

# The evolution, chemistry and applications of chelated iron hydrogen sulfide removal and oxidation processes

Derek McManus<sup>a,\*</sup>, Arthur E. Martell<sup>b</sup>

<sup>a</sup> Wheelabrator Clean Air Systems, Inc., Plainfield, IL 60544, USA

<sup>b</sup> Texas A & M University, College Station, TX 77843, USA

Received 24 April 1996; accepted 30 May 1996

## Abstract

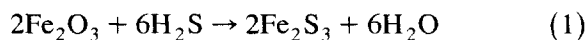
The evolution of the use of Fe(III) as a regenerable oxidant for the conversion of H<sub>2</sub>S to S is traced from inception as solid phase oxidation processes employing Fe<sub>2</sub>O<sub>3</sub>, through aqueous, alkaline Fe(OH)<sub>3</sub> suspensions and [Fe(CN)<sub>6</sub>] based processes to modern, aqueous, homogeneous redox catalysts using amino polycarboxylate chelated iron. In the current process, H<sub>2</sub>S bearing gas streams are contacted with a dilute ([Fe] = 5 mM to 0.5 M), aqueous, mildly alkaline (pH 7 to 9) amino or polyamino polycarboxylate iron (III) solution using a variety of application dependent gas–liquid contactors. S is precipitated and continuously removed. The Fe(III)L is concomitantly reduced to Fe(II)L and is regenerated by aeration in the same or a separate vessel, depending on the need to prevent admixture of reoxidation air with the H<sub>2</sub>S free process gas stream. Basic process chemical reactions are reviewed and both beneficial and troublesome side reactions, including ligand degradation by dioxygen reduction products and control thereof by in-situ generated S<sub>2</sub>O<sub>3</sub><sup>2-</sup>, are discussed together with associated mechanistic studies. Selection criteria for ligands suitable for use in this process are described and new chelating agents with significantly increased in-process stability are disclosed. The current global scope of applications is presented, advantages and limitations of the process relative to alternate technologies are discussed and the present status and future expectations are reviewed.

**Keywords:** Hydrogen sulfide; Sulfur; Iron chelates; Gas sweetening; Amino polycarboxylic acids

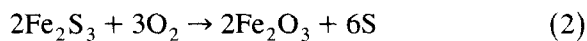
## 1. Introduction

Although the Fe(II)/(III) redox couple has no doubt served for eons to facilitate the oxidation of H<sub>2</sub>S to S by air in volcanic and geothermally active areas, the industrial use of Fe(III) as a regenerable oxidant for gas stream hydrodesulfurization commenced around 1830 with

the introduction of Fe<sub>2</sub>O<sub>3</sub> as a replacement for Ca(OH)<sub>2</sub> based processes [1].



Fe<sub>2</sub>O<sub>3</sub> was regenerated by simply exposing the Fe<sub>2</sub>S<sub>3</sub> to air.



Subsequently, suspensions of Fe(OH)<sub>3</sub> in aqueous Na<sub>2</sub>CO<sub>3</sub> solution were introduced [2]. Process reactions were similar to Eqs. (1) and (2) but some Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub> was also produced.

\* Corresponding author.

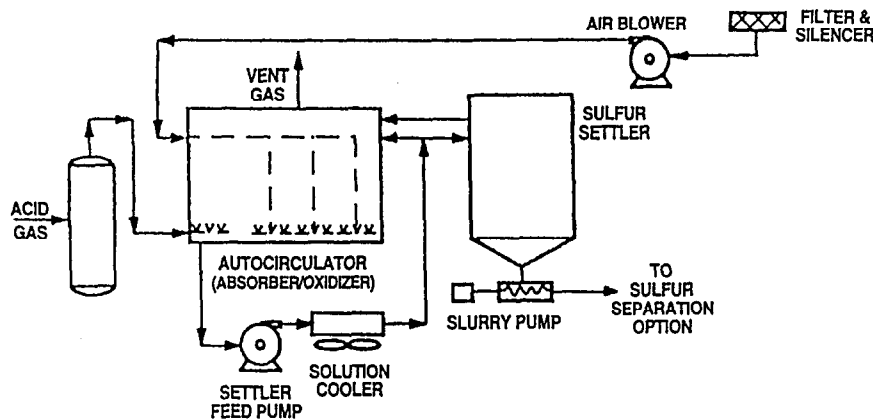


Fig. 1. Process flow diagram, LO-CAT II<sup>®</sup> autocirculator.

Iron based technology further evolved through hexacyanoferrate based processes [3] to current, aqueous, homogeneous redox catalysts employing amino and polyamino polycarboxylate chelated iron [4] (FeL, where L = NTA, EDTA or HEDTA). A superior catalyst [5] also contains a hexitol which deprotonates at high pH values and, in the presence of iron (III), forms a

very stable complex thereby preventing precipitation of Fe as  $\text{Fe}(\text{OH})_3$ .

Chelated iron process plant configurations can be many and varied [6] but all incorporate the basic unit operations of  $\text{H}_2\text{S}$  absorption, Fe(II)L oxidation and S removal. Typical process flow diagrams are shown in Figs. 1–3.

In operation, the  $\text{H}_2\text{S}$  containing gas stream

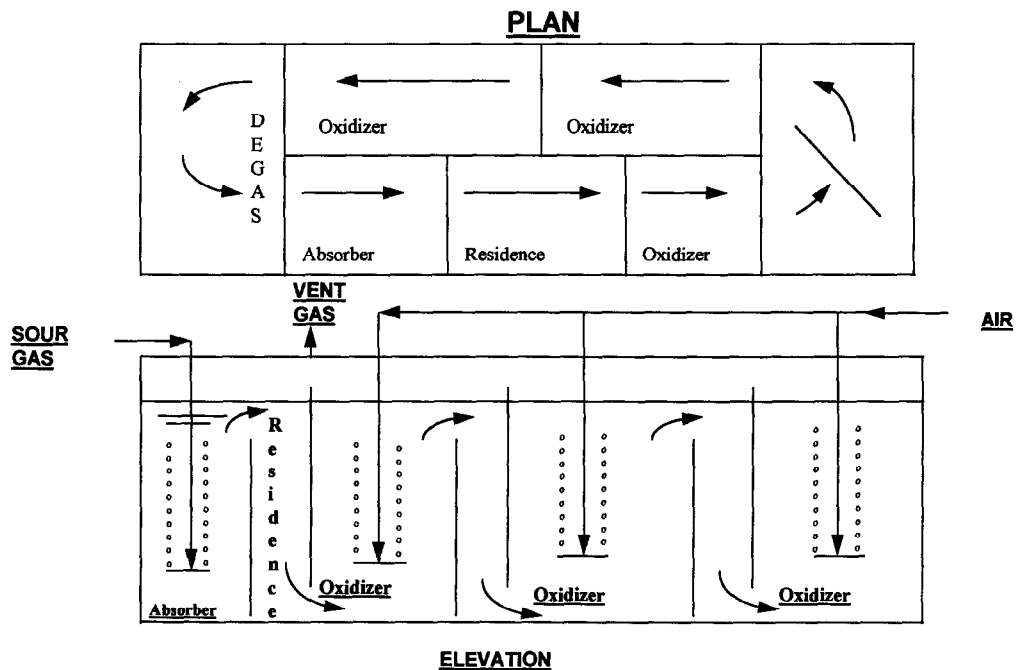


Fig. 2. Schematic, LO-CAT II<sup>®</sup> autocirculator design, plan and elevation.

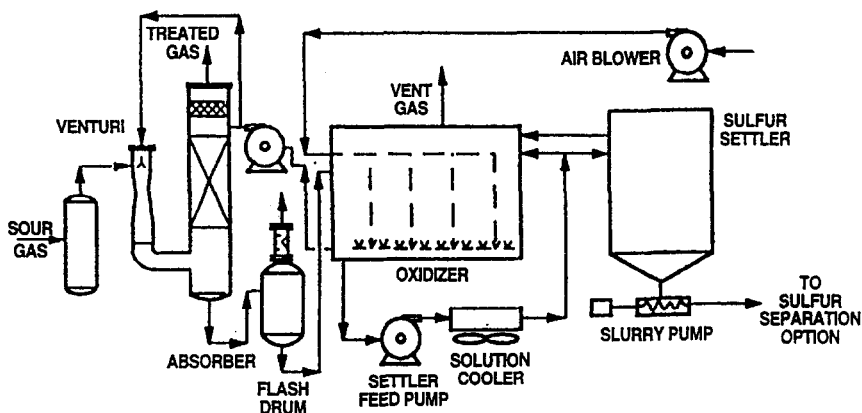
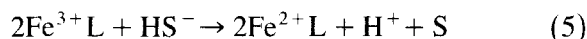
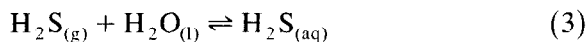


Fig. 3. Process flow diagram. LO-CAT II<sup>®</sup> conventional design.

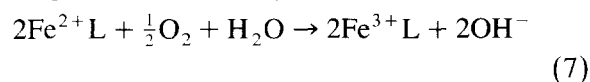
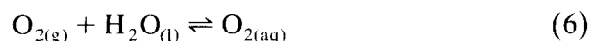
is contacted with aqueous, mildly alkaline (pH 7 to 9) amino polycarboxylato iron (III) solution ( $[\text{Fe}] = 5 \text{ mM to } 0.5 \text{ M}$ ) in the absorber vessel or zone where the below reactions take place.



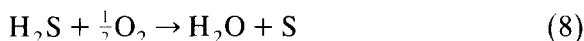
Absorbers are usually custom designed in order to accommodate the large range of sour gas stream compositions ( $[\text{H}_2\text{S}] = 50 \text{ ppm to } 100\%$ ), pressures (atmospheric to over 7 MPa (1000 psi)) and flow rates (up to  $65 \text{ m}^3 \text{ s}^{-1}$  (200 MMSCFD)). Absorber devices include spray chambers, packed towers with a variety of packing, both fixed and mobile, static mixers, venturis or eductors and liquid filled columns. Series combinations are also used.

Precipitated S is continuously removed by alternative methods including sedimentation, filtration or melting [7] under pressure.

The  $\text{Fe(II)L}$  formed in the absorber is oxidized by air in an oxidizing vessel or zone which can be integral with or separate from the absorber vessel depending on the need to avoid admixture of the  $\text{H}_2\text{S}$  free process gas with used oxidation air. Reactions occurring in the oxidizer are:



thus, the overall reaction is:



The introduction of chelated iron processes can be traced to the early 1960's [8] but no successful industrial operation emerged until small capacity systems were commissioned in the 1970's [9]. It was not until the first large (10 to 15 metric tons S/day) plants [10] equipped with sulfur melters and essentially operating with a captive catalyst inventory became operational around 1980 that the problem of chelate degradation was recognized. Operation could only be sustained by the continuous addition of considerable quantities of costly EDTA and HEDTA.

An urgent research program was initiated with the objectives of first defining the chemistry responsible for the prohibitive operating expense and second to correct the situation. Significant aspects of this work are described below.

## 2. Experimental

### 2.1. Instrumentation

Major instrumentation employed in this research included an Extrel (Madison, WI) Fourier transform mass spectrometer, FTMS-2001 (sulfide and polysulfide analyses).

The HPLC equipment used for quantitative amino polycarboxylate ligand determination included an IBM pump No. 771008, a variable wavelength UV–visible absorbance detector LDC analytical No. 3100 and a Kipp and Zonen strip chart recorder No. 6885.

The ESR spectrometer used to detect hydroxyl radicals in the iron chelate catalyst solution was manufactured by Bruker, model No. ESP300.

All other instrumentation, techniques and reagents are fully described in the provided references.

### 3. Results and discussion

#### 3.1. Iron chelate degradation studies

A 1000 cm<sup>3</sup> process simulation reactor [11], Fig. 4, was constructed and used to evaluate the stability of several, common amino and polyamino polycarboxylate iron chelates.

Ligands evaluated and ligand to iron mole ratios were nitrilotriacetic acid, NTA (1:1 and 2:1), ethylenediamine tetra acetic acid, EDTA (1.1:1), imino diacetate, IDA (3.2:1), diethylenetriamine penta acetic acid, DTPA (1:1) and cyclohexanediamine tetra acetic acid, CDTA (1:1).

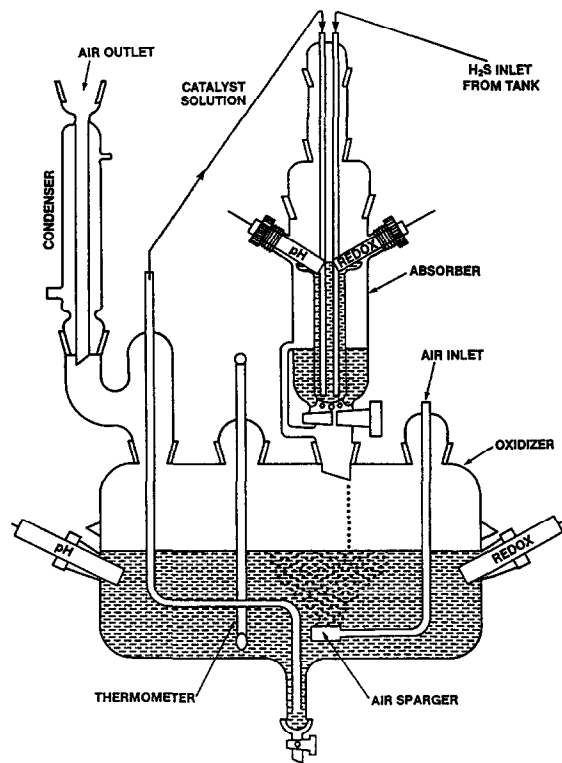


Fig. 4. Process simulation reactor.

Reactor operation was conducted at ambient temperature and pressure. H<sub>2</sub>S flow rate was held at 2.0 cm<sup>3</sup> min<sup>-1</sup> to provide a constant but unknown instantaneous concentration of hydro-sulfide ions in solution. Iron concentration was

Table 1  
Relative degradation rates of amino and polyaminopolycarboxylato iron (III)

Ligand	Ligand to iron mole ratio	Iron (mM)	Test duration (h)	Ligand degradation	
				1st order rate constant (h <sup>-1</sup> )	ligand half life (h)
NTA	2:1	18.2	51.6	0.023	30.6
NTA	2:1	18.0	62.0	0.024	29.5
NTA	2:1	15.7	47.5	0.023	30.7
NTA	1:1	18.8	46.8	0.062	11.1
EDTA	1.1:1	18.0	19.0	0.053	13.2
EDTA	1.1:1	18.0	120.5	0.059	11.8
IDA	3.2:1	18.0	130.0	0.027	25.3
DTPA	1:1	18.8	74.0	0.070	9.9
CDTA	1:1	18.0	12.6	0.053	13.1

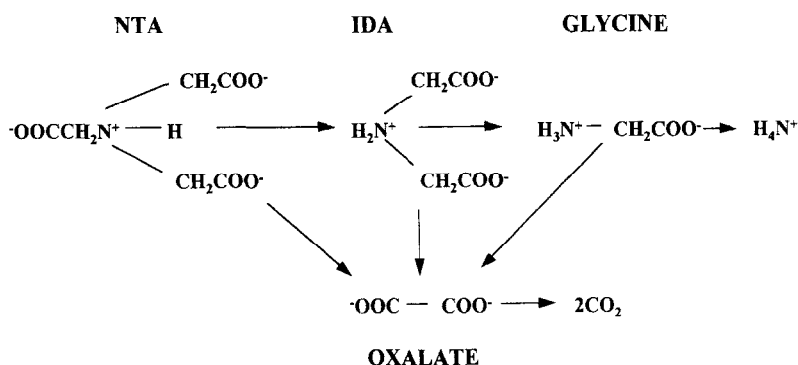


Fig. 5. Proposed degradation scheme for NTA.

18 mM. Aeration rate was  $1000 \text{ cm}^3 \text{ min}^{-1}$ , liquid circulation rate was  $80 \text{ cm}^3 \text{ min}^{-1}$  and the pH was held between 7 and 8.

Samples were periodically removed and prepared for HPLC analysis [12] by quantitative conversion of the iron (III) to the copper (II) chelate in order to produce a negative charge on the metal chelate as necessary to form an ion-pair with the quaternary counterion of the mobile phase. Invariably, a rapid loss of each of the above ligands was observed, Table 1.

HPLC also revealed the progressive formation of chelate degradation products. In the case of NTA, the retention times of two new peaks were consistent with those of IDA and oxalate. Subsequent gas chromatographic analysis [13] of the butylated and trifluoroacetylated derivatives of the two degradation products confirmed the assigned identities.

A further degradation product, glycine was subsequently identified by HPLC [11] in an analogous manner except that the iron (III) chelate was quantitatively converted to the nickel (II) chelate prior to analysis.

The proposed degradation scheme for NTA is shown in Fig. 5. No evidence of the probable intermediate, glyoxylate was observed. However, glyoxylate is reported elsewhere as a minor NTA degradation product together with *N*-oxalyl iminodiacetate and *N*-oxalyl glycine [14].

### 3.2. Iron chelate stabilization experiments

Parallel efforts aimed at providing an economic solution to intolerably high chemical operating costs encountered in several, early industrial plants proceeded on an urgent basis.

Additives such as antioxidants, buffers and

Table 2  
NTA stabilization by additives, system;  $(\text{NTA})_2 \text{ Fe(II)/(III)}$ ,  $[\text{Fe}] = 18 \text{ mM}$

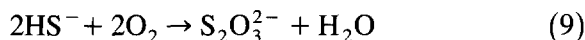
Additive	Additive concentration ( $\text{g L}^{-1}$ )	Test duration (h)	NTA degradation	
			1st order rate constant ( $\text{h}^{-1}$ )	NTA half life (h)
NTA (NTA:Fe, 5:1)	14.8	26.5	0.0270	25.8
Ethylene glycol	100.0	26.0	0.0034	156.8
Potassium citrate	50.0	23.5	0.0110	64.5
Sodium thiosulfate	50.0	24.0	"	"
Sodium thiosulfate	50.0	92.5	0.0013	531.0
Sodium hexametaphosphate	50.0	24.0	0.0420	16.5
Sodium thiocyanate	50.0	24.0	0.0034	205.0
Sodium <i>meta</i> and tetra borates	25.0 each	24.0	0.0240	28.6
<i>t</i> -butanol	100.0	26.0	"	"
Butylated hydroxyanisole	1.0	26.0	0.0380	18.4

" No measurable degradation observed.

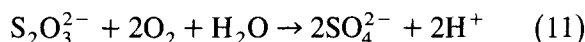
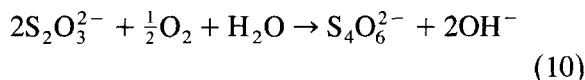
free radical scavengers were screened in a series of 24 h tests using the process simulation reactor. Several agents, particularly thiosulfate and thiocyanate anions and *t*-butanol were demonstrated to remarkably increase the stability of NTA, Table 2. Thiosulfate was selected as the agent of choice based on its low cost, chemical compatibility, low toxicity, superior performance and the fact that it can be generated in-situ from H<sub>2</sub>S.

### 3.3. Sulfur oxo-acid salt formation

Alkali metal thiosulfates are generated in the process and are beneficial at concentrations up to 0.5 M in protecting the iron chelates from degradation by oxygen radical attack. Higher concentrations of thiosulfate provide little further improvement in ligand stabilization.



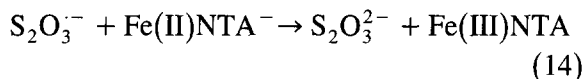
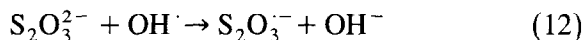
Thiosulfate has been shown to oxidize in the process to sulfate, via tetrathionate, by ion-chromatography.



Accumulation of collective sulfur oxo-acid salts over time previously required the irrevocable discarding of valuable catalyst solution to maintain an adequately low total solute concentration for facile sulfur separation, efficient gas to liquid mass transfer and avoidance of salt crystallization in the plant.

Recent process design innovations allow thiosulfate to be generated on demand to reach the optimum concentration for maximum stabilization of the iron chelate but no more than that amount. This is achieved by maximizing the sulfur forming reaction (Eq. (5)) and utilizing the thiosulfate generating reaction (Eq. (9)) only when required by controlling access of HS<sup>-</sup> ions to liquid zones possessing significant dissolved oxygen contents.

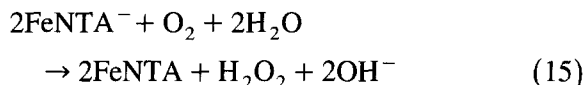
The possibility that thiosulfate ions can be regenerated in the process subsequent to reacting to scavenge hydroxyl radicals has recently been suggested [15] and is supported by earlier work involving pulsed radiolysis of aqueous thiosulfate [16].



Further studies will be required to determine whether these mechanisms are valid.

### 3.4. Chelate degradation mechanistic studies [17]

A series of stoichiometric reactions, each separately representing a single process stage, were conducted to establish exactly where the chelate degrades in the process. It was conclusively demonstrated that FeNTA degrades during oxidation of Fe(II) NTA by air to Fe(III) NTA.



Eqs. (15) and (16) are consistent with the experimental observation that lower pH values increase the rate of ligand degradation in that more acidic conditions favor hydroxyl radical formation.

Furthermore, addition of catalase which, by destroying precursor H<sub>2</sub>O<sub>2</sub>, was demonstrated to significantly reduce the rate of ligand degradation supports the contention that hydroxyl radicals are responsible for the destruction of iron chelates in the process.

Additional evidence implicating hydroxyl radicals was obtained by the formation of *o*-, *m*-, and *p*-hydroxybenzoates from added benzoate (molecular probe) [17], these same products

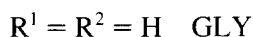
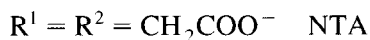
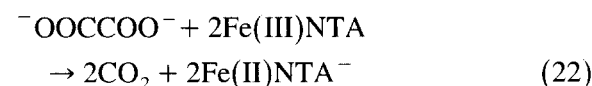
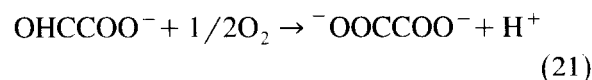
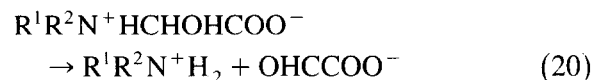
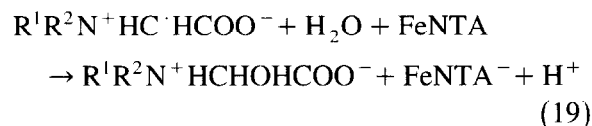
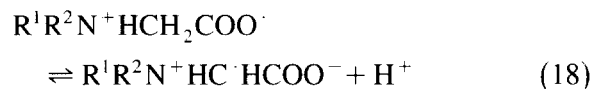
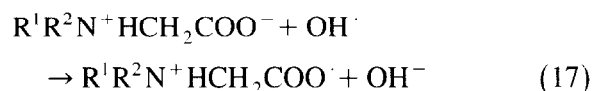
being formed from benzoate by hydroxyl radicals generated by pulse radiolysis [18].

ESR provided further experimental evidence that ligand degradation is initiated by the hydroxyl radical. Phenyl *t*-butyl nitron was used as a spin trap and the radical generated after hydroxylation by OH $\cdot$  was detected in both control (Fe(II)NTA, H<sub>2</sub>O<sub>2</sub>) and test (Fe(III)NTA, H<sub>2</sub>S, air) systems at pH 8.5.

Although some controversy endures [19] as to whether free OH $\cdot$  and not high valent iron-oxo species such as FeO<sup>3+</sup> (formerly Fe (V)), ferryl complexes (L) Fe(IV)=O or (L<sup>+</sup>)Fe(IV)=O and (L<sub>*n*</sub>)Fe(IV)(OH)(OOH) is primarily responsible for ligand degradation, the above evidence plus the fact that SO<sub>4</sub><sup>2-</sup> and not S would be produced and that the reaction occurs in aqueous solution again implicate OH $\cdot$ .

This debate will likely continue until the presence of H<sub>2</sub>O<sub>2</sub> in the reaction solution is unambiguously confirmed. Attempts to detect H<sub>2</sub>O<sub>2</sub> by chemiluminescence have been made [20] but results to date remain inconclusive due to the dark color of the solution and the low anticipated concentration of H<sub>2</sub>O<sub>2</sub> (10<sup>-5</sup> M).

Based on the above stated evidence, the following sequence of reactions is proposed to explain the degradation of FeNTA.



Other similar polyamino polycarboxylato-iron chelates are presumed to degrade similarly in the process. The point of attack being the CH<sub>2</sub> groups of the acetate chains.

### 3.5. Sulfur formation

Several mechanisms have been postulated to account for the formation of elemental sulfur in the circulating, chelated iron catalyst solutions. It is reported [21] that iron (III) *N*-hydroxyethyl ethylenediamine triacetate reacts with hydrogen sulfide in aqueous solution forming a pink unstable sulfido bridged complex, (Fe(III)HEDTA)<sub>2</sub>S<sup>2-</sup>, which probably has a similar structure to that of the well characterized  $\mu$ -oxo dimer [22]. However, speculation about the formation of a similar sulfido bridged dimer complex (FeL)<sub>2</sub>S when nitrilotriacetic acid (NTA) is the ligand has not been borne out by laser desorption Fourier transform, ion cyclotron resonance mass spectrometry [23]. Nevertheless, the  $\mu$ -oxo dimer of Fe(III)NTA has also been described [24] and the corresponding  $\mu$ -sulfo dimer could be present in trace quantities.

Recent potentiometric titrations and spectrophotometric studies have shown that the hydroxo-bridged dimeric iron (III) NTA complex, (FeNTA)<sub>2</sub>OH does not exist at millimolar concentrations at the process pH of 8 nor does Fe(NTA)<sub>2</sub>(OH) [25]. It is therefore unlikely that sulfide or polysulfide could displace a hydroxyl anion of a hypothetical bridged dimeric iron-NTA chelate in the manner proposed for iron-EDTA chelates.

Although further studies are needed to fully elucidate the role of iron (III) NTA in the conversion of monosulfide to cyclo octasulfur, it has been positively established that polysulfides are reactive intermediates in the catenation and cyclization process [23].

Much additional work will be necessary in order to develop a comprehensive understanding of the reaction mechanisms and pathways leading to elemental sulfur formation.

### 3.5.1. Ligand selection criteria [26] and novel chelates

Ligands useful in chelated iron hydrogen sulfide oxidation processes must meet the below chemical criteria and additionally must be available in commercial quantities at moderate cost, possess low toxicity and present no environmental problems.

As gas to liquid mass transfer of  $H_2S$  is much more efficient in aqueous systems at high pH, the iron chelates must possess sufficiently high thermodynamic stabilities to avoid precipitation of  $Fe(OH)_2$  and  $Fe(OH)_3$  at pH values up to 9 or more.

Similarly, the stability of the iron (II) chelate must be great enough to prevent precipitation of  $FeS$  under mildly alkaline conditions.

The difference in stabilities of the iron (II) and iron (III) chelates must be low enough to allow reduction of the iron (III) chelate by  $H_2S$  to the iron (II) chelate ( $\Delta pM$  should be between 5 and 16). If the iron (III) chelate is too stable,  $\Delta pM > 16$ , the chelate remains in the iron (III) state and no oxidation of  $H_2S$  will occur.

Additionally, the stability of the iron (III) chelate must be greater than that of the iron (II) chelate for oxidation of the iron (II) chelate by dissolved oxygen to be a highly favored reaction. Chelating agents that stabilize iron (II) over iron (III) are not useful in the process.

A final criterion relates to the inherent resistance of the iron chelate to in-process degradation by dioxygen reduction products. Amino polycarboxylate ligands are especially vulnerable to oxidation via hydroxyl radical attack at  $\alpha\text{-CH}_2$  sites [17]. Ligands lacking this structural component have been demonstrated to degrade to a lesser extent in process simulation reactor experiments. For example, pyridine 2,6-dicarboxylato iron (II)/(III) exhibits superior stability in the process [27], but unfortunately

fails to maintain adequate concentrations of the iron (III) complex in solution under the prevailing alkaline conditions, even with L:M ratios as high as 5:1.

However, pyridine 2,6-diacetato iron (II)/(III) degrades rapidly under identical experimental conditions [28].

Research continues with the synthesis and evaluation of similar compounds with different substituent functional groups on the pyridine ring.

## 4. Summary

Chelated iron process applications [29] include natural gas sweetening (amine acid gas, direct treating), oil refinery operations (hydro-treater off-gas, fuel gas, sour water stripper gas), enhanced oil recovery (carbon dioxide recycle), marine vessel loading, underground oil shale retorting, landfill gas treating, waste water treating plant odor control, biogas treating, geothermal electric power generation, coke oven gas treating, beverage quality carbon dioxide production and a large variety of chemical manufacturing operations (viscose, rayon, phosphorus pentasulfide, phosphoric acid, silicon carbide, titanium dioxide).

Advantages over alternate technologies include a non-toxic, environmentally benign catalyst solution as opposed to caustic non-regenerable reagents, ease of operation, fast kinetics, relatively high selectivities for both gas absorption and sulfur formation, no turn-down or gas compositional limitations and cost effectiveness up to a plant capacities of 30 tons of sulfur per day. Approximately 150 chelated iron plants with a total estimated annual sulfur production of 150,000 metric tons have been installed.

Ongoing research and development continues with programs aimed at gaining further improvements in the process economics. Both plant capital expense and process operating cost reductions are anticipated from successful conclusion of current research projects at both Texas



A&M University, College Station, Texas and WCAS's new research facility at Plainfield, IL.

It is anticipated that the market for the chelated iron processes will continue to grow well into the next century as dictated by consumer demands and environmental requirements.

## Acknowledgements

A substantial fraction of this research was supported by Wheelabrator Clean Air Systems, Inc. (WCAS) Schaumburg, IL, USA and ARI Technologies, Inc., a WCAS predecessor company. Additionally, the authors appreciate helpful discussions with Professor D.H.R. Barton of Texas A&M University (College Station).

## References

- [1] A.L. Kohl and F.C. Riesenfeld, *Gas Purification* (McGraw-Hill, New York, 1960) pp. 241–259.
- [2] A.L. Kohl and F.C. Riesenfeld, *Gas Purification* (McGraw-Hill, New York, 1960) pp. 283–295.
- [3] A.L. Kohl and F.C. Riesenfeld, *Gas Purification* (McGraw-Hill, New York, 1960) pp. 303–308.
- [4] R.B. Thompson, U.S. Patent 4,189,462, Feb. 19 (1980); L.C. Hardison, U.S. Patent 5,139,753, Aug. 18 (1992); 5,160,714, Nov. 3 (1992).
- [5] R.B. Thompson, U.S. Patent 4,218,342, Aug. 19 (1980).
- [6] D. McManus and W.J. Niemiec, 1996 AIChE Spring Natl. Meet., New Orleans, LA, USA, Paper 46b; D. McManus and M. Reicher, 1995 AIChE Spring Natl. Meet., Houston, TX, USA, Paper 54a; D. McManus, 1994 AIChE Spring Natl. Meet., Atlanta, GA, USA, Paper 27b.
- [7] D. McManus, Gas Research Institutes (Chicago) 7th Sulfur Recovery Conf., Austin, TX, USA, September (1995).
- [8] W. Hartley, R.S. Craig and R.H. Sapiro, U.S. Patent 3,068,065, Dec. 11 (1962).
- [9] Amax, Golden, CO, USA and Hooker Chemical Company, Columbia, MS, USA.
- [10] R.K. Leicht et al., *Chemical Processing* (Fletcher Oil and Refining Company, Carson, CA, USA, August 1986); J. Cabodi et al., *Oil and Gas Journal* (U.S. Oil and Refining Company, Tacoma, WA, USA, July 5, 1982).
- [11] D. Chen, R.J. Motekaitis, A.E. Martell and D. McManus, *Can. J. Chem.* 71 (1993) 1524–1531.
- [12] D. McManus, 41st Pittsburgh Conf. and Exposition on Anal. Chem. and Appl. Spectrosc., New York City (1990); 211th Am. Chem. Soc. Natl. Meet., New Orleans, LA, USA, March 24–28 (1996), Paper 31; G.A. Perfetti and C.R. Warner, *J. Assoc. Off. Anal. Chem.* 62(5) (1979) 1092; D.G. Parks, M.G. Caruso and J.E. Spradling, *Anal. Chem.* 53 (1981) 2154.
- [13] C.B. Warren and E.J. Malec, *J. Chromatogr.* 64 (1972) 219–237.
- [14] E.F. Lutz, U.S. Patent 4,443,418, April 17 (1984).
- [15] D.W. DeBerry, 1996 AIChE Spring Natl. Meet., New Orleans, LA, USA, paper 46a.
- [16] R. Mehnert, O. Brede and I. Janovsky, *Radiat. Phys. Chem.* 23 (1984) 463–468.
- [17] D. Chen, A.E. Martell and D. McManus, *Can. J. Chem.* 73 (1995) 264–274; D. Chen, R.J. Motekaitis, A.E. Martell and D. McManus, *Can. J. Chem.* 71 (1993) 1524–1531.
- [18] G.W. Klein, K. Bhatia, V. Madhavan and R.H. Schuler, *J. Phys. Chem.* 79 (1975) 1767.
- [19] X. Liu, D.T. Sawyer, S.A. Bedell and C.M. Worley, Gas Research Institutes (Chicago), 7th Sulfur Recovery Conf., Austin, TX, USA, September (1995).
- [20] D.W. DeBerry and D. McManus, private communication.
- [21] C.V. Phillip and D.W. Brooks, *Inorg. Chem.* 13(2) (1974) 384–386; D.W. DeBerry, B. Petrinec and T. Trofe, Gas Res. Institutes Liquid Redox Sulfur Recovery Conf., Austin, Texas, May 5–7 (1991).
- [22] H. Schugar, C. Walling, R.B. Jones and H.B. Gray, *J. Am. Chem. Soc.* 89 (1967) 3712.
- [23] E.T. Clarke, T. Solouki, D.H. Russell, A.E. Martell and D. McManus, *Anal. Chim. Acta* 299 (1994) 97–111.
- [24] G. McLendon, R.J. Motekaitis and A.E. Martell, *Inorg. Chem.* 15 (1976) 2306.
- [25] R.J. Motekaitis and A.E. Martell, *J. Coord. Chem.* 31 (1994) 67–78.
- [26] A.E. Martell, R.J. Motekaitis, D. Chen, R.D. Hancock and D. McManus, 1995 AIChE Spring Natl. Meet., Houston, TX, USA, Paper 54c.
- [27] D.T. Sawyer and A. Sobkowiak, U.S. Patent 5,273,734, Dec. 28 (1993).
- [28] D. Chen, A.E. Martell and D. McManus, unpublished work.
- [29] G.J. Nagl, Gas Research Institutes (Chicago) 7th Sulfur Recovery Conf., Austin, TX, USA, September (1995).