

The Fractionated Precipitation Process of Zinc and lead Compounds in Ammonical Ammonium Carbonate Solutions

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Abstract: Chemical and metallurgical industries produce each year millions of tons of waste products containing low grade oxidized Zinc and other materials which contain toxic and harmful elements depending on their sources. This paper attempts to develop a process to fractionate precipitation of Zinc and Lead compounds using solvents of ammonia and ammonium carbonate forming different levels of leach types to which Electric Arc Furnace Dust (EAF) is added. A total mass of suspension was composed of mass of distilled water, mass of ammonium carbonate, mass of ammonium hydroxide and mass of EAF dust making 26 mass suspensions to each of which several parameters were applied. The parameters included system temperatures, pH values, carbon dioxide flow rate, using a reactor where these suspensions were subjected to different speeds of the stirrer, different conductivity and different time. The research concluded by reaching an appropriate combination of the parameters which simultaneously achieved a break point that produced the desired optimum recovery of the heavy metal. This break point has the following specification: the optimum solution was 2746.9 (g) being composed of 39.14% distilled water, 19.5% Ammonium carbonate 16.33% ammonium hydroxide and 24.99 % EAF dust while the optimum level of carbon dioxide flow rate was 5 in/min, rotation speed was 700 rpm and temperature was 50 °C.

1 Introduction

Chemical and metallurgical industries produce each year millions of tons of waste products containing low grade oxidized zinc and other materials added to the already huge amounts accumulated over the years in the world. These waste products contain toxic and harmful materials depending on their sources. These sources include dry and wet galvanizing plants, Electric Arc Furnace dust (EAF), steel mills, smelting operations etc. However, keeping the environment clean from these toxic and harmful materials is becoming one of the hot issues facing these chemical and metallurgical industries. Local regional and international authorities and organizations who are very much concerned about the environment have set stringent environmental regulations and penalties in case of dumping these materials in landfill. The chemical and metallurgical industries being aware of these regulations and penalties have tried to develop treatments at various levels to recover some of the metals contained in these waste products and solve the environmental problem as well. These treatments used pyrometallurgical and hydrometallurgical processes. These processes have been investigated by many researchers and some of them have been tried commercially. However pyrometallurgical processes were said to be very costly both in terms of capital investment and energy (De. Juan, et al 1996) On the other hand hydrometallurgical processes were considered friendly, more efficient, environmentally safe and flexible (Jha, et al; 2001). Both types of processes use a range of lixiviants/extractants/solvents, some of which

were considered selective. They ranged from concentrated acids to concentrated alkalines. Not only that; they were also performed under varied temperatures and atmospheric pressures.

Several scientific papers and publications have been written on the subject. They dealt with the different treatments which were recognized in recovering zinc from industrial waste products. This paper is concerned with those papers and publications which dealt with hydrometallurgical methods. These hydrometallurgical processes used a range of lixiviants depending on the source of the waste products. Among these lixiviants were sulphuric acid, hydrochloric acid, ammoniacal solution and sodium hydroxide:

2 Objectives of the Research

The objective of the research is to develop a hydrometallurgical process to remove the heavy metals as well as the zinc oxides from EAF dust and use the remaining inert material. This objective will be achieved through fractionated precipitation process of zinc and lead compounds in ammoniacal ammonium carbonate solutions obtained from EAF dust.

3 Materials and Methods

The research will try to reach a break point by a process consisting of simultaneous mechanical activating grinding and leaching of zinc, lead and iron in an alkaline environment using ammoniacal ammonium carbonate system combined with a high selective fractionated precipitation downstream. The fractionated precipitation condition of the hot educt solutions will be investigated to determine the simultaneous effect of several parameters which include system temperature, concentration of reactants in solution, pH value, gas partial pressure of ammonia, carbon dioxide and inter media in order to get maximum particle recovery of alkaline zinc and lead hydroxide/carbonate.

The experimental work was conducted in the laboratory and included tests with sodium carbonate and precipitation with carbon dioxide for optimization.

In the experiments two approaches for selective fractionated precipitation of zinc carbonate (ZnCO_3) were employed. The first was a thermal approach (lead and zinc carbonate complexes precipitate from the educt solution of leaching at 40°C and 80 °C, recycling by heating up the solution and the other was a chemical approach. Five pretest experiments were performed. In the first experiment the thermal approach was used while in the other four the chemical approach was used. The chemical approach used sodium carbonate solution, sodium carbonate as solid, hydrochloric acid and carbon dioxide.

In this research the experiment which was adopted used solvents of ammonia and ammonium carbonate. It is worth noting that in this set of experiments a sample was prepared by leaching different quantities of distilled water, ammonium carbonate and ammonium hydroxide forming different levels of leach types to each of which EAF dust in a powder form was added. The mixture of these leach levels and EAF dust made a suspension which was then used in separate experiments. Twenty six mass suspensions were prepared (Table 1). Each of these suspensions was used in a separate experiment using a bath reactor. Fig (1).

With continuous stirring of the suspension in the reactor, pH, temperature and conductivity were measured every two minutes for a period of one hour. Carbon dioxide was continuously

Table 1 Experiment Results Compositions of Different Types of Leaches

Type of Leach	Mass %	Mass %	Mass %	Mass %	Total Mass	Total Mass of
	H2O	[(NH ₄) ₂ CO ₃]	[NH ₄ OH] 25%	EAF-Dust	of Leach [g]	Suspension [g]
V 6.1	52.62	19.07	8.91	19.37	1872.0	2322.0
V 7.1	51.60	19.07	9.84	19.48	3305.6	4105.6
V 8.1	51.72	18.92	10.10	19.24	3356.8	4156.8
V 10.1	52.07	19.56	8.53	19.83	3233.4	4033.4
L G.1	52.97	18.87	9.07	19.07	3393.9	4193.9
V 11.1	52.75	19.69	7.91	19.97	3206.0	4006.0
L G.2	52.52	19.71	7.77	19.98	3202.3	4002.3
V 12.1	52.47	19.72	7.8	19.99	3201.6	4001.6
V 13.1	52.95	19.4	7.65	19.98	4805.5	6005.5
V 14.1	52.85	19.47	7.67	19.98	5403.8	6753.8
V 15.1	51.30	15.30	13.20	20.2	5413.8	6785.5
V 17.1	52.23	17.71	10.14	19.9	5430.9	6780.9
V 18.1	49.90	12.42	17.71	19.96	4811.8	6011.8
V 19.1	62.80	12.10	10.10	15.00	5106.0	6006.0
V 20.1	39.16	19.50	16.33	24.99	3600.2	4800.2
L G.3	50.44	16.09	13.46	19.99	3600.5	4500.5

Source: The researcher.

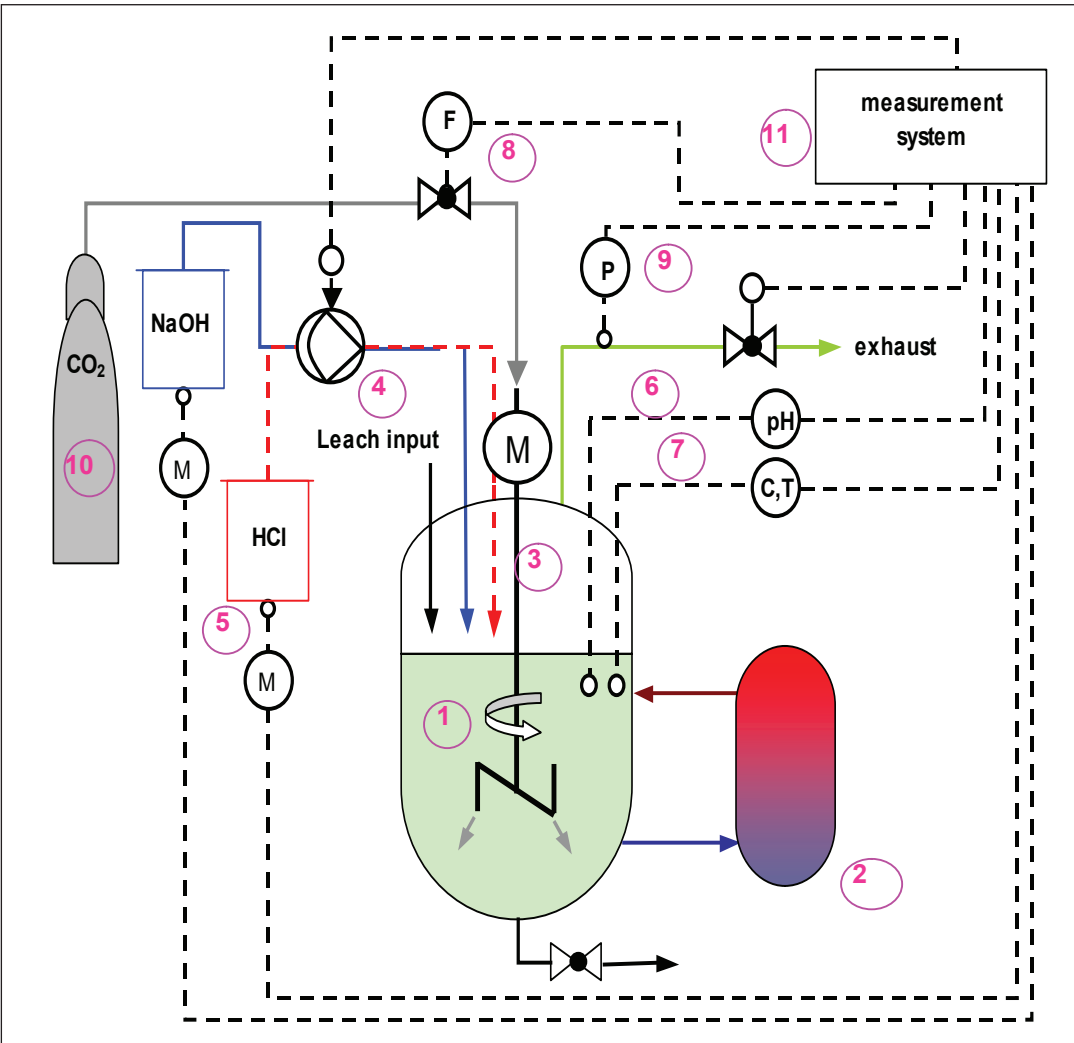
added into the reactor at a predetermined flow rate and the suspension was closely monitored for any changes. The time of the first precipitation was recorded together with the levels of the associated parameters. These parameters included the rotation speed of the stirrer and the initial temperature. At the end of the experiment the precipitate was filtered, dried and weighted for each type of leach (Table 2).

Results and Discussions

The results included the following percent precipitation from different leach samples, same leach composition with different set parameters (mass of solution, carbon dioxide flow in min, rotation speed rpm and temperature. The relationship of the type of leach to the different parameters and the resulting precipitation was recorded and plotted for each type of leach and it's associated parameters. The results also included pH versus time and conductivity versus time. At a later state a comparison between the different types of leach related to temperature carbon dioxide flow rate, rotation speed and precipitation percentage was made.

Higher and lower temperatures did not influence the selectively. In general a higher precipitation ratio did not mean a higher precipitation rate and the optimal temperature to get high precipitation rate or recovery of zinc and high amount of precipitation was about 15 °C. Increasing or decreasing the pH. Precipitation depended on the temperature. The effect of high amount of carbon dioxide flow rate (about 600 in/min) led to an earlier precipitation and an increase of the amount. Lower specific carbon dioxide input could decrease the recovery of zinc. The effect of a high ratio of ammonium carbonate led to increase the precipitation pH while a small ratio of

Fig. 1 Flow Diagram of the Precipitation Reactor System



Labels Description

1. Precipitation reactor.
2. Thermostat.
3. Stirrer drive with gassing stirrer.
4. Dosage pump.
5. Balance with receiver of NaOH / HCl.
6. Sensor pH.
7. Sensor C, T.
8. Mass flow controller CO₂.
9. Pressure controller.
10. Receiver CO₂.
11. Measurement data logging and analysis.

Table 2 Mass Precipitate

Type of Leaching	Mass of Solution [g]	Mass of Precipitate [g]
V 7.1	2167.4	151.4
V 18.1a	1874.7	155.3
V 11.1	2118.4	158.6
L G.2	2267.6	168.8
V 13.1c	1906.1	138.6
V 13.1b	2045.1	144.7
V 12.1	2219.5	154.9
V 18.1b	1972.2	132.9
L G.3	1843.2	116.6
V 20.1b	2415.2	151.2
V 19.1c	1992.1	124.3
V 20.1a	2746.9	170.6
V 19.1b	1739.6	100.8
L G.1	2212.5	112.7
V 20.1c	1867.1	84.7
V 10.1	2213.0	100.2
V 15.1b	2144.3	86.7
V 15.1a	2194.6	88.5
V 14.1b	2208.3	80.1
V 14.1a	2068.8	73.5
V 17.1b	2091.0	61.7
V 17.1a	2141.5	51.8
V 8.1	2196.1	40.2
V 6.1	1988.9	28.6
V 19.1a	1928.2	17.1
V 13.1a	2045.1	0.0

Source: The researcher.

ammonium carbonate led to increase the selectivity of zinc in the precipitation. It should always be considered that a large excess of precipitating agent might lead to the formation of complex ions and consequent partial solution of the precipitate. The effect of rotation speed did not lead to big gains in precipitation. Grinding of EAF dust to diminutive particles was to expand the contact area between the leach and dust and subject the zinc compound to the selective solvent for more reaction and therefore more precipitate. It should be noted that single parameters such as carbon dioxide, flow rate, rotation speed, temperature, pH value, conductivity and composition of the leach do not by themselves lead to a higher recovery of heavy metals. Hence to achieve the higher recovery of zinc and lead an appropriate combination of the mentioned parameters should be made. This was achieved through varying one parameter while holding the others constant. This was tried for all the parameters and in the mean time retaining those with highest precipitate on going. A break point was finally reached where only one optimum combination of

the parameters simultaneously produced the desired optimum recovery. This break point has the following specification:

Optimum mass of solution was 2746.9(g) being composed of 39.14% distilled water, 19.5% Ammonium carbonate 16.33% ammonium hydroxide and 24.99% EAF dust while the optimum level of carbon dioxide flow rate was 5 in/min, rotation speed was 700 rpm and temperature was 50°C.

Appendix A and B refer to some of the experimental work.

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Appendix A

Mass of Solution [g]: 2746.9

Parameters

CO₂ Flow [ln/min]: 5

Rotation Speed [rpm]: 700

Temperature [c]: 50

Time [min]	pH-Value	Temperature [°C]	Conductivity [ms/cm]
0	9.49	5.4	71.6
2	9.41	6.11	71.5
4	9.36	6.65	71.9
6	9.32	7.14	72.9
8	9.27	7.65	73.4
10	9.24	8.03	74
12	9.2	8.41	75
14	9.16	8.78	75.6
16	9.13	9	76.8
18	9.11	9.14	77.2
20	9.08	9.28	77.9
22	9.05	9.45	78.1
24	9.03	9.53	78.3
26	9	9.57	78.9
28	8.97	9.6	79.4
30	8.94	9.62	79.6
32	8.91	9.59	79.9
34	8.88	9.59	80.2
36	8.89	10.3	79.5
38	8.92	11.2	77.7
40	8.91	11.3	76.6
42	8.9	11.4	75.7
44	8.89	11.4	74.7
46	8.89	11.5	74.3
48	8.94	11.5	75.8
50	8.95	11.4	77.3
52	8.96	11.3	77.8
54	8.96	11.2	78.2
56	8.96	11	77.9
58	8.96	10.9	77.7
60	8.95	10.7	77.2

First precipitation [min]: 36

Mass of precipitate [g]: 170.6

Appendix B

Description of Leach V 20.1:

Mass [g] %			
Distilled Water	Ammonium Carbonate	Ammonium Hydroxide	EAF-Dust
H2O	[(NH4)2CO3]	[NH4OH] 25%	
39.16	19.5	16.33	24.99

Input Parameters:

Average Temperature	Rotation Speed	leaching time
[°C]	[rpm]	[min]
50	1000	90

Results:

Total Mass of Leach	Total Mass of Suspension	Density of Solution
[g]	[g]	[g / ml]
3600.2	4800.2	0.75