Chemical Treatment of Pesticide Wastes. Evaluation of Fe(III) Chelates for Catalytic Hydrogen Peroxide Oxidation of 2,4-D at Circumneutral pH

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A number of structurally diverse organic and inorganic polydentate chelators were tested for their ability to solubilize Fe(III) at pH 6 and catalyze the oxidation of 0.1 mM (22 ppm) 2,4-dichlorophenoxyacetic acid (2,4-D) by 10 mM (340 ppm) hydrogen peroxide in aerated aqueous solution. Of 50 compounds tested, 20 organic compounds gave soluble complexes capable of oxidizing 2,4-D in periods ranging from minutes to less than 5 h. The reaction rate was proportional to the concentration of H_2O_2 and inversely proportional to the concentration of chelator. 2,4-Dichlorophenol was a transient intermediate. Chloride was released concurrently with 2,4-D disappearance. Activities of individual complexes toward 2,4-D transformation and hydrogen peroxide decomposition were similar. Several of the most active complexes gave about 80% mineralization of [*ring*-^{14}C]- or [*carboxy*-^{14}C]-2,4-D within 4 h. Additional supplements of H_2O_2 increased the extent of mineralization. All active organic ligands were themselves oxidized, but in many cases the resulting iron complexes remained solubilized and reactive, often more reactive than the original complex. From a practical perspective, degradation of the ligand may be viewed as desirable.

INTRODUCTION

Pesticide wastes generated on-site in the form of aqueous rinsates and unused product are a potential threat to water supplies and have come under increasingly stringent regulation. An additional concern is contaminated soil resulting from accidental spills or improper handling or disposal at mixing/loading sites. Since transport to off-site commercial incinerators is costly for small-scale wastes, alternative chemical or microbiological treatment methods are needed. A number of approaches have been investigated (Nannipieri and Bollag, 1991; Somich et al., 1990; EPA, 1985, 1986; Krueger and Seiber, 1984; Joshi et al., 1985; Pignatello, 1992).

A promising chemical treatment method is hydrogen peroxide oxidation in the presence of Fe salts, which generates hydroxyl radical (OH•) and probably other strong oxidants. It was recently shown that acidic aerated solutions of dilute H_2O_2 and Fe^{3+} rapidly degraded chlorophenoxyalkanoic acids, 2,4-dichlorophenoxyacetic acid (2,4-D), and 2,4,5-trichlorophenoxyacetic acid (2,4,5-T), to carbon dioxide and chloride (Pignatello, 1992). However, the reaction rate drops off sharply above and below the optimum pH of about 2.8. At higher pH, loss of reactivity is due to precipitation of Fe^{3+} as hydrous oxyhydroxides, $Fe_2O_3 \cdot nH_2O$. The narrow effective pH range limits the utility of Fe/H_2O_2 for treating contaminated soil and certain compounds that may be less reactive or less soluble in acidic media. Acidification of soil to the proper pH is cumbersome in a batch mode and may preclude or severely restrict implementation of Fe/H_2O_2 for in situ treatment. Moreover, acidification and subsequent neutralization will add two additional steps to any scheme.

Iron(III) may be solubilized at circumneutral pH by chelation (Anderson and Hiller, 1975; Martell, 1980; Porter, 1989). To be useful, an Fe(III) chelate must also (i) have catalytic activity toward oxidation of the waste component, i.e., be capable of generating hydroxyl radical or other reactive oxidant from H_2O_2; (ii) be resistant to oxidation in the medium; and (iii) be environmentally safe. With these considerations, we searched for a suitable chelating agent from among several types of polydentate organic and inorganic compounds.

We chose 2,4-D as a model owing to our previous experience with it in Fe/H_2O_2 media under acidic conditions. 2,4-D is commercially important for control of broad-leafed weeds on lawns and crops, and its health hazards have been well documented (EPA, 1987; VNR Information Services, 1987). Other chemical reagents that have been investigated for 2,4-D destruction include ozone (Joshi et al., 1985), potassium polyethylene glycolate (Taylor et al., 1990), and concentrated nitric acid (Leavitt and Abraham, 1990).

MATERIALS AND METHODS

Materials. Hydroxyethyliminodiacetic acid (HEIDA), [ethylenebis(oxyethylenenitrilo)]tetraacetic acid (EGTA), nitrilotriacetic acid (NTA), alizarin red s, and citric acid were obtained from J. T. Baker; oxalic acid, sodium pyrophosphate, sodium tripolyphosphate (STPP), and ethylenediaminetetraacetic acid (EDTA) from Mallinckrodt; ethylenediaminedi-*o*-hydroxyphenylacetic acid (EDDHA) and *N*-(hydroxyethyl)ethylenediaminetriacetic acid (HEDTA) from K & K Laboratories; acetohydroxamic acid, picolinic acid (pica), rhodizonic acid, tetrahydroxy-1,4-quinone hydrate (THQ), gallic acid, quercetin, rutin, adenosine 5′-diphosphate (ADP), adenosine 5′-triphosphate (ATP), hemin, hematin, and hematoporphine X from ICN Biochemicals; mucic acid, 1,2-diacetylhydrazine, pyrazinecarboxylic acid, oxonic acid, 5-nitroorotic acid, catechol, hexaketocyclohexane (HKCH), and violuric acid from Aldrich Chemical Co.; DL-tartaric acid, malonic acid, diurea, and biuret from Matheson Coleman & Bell; dipicolinic acid (dipica) and pyrocatechol violet from Kodak; 2,3-dihydroxybenzoic acid (DHBA), sodium trimetapolyphosphate, sodium carbamylphosphate, sodium imidodiphosphate, trisodium phosphonoformate, dodecasodium phytate, and sodium ketomalonate from Sigma; and sodium pyrosulfate and sodium dithionate from Pfaltz & Bauer (Waterbury, CT). L-Malic acid was obtained from Mann Research Laboratories and ascorbic acid from Fisher. All chemicals were used as received.

Disodium hypophosphate was synthesized from red phosphorus (Mallinckrodt) and H_2O_2 in concentrated aqueous ammonia (Dragulescu et al., 1963). Disodium carbonyldiphosphate was synthesized from tetraisopropyl methylenediphosphate (Lan-

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Table I. Chelating Ability of Ligands for Iron(III) at pH 6 and Activity of the Chelates with Respect to Oxidation of 2,4-D by H_2O_2[a]

ligand	soluble?	activity
aminopolycarboxylates		
EDTA	yes	inactive
HEDTA	yes	inactive
EDDHA	yes	inactive
EGTA	yes	inactive
NTA	yes	high
HEIDA	yes	high
polycarboxylates		
citric acid	yes[b]	low
mucic acid	yes	moderate
malonic acid	yes	moderate
L-malic acid	yes	inactive
ketomalonic acid	yes[b]	low
DL-tartaric acid	yes	low
oxalic acid	yes[b]	low
hydroxamates		
acetohydroxamic acid	yes[b]	inactive
violuric acid	no[b]	nt[c]
N-heterocyclic carboxylates		
picolinic acid	yes	high
dipicolinic acid	yes	moderate
pyrazinecarboxylic acid	no[b]	nt
oxonic acid	no[b]	nt
5-nitroorotic acid	no[b]	nt
polyhydroxy aromatics		
catechol	yes	moderate
1,2-dihydroxybenzoic acid	yes	moderate
gallic acid	yes	high
quercertin	yes	moderate
rutin	yes	inactive
pyrocatechol violet	yes	moderate
alizarin red	yes	low
porphyrins		
hemin	yes	inactive
hematin	yes	inactive
hematoporphine X	yes	inactive
sulfur compounds		
pyrosulfate	no[b]	nt
dithionate	no[b]	nt
phosphates		
pyrophosphate	yes	inactive
hypophosphate	yes[b]	inactive
tripolyphosphate	yes	inactive
trimetapolyphosphate	yes[b]	inactive
phytate	yes[b]	inactive
imidodiphosphate	yes[b]	inactive
carbonyldiphosphate	yes[b]	inactive
carbamylphosphate	no[b]	nt
phosphonoformate	yes[b]	inactive
adenosine 5'-diphosphate	yes	inactive
adenosine 5'-triphosphate	yes	inactive
miscellaneous		
rhodizonic acid	yes	high
tetrahydroxy-1,4-quinone	yes	high
ascorbic acid	yes[b]	moderate
hexaketocyclohexane (HKCH)	yes	high
biuret	no[b]	nt
diurea	no[d]	nt
1,2-diacetylhydrazine	no[b]	nt

[a] [Fe(III)] = 1.0 mM; 1:1 ligand–Fe molar ratio unless noted otherwise. [b] At 10:1 ligand–Fe molar ratio. [c] nt, not tested. [d] Only slightly soluble. Saturated diurea solution was used.

caster) and sodium hypochlorite (Fisher) in 1,1,2,2-tetrachloroethane (Kodak) (Quimby et al., 1967).

Procedures. Iron chelate solutions were made by directly mixing aqueous solutions of Fe^{3+} and the corresponding ligand. Aqueous Fe^{3+} was prepared fresh daily by dissolving anhydrous $Fe(ClO_4)_3$ (GFS Chemicals) in 0.1 M perchloric acid. The Fe chelate solutions were adjusted to pH 6 with NaOH. Ionic strength was maintained at 0.2 M by $NaClO_4$. The neutral ADP solution formed a white milky suspension upon the addition of the acidic Fe^{3+} solution. The mixture became clear when neutralized to pH 6.0. The hemin and hematin porphyrins already contain iron and were used directly after the pH was adjusted. Hematoporphine X was first dissolved in NaOH, mixed with Fe^{3+} solution, and then acidified to pH 6 with $HClO_4$.

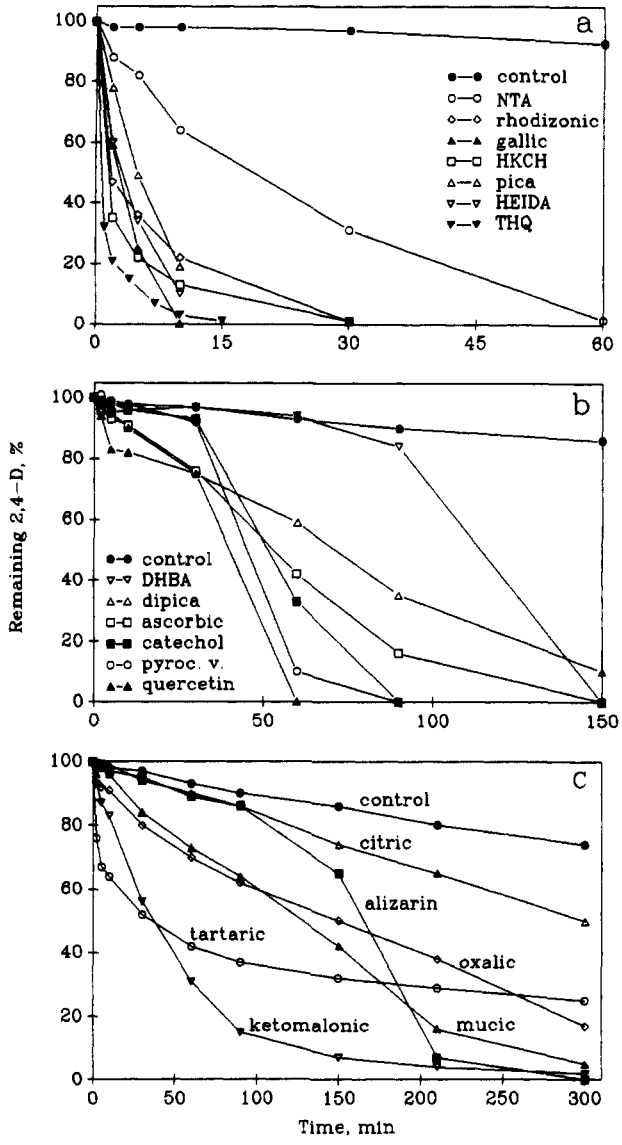

Figure 1. Transformation of 2,4-D by active iron(III) chelates: (a) high, (b) moderate, and (c) low activity. [2,4-D] = 0.1 mM; [chelate] = 1.0 mM; [H_2O_2] = 10 mM; pH 6.0.

Reactions were carried out in open Erlenmeyer flasks with continuous stirring at room temperature. Control experiments showed no appreciable volatilization of 2,4-D. Reactions were initiated by adding hydrogen peroxide. Aliquots (2 mL) for HPLC analysis were added to 3 mL of acidified ($HClO_4$) methanol, which quenched the reaction.

Some reactions were supplemented with 10^4 DPM/mL [*ring*-UL-^{14}C]-2,4-D (10.0 mCi/mmol) or [*carboxy*-^{14}C]-2,4-D (9.0 mCi/mmol) (both from Sigma). Solution-phase radioactivity was determined by scintillation counting as described previously (Pignatello, 1992) after a 1-mL aliquot of the reaction mixture was quenched in 1 mL of 2 M $HClO_4$ plus 0.5 mL of methanol and sparged to remove $^{14}CO_2$.

Analyses. 2,4-D and 2,4-dichlorophenol (DCP) were monitored by HPLC, and hydrogen peroxide was analyzed iodometrically in the presence of NaF, as previously described (Pignatello, 1992). UV–visible spectra were recorded on a double-beam Perkin-Elmer spectrophotometer (Hitachi 200).

RESULTS

Chelating Ability and Catalytic Activity. Many of the 50 compounds investigated formed soluble 1 mM 1:1

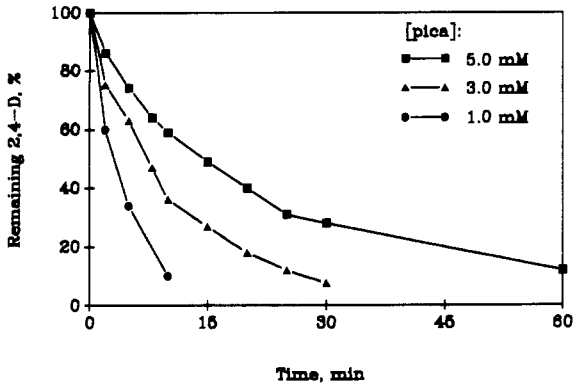

Figure 2. Dependence of 2,4-D transformation on picolinic acid concentration. Concentrations of other reagents are the same as in Figure 1.

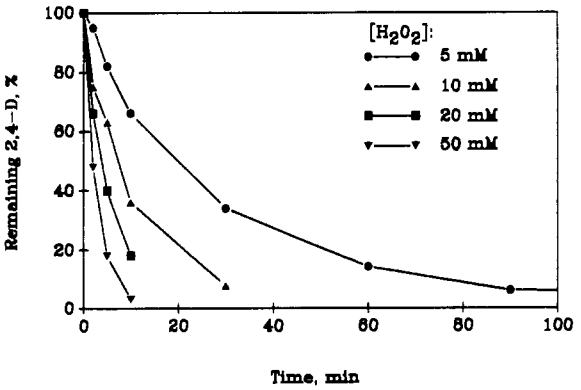

Figure 3. Dependence of 2,4-D transformation on hydrogen peroxide concentration. [pica] = 3.0 mM; other conditions are as in Figure 1.

Fe(III) complexes at pH 6 in aqueous solution (Table I). Others became soluble at higher ligand–metal ratios. The soluble complexes were further tested for activity in oxidation of 2,4-D by 10 mM H_2O_2 at pH 6. Table I indicates the relative activity of each complex during a 300-min reaction period. "Inactive" means that the complex was less effective, or only marginally more effective, than the control, which consisted of precipitated Fe(III) at pH 6 in the absence of a complexing ligand. High, moderate, and low activities indicate effectiveness in removal of 2,4-D within approximately 60, 150, and 300 min, respectively. The 2,4-D disappearance curves for the 20 active complexes are presented in Figure 1.

With the most active chelates (NTA, HEIDA, rhodizonic acid, gallic acid, HKCH, pica, and THQ), 2,4-D disappeared in minutes, which is comparable in rate to unchelated Fe(III) at pH 2.8 (Pignatello, 1992). Using the pica chelate, the rate of disappearance of 2,4-D decreased with increasing ligand concentration (Figure 2) and increased with increasing hydrogen peroxide concentration (Figure 3).

The only peak in the HPLC chromatogram attributable to organic degradation products corresponded to 2,4-dichlorophenol (DCP). DCP always reacted within the lifetime of 2,4-D in the particular system and never exceeded 8% of theoretical maximum yield (Figure 4). Similar results were obtained in the unchelated systems at pH 2.8 (Pignatello, 1992).

It has been reported that chlorine was released concurrently with the disappearance of parent compounds in unchelated systems in acidic media (Pignatello, 1992) as inorganic chloride ion, measured potentiometrically. However, in the chelated systems, the presence of some chelating agents interfered with the determination. Nevertheless, in one system (1 mM gallate chelate, 10 mM H_2O_2, and 0.1 mM 2,4-D, pH 6), gravimetric determination indicated a 95% yield of chloride ion 15 min after the reaction started.

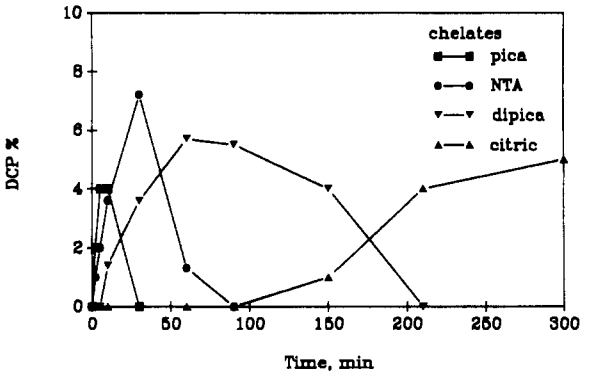

Figure 4. Production and disappearance of 2,4-dichlorophenol during 2,4-D transformation. Conditions are the same as in Figure 1.

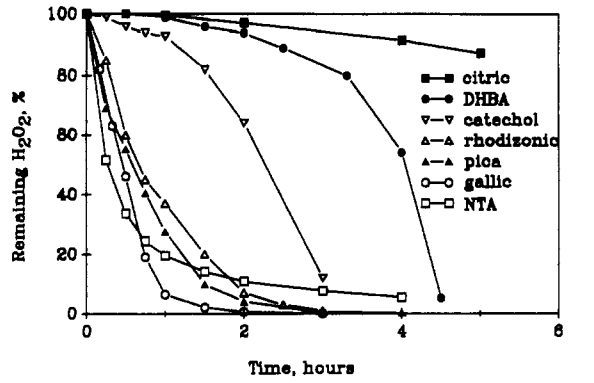

Figure 5. H_2O_2 decomposition by iron(III) chelates. [H_2O_2] = 10 mM; [chelate] = 1.0 mM; pH 6.0.

Active Fe(III) chelates also catalyzed the decomposition of H_2O_2; a few examples are given in Figure 5. The decomposition of H_2O_2 by Fe–NTA was initially fast but then slowed concurrent with degradation of the ligand and precipitation of Fe(III) (see below). Comparison of Figure 5 with Figure 1 reveals that, except for the NTA chelate which precipitated during the reaction, chelate activities toward both decomposition of H_2O_2 and transformation of 2,4-D followed the same order:

$$\text{citric} < \text{DHBA} < \text{catechol} < \text{rhodizonic} \approx \text{pica} < \text{gallic}$$

Chelate Stability. Virtually all active Fe(III) complexes were themselves unstable to the oxidant, presumably due to transformations of the ligand. In the absence of 2,4-D, most complexes showed color changes on addition of H_2O_2 (Table II). The absorbance spectrum of the catecholate complex, for example, changed rapidly on addition of H_2O_2 (Figure 6); the absence of an isosbestic point suggests that the ligand underwent more than a single transformation. For some chelates, a color change occurred slowly even without H_2O_2; the brown colors of rhodizonic acid and THQ complexes changed slowly to yellow. This behavior may indicate air oxidation or oxidation by Fe(III) itself.

Ligand oxidation was usually followed eventually by precipitation of orange-brown Fe(III) oxyhydroxides, after times ranging from hours to days in 10 mM H_2O_2 (Table II). Typically, the pH then fell to about 4 due to release of protons from hydrolyzed iron. All chelates precipitated overnight at high H_2O_2 concentration (0.1–0.5 M). Pre-

Table II. Stability of Active Fe(III) Chelates in 10 mM H_2O_2 at pH 6

chelate	color change	precipitation within
NTA	yellow[a]	2–3 h
HEIDA	yellow[a]	3–4 h
mucic acid	yellow[a]	3 days
malonic acid	yellow[a]	1 days
malic acid	yellow[a]	3 days
ketomalonic acid	yellow[a]	1 days
tartaric acid	yellow[a]	3 days
oxalic acid	yellow[a]	1 days
pica	orange to yellow[b]	7 days
dipica	yellow[a]	>7 days
catechol	dark brown to yellow[b]	3 days
DHBA	dark blue to yellow	7 days
ascorbic acid	yellow[a]	<16 h
gallic acid	dark blue to yellow	7 days
rhodizonic acid	brown to yellow	5 days
HKCH	yellow[b]	3 days
THQ	brown to yellow	5 days
quercetin	dark brown to yellow	>7 days
pyrocatechol violet	dark brown to yellow	>7 days
alizarin red	dark brown to yellow	7 days
citric acid	yellow[a]	>7 days

[a] No color change on addition of hydrogen peroxide; however, precipitation indicates eventual oxidation. [b] UV spectral changes were observed.

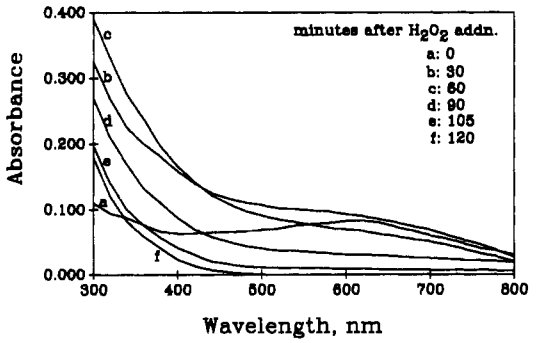

Figure 6. UV–visible spectral change of catechol–iron(III) chelate during oxidation by H_2O_2. [H_2O_2] = 10 mM; [chelate] = 1.0 mM; pH 6.0.

cipitated solutions lost their ability to effect 2,4-D degradation (data not shown).

The transformation of 2,4-D displayed an induction period when complexes of catechol, DHBA, alizarin red, quercetin, and pyrocatechol violet were used (Figure 1b,c). DHBA and catechol caused an induction period for H_2O_2 decomposition as well (Figure 5). Evidently, the lag is due to oxidation of the original complex by H_2O_2 to a more active form, since preincubation of the complex (quercetin, catechol, and pyrocatechol violet) with H_2O_2 led to elimination of the lag and, moreover, gave rise to an accelerated rate (Figure 7). Thus, provided they remained soluble, oxidized chelates normally retained their ability to catalyze 2,4-D oxidation. In fact, some chelates became more reactive after ligand oxidation.

2,4-D Mineralization. Mineralization of ring-^{14}C-labeled 2,4-D by H_2O_2 in the presence of various complexes is shown in Figure 8a. Loss of solution radiolabel was taken to be a direct measurement of conversion to CO_2, as was demonstrated for the unchelated systems under acidic conditions (Pignatello, 1992). Mineralization usually ceased within about 4 h. However, mineralization by the citrate complex continued for several days; hydrogen peroxide was verified to persist during that period. Invariably, about 80% mineralization of 0.1 mM 2,4-D was achieved at 10 mM initial H_2O_2. Two additional supplements of 10 mM H_2O_2 gave more extensive mineralization (to 92%) in the case of the pica complex. The DHBA complex caused an induction period for 2,4-D mineralization, just as it did for 2,4-D transformation. Using the pica complex, carboxy-labeled 2,4-D was mineralized to a slightly greater extent than ring-labeled 2,4-D (Figure 8b).

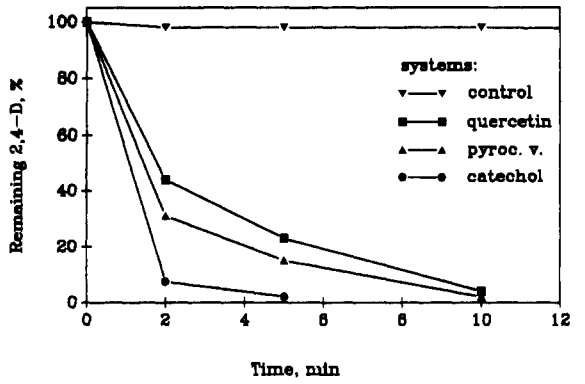

Figure 7. 2,4-D transformation by preoxidized iron(III) chelates. Chelates were incubated with 10 mM H_2O_2 for 4 h. Other conditions are the same as in Figure 1. Control is precipitated iron oxyhydroxides at pH 6.0.

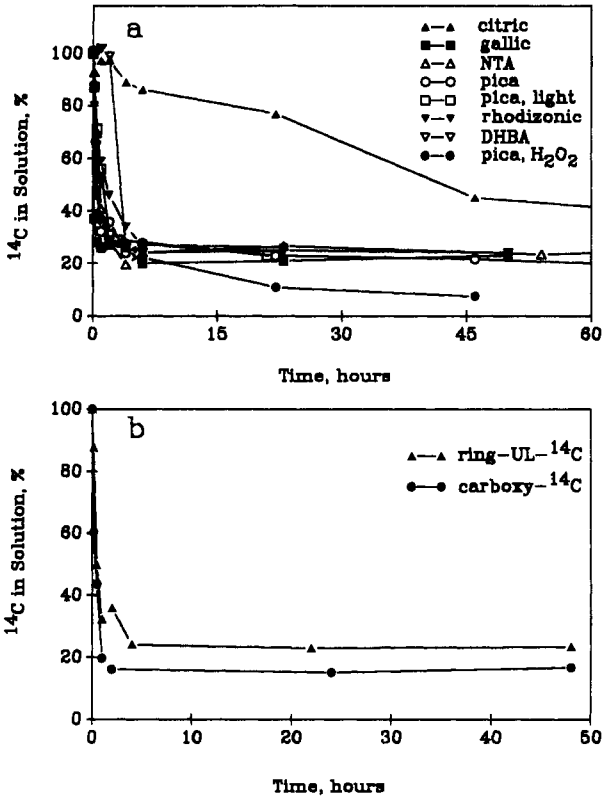

Figure 8. Mineralization of [*ring*-UL-^{14}C]- (a) and [*carboxy*-^{14}C]- (b) 2,4-D by H_2O_2 and iron(III) chelates. Conditions are the same as in Figure 1 unless stated otherwise. "pica, light" indicates pica system irradiated under conditions previously described (Pignatello, 1992); "pica, H_2O_2" presents picolinic acid system with two additional supplments of 10 mM H_2O_2 each. All pica systems used 3:1 ligand–Fe molar ratio.

In contrast to the unchelated system at pH 2.8, mineralization of 2,4-D using the gallic acid, pica, and rhodizonic acid chelates at pH 6 was insensitive to light. Figure 8a shows the pica chelate reaction with and without irradiation by visible fluorescent light containing a small UV component.

Mineralization of 2,4-D by H_2O_2 and Fe^{3+} at pH 2.8 in the presence of gallic acid, pica, or rhodizonic acid behaved much like the unchelated system at pH 2.8 in that only

about 50% mineralization occurred in 4 h (results not shown) compared to about 80% at pH 6.

DISCUSSION

Two mechanisms have been proposed to account for the powerful oxidizing ability of Fe(III)/H_2O_2 systems. The classical "radical" mechanism, abbreviated in eqs 1 and 2, generates OH•, which is one of the strongest oxidants

$$Fe^{3+} + H_2O_2 \rightleftharpoons FeOOH^{2+} \rightarrow Fe^{2+} + HO_2^{\bullet} + H^+ \quad (1)$$

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + OH^{\bullet} \quad (2)$$

known and reacts nonselectively with organic compounds (Barb et al., 1951; Walling, 1975; Walling and Cleary, 1977; Buxton et al., 1988). Equation 2 is known as the Fenton reaction.

Recently, however, there has been strong evidence for formation of high-valent iron–oxo species originating from either Fe(II) or Fe(III), especially when the iron is complexed. These species may include the ferryl ion Fe(IV)=O and the one-electron ligand oxidized ferryl ion $[L^{\bullet+}]$Fe(IV)=O (Rush and Koppennol, 1986; Rahhal and Richter, 1988; Arasasingham et al., 1989; Leising et al., 1991). It is conceivable that a combination of the above oxidants is involved here, and we will refer to them collectively as simply the "oxidant".

Most of the compounds tested (40 of 50) formed soluble Fe(III) complexes at pH 6. We did not characterize any of the complexes, since the active ones, which all contained organic ligands, were transformed in the medium. It is likely that the ligands were attacked by the oxidant. Hydroxyl radical reacts rapidly with aromatic rings, double bonds, and aliphatic C–H bonds, with rate constants as high as the diffusion-controlled limit (Buxton et al., 1988). Ferryl species also are believed to be reactive (Rush and Koppenol, 1986; Groves and Van Der Puy, 1976). Either free or complexed organic ligand may be attacked by the oxidant. Walling et al. (Walling, 1975; Walling et al., 1972) suggested that OH• generated in H_2O_2 media attacks the EDTA methylene hydrogen of Fe(III)–EDTA. Walling and Amarnath (1982) gave evidence for intramolecular attack by a solvent "caged" hydroxyl radical on mandelic acid dianion coordinated to Fe(III). The ferryl group might also be expected to intramolecularly oxidize a coordinated ligand (Groves and Van Der Puy, 1976).

From a practical standpoint, oxidation of the chelating ligand may be viewed as advantageous, provided the iron remains soluble and catalytically active long enough to carry out degradation. Destruction of the ligand may alleviate concerns about the introduction of another component into the waste. We found in several cases, moreover, that the activity of the oxidized complex was enhanced compared to that of the original complex (Figures 1 and 7).

A ligand may influence the reactivity of the complex in at least three ways: (i) by ligand field effects on the redox properties of the metal; (ii) by allowing a labile coordination position on the metal for complexation of H_2O_2 (Graf et al., 1984); and (iii) by its competition with 2,4-D for reaction with the oxidant. It is difficult to evaluate these factors due to the almost universal instability of the ligands and lack of structural and thermodynamic data on the complexes; however, some conclusions are possible. The third factor (may also include the second) is supported by the effects of ligand concentration on the reaction rate (Figure 2). The contribution of either of the first two is supported by the fact that the soluble inorganic phosphate chelates at 1:1 metal–ligand ratio were inactive, despite the comparative stability of phosphates toward oxidation by OH• (Buxton et al., 1988). Finally, the first factor is supported by the inactivity of the 1:1 catechol chelate in both 2,4-D oxidation (Figure 1b) and H_2O_2 decomposition (Figure 5) until the ligand itself was oxidized (i.e., the second factor is ruled out because catechol is bidentate, and the third factor is irrelevant to H_2O_2 decomposition).

Of the aminopolycarboxylates, NTA and HEIDA were the most reactive but were relatively unstable in the reaction media compared to many of the other compounds. The reactivity of NTA was consistent with earlier papers concerning the ability of its Fe(III) complexes to co-oxidize organic compounds by generating OH• or other oxidants from H_2O_2 (Graf et al., 1984; Singh and Hider, 1988; Hamazaki et al., 1989). However, the negligible reactivity of the EDTA and EGTA complexes contradicts previous papers (Graf et al., 1984; Winterbourn and Sutton, 1986; Walling et al., 1970). The Fe(III) ADP complex was also reportedly active (Graf et al., 1984) but was found inactive here. None of the porphyrin complexes were active, despite their known ability to form transient high-valent iron–oxo intermediates (Arasasingham et al., 1989).

With several complexes (pica, gallic acid, rhodizonic acid, NTA, HKCH, and DHBA) it was possible to achieve about 80% mineralization of 2,4-D using a relatively low concentration of peroxide. This is an improvement over unchelated Fe^{3+} in acid solution at pH 2.8, which gave only about 40% mineralization in the dark (Pignatello, 1992). However, the unchelated system was quite sensitive to light, yielding complete mineralization within 2 h on irradiation with visible light containing a small near-UV component (300-nm cutoff).

This study demonstrates the potential of several soluble organic complexes of Fe(III) to transform and extensively mineralize 2,4-D in dilute aqueous hydrogen peroxide at circumneutral pH within a few hours. The organic ligands were also oxidized, but in many cases the resulting complex containing oxidized ligand remained soluble and active. A forthcoming paper will show that three of these chelates (pica, rhodizonic acid, and gallic acid) are capable of promoting rapid oxidation of 2,4,5-T, atrazine, baygon, carbaryl, and picloram by H_2O_2. Further studies will investigate the identities of the oxidized ligands and the potential of these systems for soil remediation.

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