrubber, RH meter, temperature measuring thermister in each device is installed.
9. **RH meter + Temp. Thermister:** Relative humidity and temperature of the entering gas is measured near the sampling points of the adsorber.
10. **Gas Analyzers:**
  - 1.ATS 103M in range 0-200 ppm level and 2.ATS 101M CO in range 0-10,000 ppm

### Working:

#### Firing of the furnace:

Keep all the valves closed. Keep door of the furnace open. Place 3-4 kg coal and 400-500 gm briquette as fuel on the grid. Ignite the fuel switch on the FD fan and slowly rise its speed. Wait till the smoke gets reduced and coal is red hot. Close the door of furnace, switch off FD fan and open all the valves of pipeline. Fit the RH meter and gas analyzers to their points. Wait for 2 minutes and note down the steady values of concentration of 'in' and 'out' for every gas. Note the observation after every four minutes. The graph of concentration in ppm with time is plotted in the same graph paper.

The equation used to calculate the removal capacity is

$$\% \text{ Noxious gas removed} = \left( \frac{[\text{Noxious gas}]_{\text{in}} - [\text{Noxious gas}]_{\text{out}}}{[\text{Noxious gas}]_{\text{in}}} \right) \times 100$$

Another graph is plotted as % removal Vs Time in minutes.

**Table1:Basic Sorbent :A: 4 kg in each layer:**

Time minutes	CO in ppm		Time minutes	CO in ppm	
	IN	OUT		IN	OUT
0	1636	1258	32	1428	1188
4	1604	1234	36	1383	1175
8	1591	1248	40	1353	1156
12	1537	1228	44	1307	1151
16	1480	1211	48	1293	1139



20	1470	1211	52	1282	1131
24	1452	1200	56	1222	1098
28	1447	1199	60	1197	1081

Graph:1: Removal by Slaked lime

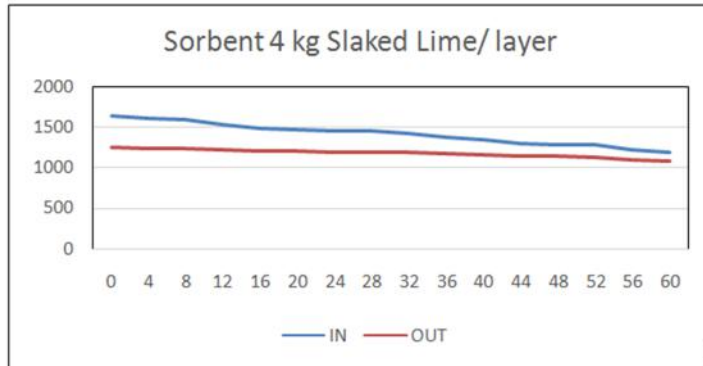
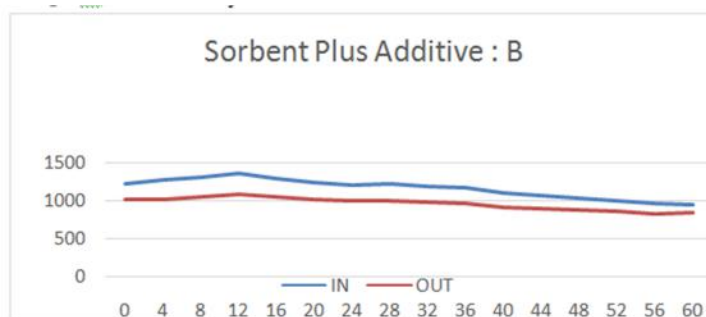


Table 2: Removal by 4 kg slaked lime plus 1kg salt :Sorbent Plus Additive Mixture B:

Time in minutes	CO in ppm		Time in minutes	CO in ppm	
	IN	OUT		IN	OUT
0	1235	1021	32	1198	986
4	1278	1032	36	1179	981
8	1307	1056	40	1103	921
12	1364	1096	44	1070	906
16	1303	1067	48	1036	889
20	1252	1023	52	1002	865
24	1217	1007	56	976	834
28	1236	1002	60	955	845

Graph:2 Removal by Mixture B

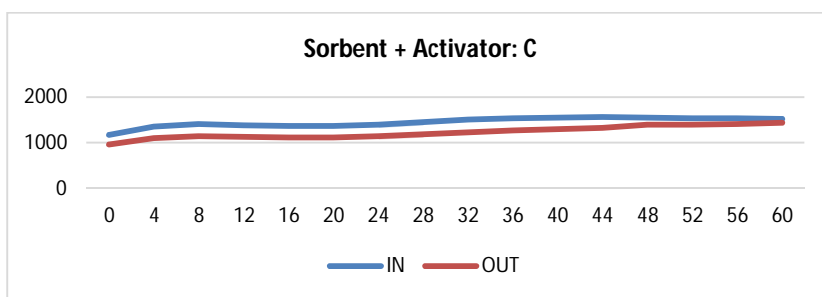




**Table 3: Removal by Sorbent + Additive+ Activator: Mixture C**

Time in minutes	CO in ppm		Time in minutes	CO in ppm	
	IN	OUT		IN	OUT
0	1165	959	32	1501	1226
4	1356	1103	36	1528	1263
8	1401	1135	40	1549	1289
12	1383	1121	44	1556	1324
16	1372	1113	48	1544	1387
20	1370	1111	52	1538	1399
24	1398	1143	56	1538	1412
28	1445	1183	60	1520	1437

**Graph 3: Removal by Sorbent + Additive+ Activator: Mixture C**

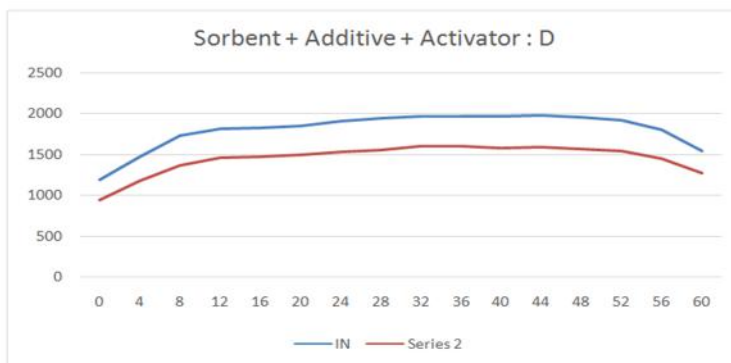


**Table 4 : Sorbent + Additive + Activator: Mixture D**

Time in minutes	CO in ppm		Time in minutes	CO in ppm	
	IN	OUT		IN	OUT
0	1198	951	32	1970	1599
4	1480	1187	36	1975	1594
8	1736	1372	40	1975	1574
12	1815	1465	44	1981	1585
16	1832	1469	48	1960	1567
20	1857	1493	52	1924	1541
24	1908	1526	56	1800	1453
28	1943	1551	60	1544	1278



**Graph 4: Sorbent + Additive + Activator: Mixture D**

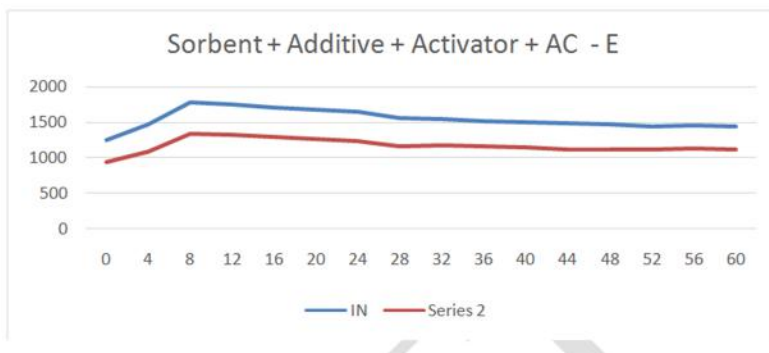


**Table 5: Sorbent + Additive + Activator + Active component (AC : 100 g) Mixture E**

Time minutes	CO in ppm		Time minutes	CO in ppm	
	IN	OUT		IN	OUT
0	1250	934	32	1553	1172
4	1466	1089	36	1512	1155
8	1783	1341	40	1502	1152
12	1755	1323	44	1486	1121
16	1713	1301	48	1470	1109
20	1677	1272	52	1438	1119
24	1651	1243	56	1456	1135
28	1562	1166	60	1437	1119

**Graph: 5: Sorbent + Additive + Activator + Active component (AC : 100 g) Mixture E**

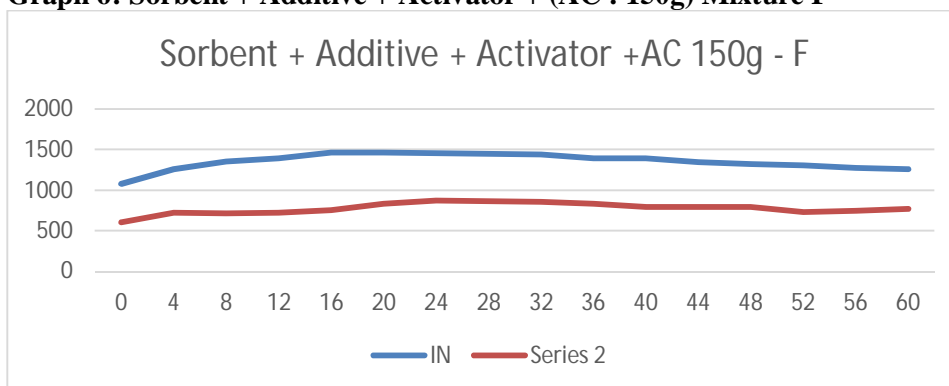




**Table 6: Sorbent + Additive + Activator + (AC : 150g) Mixture F**

Time minutes	in	CO in ppm		Time minutes	in	CO in ppm	
		IN	OUT			IN	OUT
0		1076	603	32		1443	855
4		1260	722	36		1396	835
8		1350	718	40		1389	791
12		1396	721	44		1344	795
16		1460	756	48		1325	790
20		1462	834	52		1305	733
24		1456	869	56		1275	745
28		1445	865	60		1261	770

**Graph 6: Sorbent + Additive + Activator + (AC : 150g) Mixture F**





**Table 7: Comparison of Adsorbents : A,B and C:**

Time in Minutes	Adsorbent			
	4 kg S. Lime	4 kg S. Lime + 1 kg Common Salt	4 kg S. Lime + 0.1 kg Bleaching Powder	4 kg S. Lime + 1 kg Common Salt + 0.1 kg Bleaching Powder
0	23.11	17.33	17.68	20.62
4	23.07	19.25	18.66	19.80
8	21.56	19.20	18.99	20.97
12	20.10	19.65	18.94	19.28
16	18.18	18.11	18.88	19.81
20	17.62	18.29	18.91	19.60
24	17.36	17.26	18.24	20.02
28	17.14	18.93	18.13	20.17
32	16.81	17.70	18.32	18.83
36	15.04	16.79	17.34	19.29
40	14.56	16.50	16.79	20.30
44	11.94	15.33	14.91	19.99
48	11.91	14.19	10.17	20.05
52	11.78	13.67	9.04	19.91
56	10.15	14.55	8.19	19.28
60	9.69	11.52	5.46	17.23
64	8.12	11.37	3.80	17.70
68	7.54	11.32	3.78	15.77
72	8.99	9.16	3.33	15.68
76	8.64	8.44	4.24	15.28
80	7.64	6.41	4.80	13.16
84	7.57	4.99	2.01	11.79



Graph 7: Comparison of A,B and C:

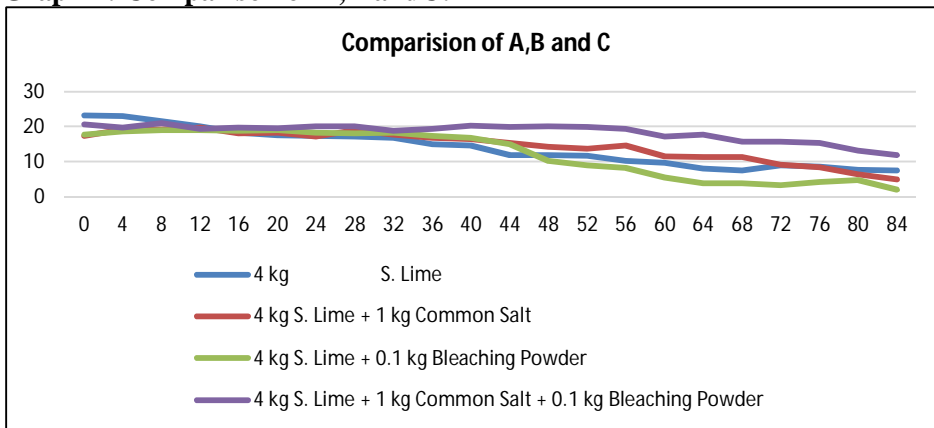


Table8: Comparison of D, E, F :

Time in Minutes	Adsorber			Time in Minutes	Adsorber		
	D	E	F		D	E	F
0	20.62	25.28	43.96	72	15.68	21.18	37.34
4	19.80	25.72	42.70	76	15.28	22.34	38.77
8	20.97	24.79	46.81	80	13.16	21.53	39.09
12	19.28	24.62	48.35	84	11.79	23.59	35.68
16	19.81	24.05	48.22	88	10.22	23.45	35.76
20	19.60	24.15	42.95	92	9.92	22.21	35.52
24	20.02	24.71	40.32	96	12.48	22.15	37.37
28	20.17	25.35	40.14	100	12.92	23.77	35.36
32	18.83	24.53	40.75	104	13.80	23.86	37.37
36	19.29	23.61	40.19	108	13.17	23.99	25.78
40	20.30	23.30	43.05	112	8.17	23.81	22.80
44	19.99	24.56	40.85	116	7.86	23.56	22.26
48	20.05	24.56	40.38	120	7.49	23.49	25.12
52	19.91	22.18	43.83	124	7.47	23.83	29.01
56	19.28	22.05	41.57	128	4.32	18.91	27.90
60	17.23	22.13	38.94	132	3.83	15.34	23.84
64	17.70	22.76	36.59	136	1.82	13.71	28.10
68	15.77	22.27	35.10	140	1.04	16.19	25.35



Graph 8: Comparison of Adsorbents D, E, F :

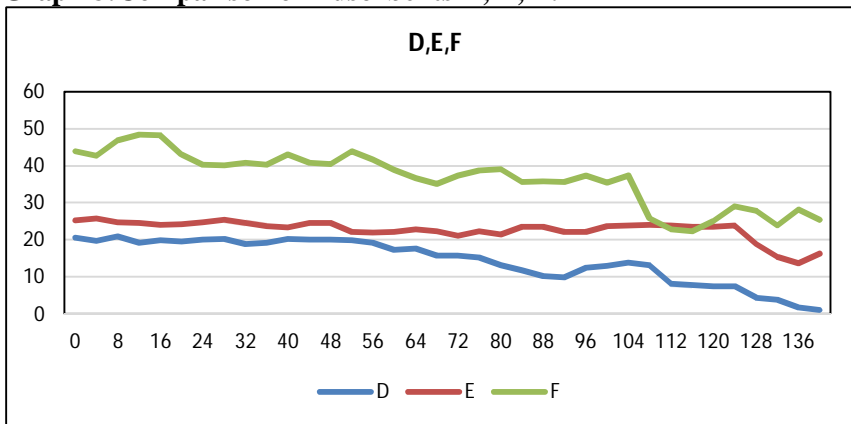
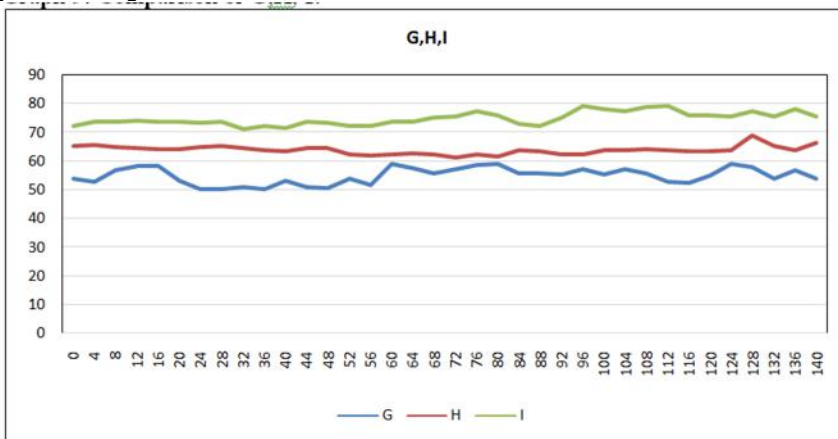


Table 9 : Comparison of G, H, I:

Time in Minute s	Adsorber			Time in Minutes	Adsorber		
	G	H	I		G	H	I
0	53.96	65.28	72.15	72	57.34	61.18	75.36
4	52.70	65.72	73.77	76	58.77	62.34	77.37
8	56.81	64.79	73.86	80	59.09	61.53	75.78
12	58.35	64.62	73.99	84	55.68	63.59	72.80
16	58.22	64.05	73.81	88	55.76	63.45	72.26
20	52.95	64.15	73.56	92	55.52	62.21	75.12
24	50.32	64.71	73.49	96	57.37	62.15	79.01
28	50.14	65.35	73.83	100	55.36	63.77	77.90
32	50.75	64.53	71.18	104	57.37	63.86	77.34
36	50.19	63.61	72.34	108	55.78	63.99	78.77
40	53.05	63.30	71.53	112	52.80	63.81	79.09
44	50.85	64.56	73.59	116	52.26	63.56	75.68
48	50.38	64.56	73.45	120	55.12	63.49	75.76
52	53.83	62.18	72.21	124	59.01	63.83	75.52
56	51.57	62.05	72.15	128	57.90	68.91	77.37
60	58.94	62.13	73.77	132	53.84	65.34	75.36
64	57.70	62.76	73.86	136	56.82	63.71	78.10
68	55.77	62.27	75.10	140	54.04	66.19	75.35



Graph 9: Comparison of G,H, I:



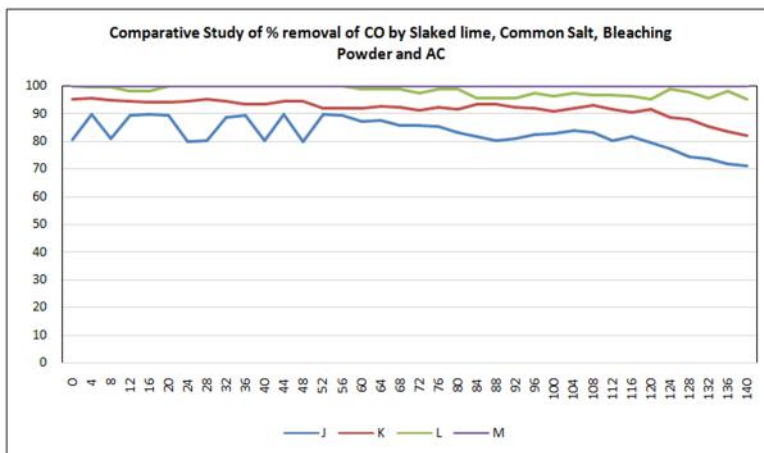
The further mixtures are prepared by adding 50 grams of active component to observe increase in percentage removal.

Table 10: Increment of AC 50 gm each

Time in minutes	Adsorbent				Time in minutes	Adsorbent			
	J	K	L	M		J	K	L	M
0	80.62	95.28	99.96	99.97	72	85.68	91.18	97.34	100
4	89.8	95.72	99.8	100	76	85.28	92.34	98.77	100
8	80.97	94.79	99.81	100	80	83.16	91.53	99.09	100
12	89.28	94.62	98.35	100	84	81.79	93.59	95.68	100
16	89.81	94.05	98.22	100	88	80.22	93.45	95.76	100
20	89.6	94.15	100	100	92	80.92	92.21	95.52	100
24	80.02	94.71	100	100	96	82.48	92.15	97.37	100
28	80.17	95.35	100	100	100	82.92	90.77	96.36	100
32	88.83	94.53	100	100	104	83.8	91.86	97.37	100
36	89.29	93.61	100	100	108	83.17	92.99	96.78	100
40	80.3	93.3	100	100	112	80.17	91.81	96.8	100
44	89.99	94.56	100	100	116	81.86	90.56	96.26	100
48	80.05	94.56	100	100	120	79.49	91.49	95.12	100
52	89.91	92.18	99.9	100	124	77.47	88.83	99.01	100
56	89.28	92.05	99.9	100	128	74.32	87.91	97.9	100
60	87.23	92.13	98.8	100	132	73.83	85.34	95.84	100
64	87.7	92.76	99	100	136	71.82	83.71	98.1	100
68	85.77	92.27	98.9	100	140	71.04	82.19	95.35	100



Graph 10: Increment of AC 50 gm each

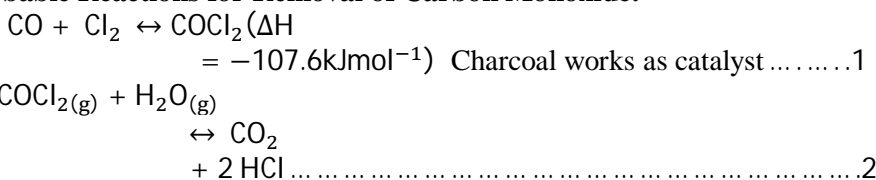


**Discussion:**

Removal of Carbon monoxide from flue gas is not attempted before using real flue gas. A few attempts are made by introducing artificial flue gas which selectively includes CO<sub>2</sub>, CO N<sub>2</sub>and moisture. Other reactive gases like NO<sub>x</sub>, SO<sub>x</sub>, volatile organic matter, particulate matter etc. are inherent components of flue gas and do affect the adsorbent. In fact, design an adsorbent to remove carbon monoxide including all these components is in state of art. Unlike NO<sub>x</sub>, SO<sub>x</sub> and CO<sub>2</sub>, which are reactive and acidic, carbon monoxide is inert. It has low dipole and quadrupole moment. Common adsorbents like activated charcoal, zeolites, earths do not adsorb carbon monoxide quantitatively since its desorption is equally faster. In presence of other components of flue gas the adsorption of carbon monoxide is negligible on these common adsorbents.

Considering the reactivity of carbon monoxide with typical compounds a basic sorbent is selected and its removal capacity is enhanced by certain additives, activators and increasing the proportion of active component in the mixture. A very small amount of free chlorine is added to mixture initially which acidifies then surface of the sorbent. The hydrated layer of sorbents allows the following reactions which result in significant adsorption of carbon monoxide.

**Probable Reactions for Removal of Carbon Monoxide:**





$\text{CO}_3^{2-} + \text{CO}_2 + \text{H}_2\text{O}$ → $\text{HCO}_3^-$ (In presence of salt and moisture) .....	3
$\text{CO} + \text{H}_2\text{O} \leftrightarrow \text{H}_2$ + $\text{CO}_2$ (water – gas shift reaction occurring in the gas phase) ...	4
$\text{CO} + \text{H}_2\text{O}$ ↔ $\text{HCOOH}$ ( Slow reaction) .....	5
$\text{CO} + \text{CaSO}_4 \leftrightarrow \text{CaO} + \text{SO}_2$ + $\text{CO}_2$ .....	6
$\text{CO} + \text{OH}^-$ ↔ $\text{HCOO}^-$ (The adsorption reaction) .....	7

### Conclusion

Reaction 1,5,6 and 7 are expected to be predominant in removal of CO. Free chlorine is made available in the sorbent which is less than 1%. Water is in form of moisture which is a constituent of flue gas.  $\text{CaSO}_4$  is formed by reaction of Sulfur oxides with slaked lime. Probability factor indicates that out of billions of physiosorbed molecules, only a few achieve activation energy and proper orientation to react with each other. To hold a gas molecule by the sorbent bed, physical adsorption is enough. Instead of measuring the chemical reactivity of sorbent towards the gas, the removal capacity is better measured as the ‘holding’ capacity of the sorbent for the specific gas. This aspect highlights the extraordinary performance by the sorbent. The exhaustive experimental work enlightens the need of removal of CO from the flue gas and a definite practicable solution with 100 % removal capacity.

### Suggestions:

Though the gas detectors are indicating removal of carbon monoxide to a sizable quantity, evidence/s to support the observations by surface analysis methods like SEM, XRD, EDX etc. should be attempted. It is also a task to find a chemical method/s which can estimate or at least indicate presence of carbon monoxide or its sorption products in the sorbent.



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