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Topic Review

Fenton- and Fenton-type Chemistry

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Overview

1. **Fenton's Discovery**
   - Reaction with tartaric acid
   - The Mechanism: The Controversy
   - Possible Mechanisms
2. **Fenton Chemistry in Biology and Medicine**
   - Cytochromes P-450; Oxidations
   - Interactions/Treatments
3. **Environmental Application**
4. **Gif Chemistry (Barton)**
5. **Applications**
   - Inactivated C-H oxidation
   - C-C Bond formation
6. **Fenton Chemistry with other Metals**

Fenton‘s Discovery
Henry John Horstman Fenton

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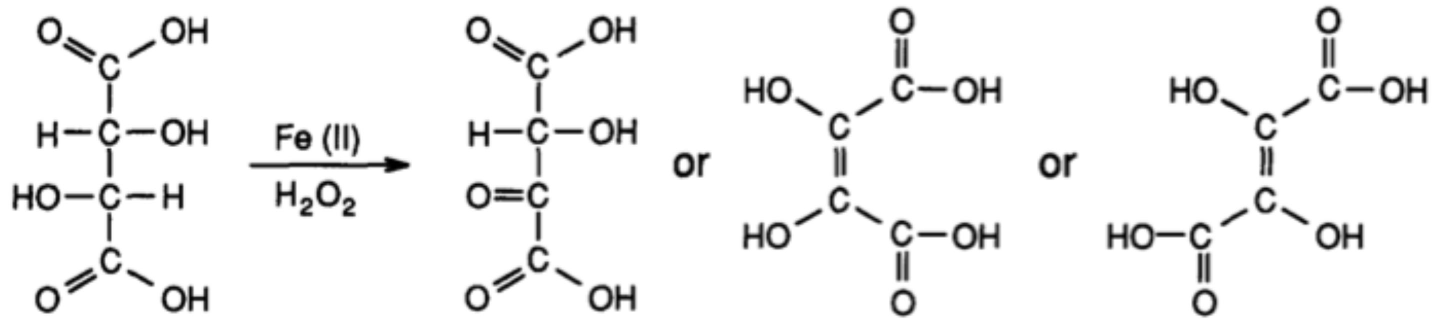
$$Fe^{2+} + HOOH \longrightarrow Fe^{3+} + {}^{-}OH + {}^{\bullet}OH$$

- Born 18th of February 1854 in Ealing, London
- Undergraduate studies at King‘s College in London
- Graduate student in group of Prof. G. D. Living (Cambridge)
- Received PhD in 1906
- Published his discovery in 1876 with title: “On a new reaction of tartaric acid”
- Discovery based on: “A fellow student was mixing reagents at random and obtained a solution with a violet colour.”
- Almost 20 years later he identified the structure of the products
- In 1894 he published “Oxidation of Tartaric Acid in Presence of Iron”

W. H. Koppenol; *Free Rad. Biol. Med.*, **1993**, *15*, 645–651
H. Fenton, *J. Chem. Soc., Trans.*, **1894**, *65*, 899–910.

Fenton's Discovery
Oxidation of Tartaric Acid

- Mixture of H_2O_2, Tartaric acid, $Fe(II)SO_4$, in water (pH 2.5 to 4)



(2*R*, 3*R*)- or L(+)-tartaric acid | 2-hydroxy-3-oxo-butanedioic acid | 2, 3-dihydroxy-maleic acid | 2, 3-dihydroxy-fumaric acid

- "[Fenton] has made the remarkable discovery that hydrogen peroxide, [...], in presence of an iron salt, at once oxidises tartaric acid and other similar acids, carbohydrates, etc., giving rise to very characteristic products – <u>a discovery of special importance in connection with plant metabolism</u>, ..."

W. H. Koppenol; *Free Rad. Biol. Med.*, **1993**, *15*, 645–651
H. Fenton, *J. Chem. Soc., Trans.*, **1894**, *65*, 899–910.

Fenton's Discovery
Fenton's Conclusions
The Controversy

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- "It seemed probable that the iron acts in a manner usually termed "catalytic""
- The oxidant may be not only hydrogen peroxide (e.g. Hypochlorous acid)
- A reduced form of a heavy metal (in this case iron) is needed, but in low concentration
- A higher oxidation state of iron may be involved as an intermediate
- Debate whether through radical intermediate or not for 80 years

Radical pathway (1932)

$Fe^{2+} + HOOH \rightarrow Fe^{3+} + {}^{-}OH + {}^{\bullet}OH$

${}^{\bullet}OH + HOOH \longrightarrow H_2O + {}^{\bullet}OOH$

${}^{\bullet}OOH + Fe^{3+} \longrightarrow Fe^{2+} + O_2 + H^+$

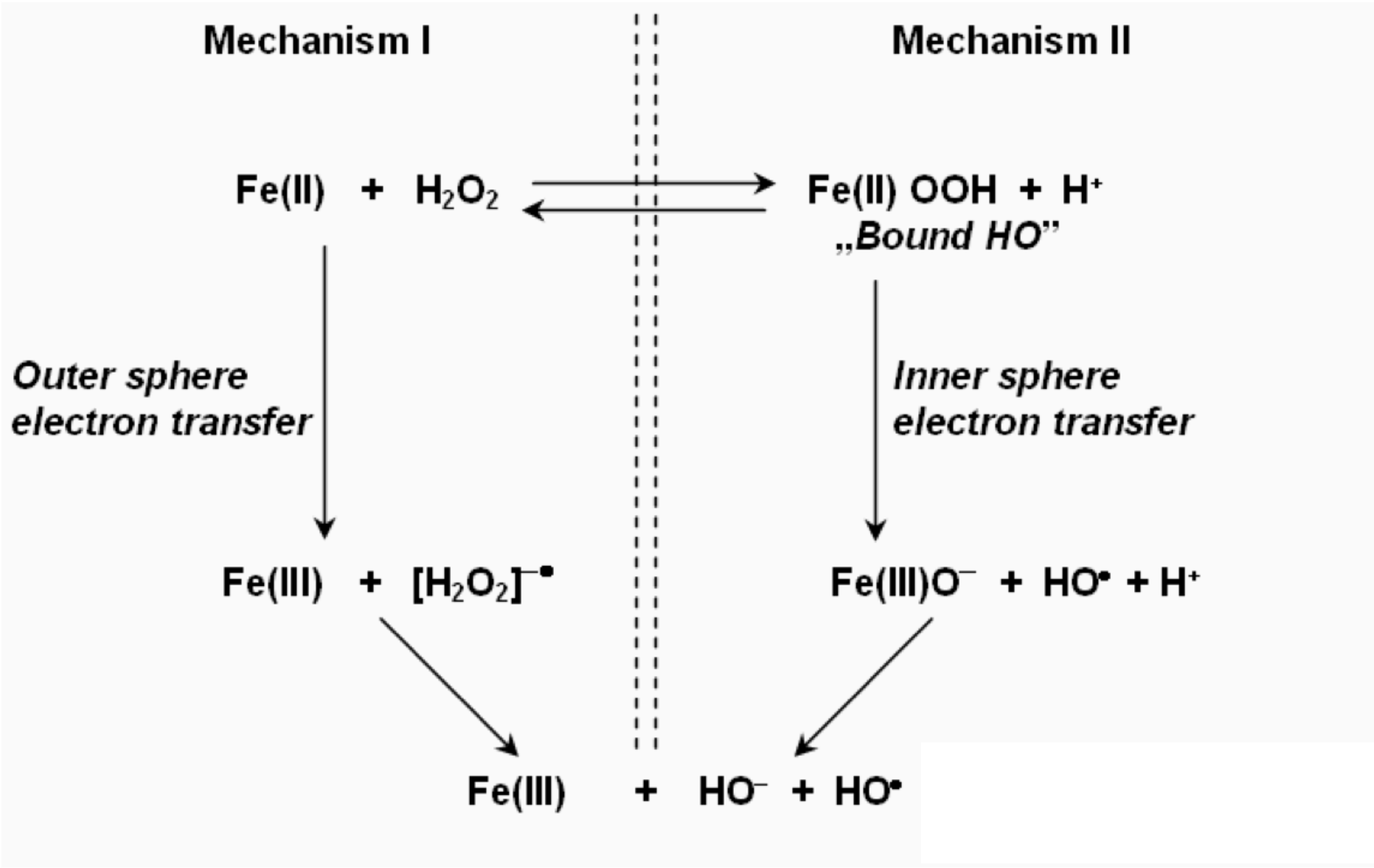
Non-Radical pathway (1934)

$Fe^{2+} + HOOH \longrightarrow FeO^{2+} + H_2O$

$FeO^{2+} + HOOH \longrightarrow Fe^{2+} + H_2O + O_2$

H. B. Dunford, *Coord. Chem. Rev.*, **2002**, *233–234*, 311–318
W. H. Koppenol; *Free Rad. Biol. Med.*, **1993**, *15*, 645–651
H. Fenton, *J. Chem. Soc., Trans.*, **1894**, *65*, 899–910.

Possible Mechanism I
Free Radical Pathway (Fe^{II})



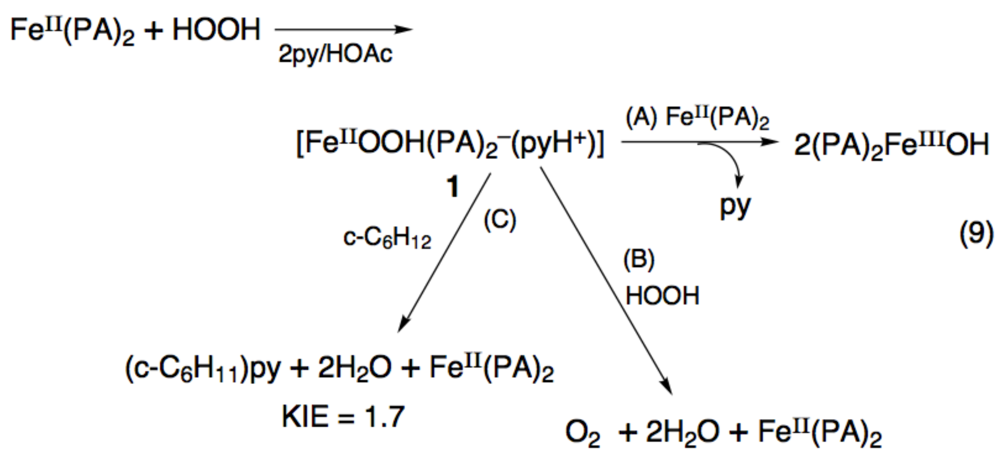
K. Barbusinski, *Ecol. Chem. Engin.*, **2009**, *16*, 347–358

Possible Mechanism II

Sawyer's Paper

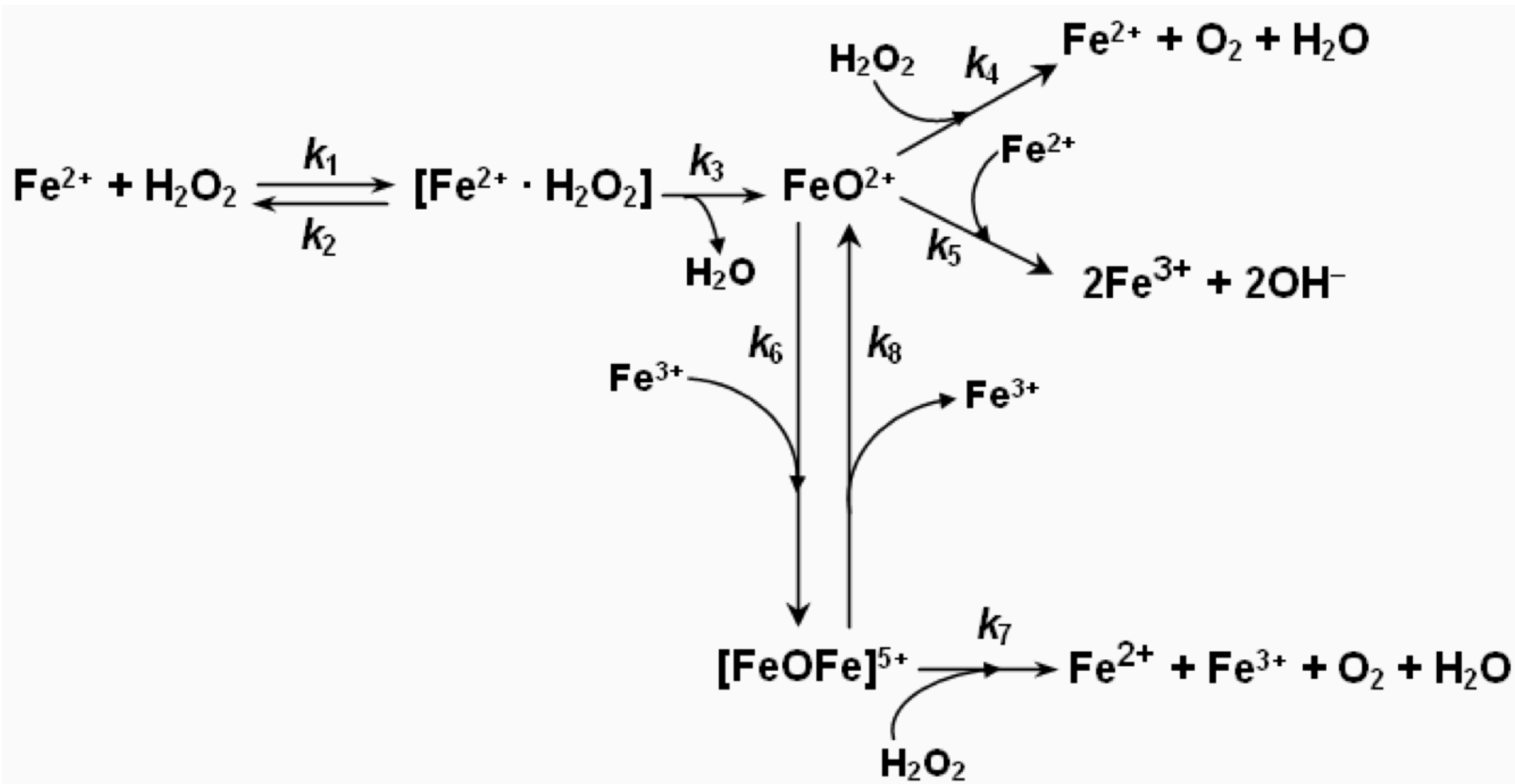
"Fenton Reagents *do not produce* •OH or R•"

- Primary activity of peroxides is nucleophilic, centred at H–OOR bond
- Dialkyl peroxides in contrast to hydroperoxides are unreactive with electrophilic substrates (e.g. SO_2, $FeCl_2$, *n*-BuI, and HOCl)
- Anhydrous HOOH in base-free media (e.g. MeCN) is unreactive
- Although •OH reacts with CH_4 ($k = 1.1 \times 10^8\ M^{-1}s^{-1}$), Fenton reagent does not
- Although radical traps (e.g. $(PhSe)_2$, $BrCCl_3$, Me_2SO) often used to prove free carbon radicals (•R), these reacts also with non-radicals



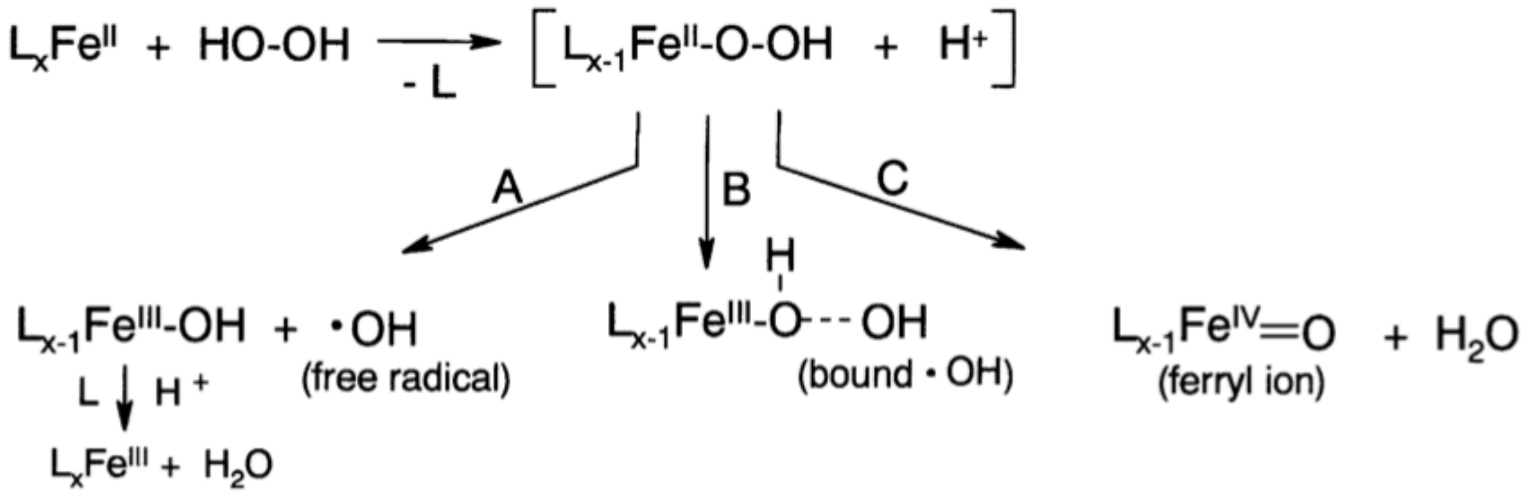
D. T. Sawyer, A. Sobkowiak, T. Matsushita, *Acc. Chem. Res.* **1996**, *29*, 409–416

Possible Mechanism III
Non-Radical Pathway (Fe^{II})



M. L. Kremer, *Phys. Chem. Chem. Phys.*, **1999**, *1*, 3595–3605

Possible Mechanism IV
The Dualism: Fe(II)

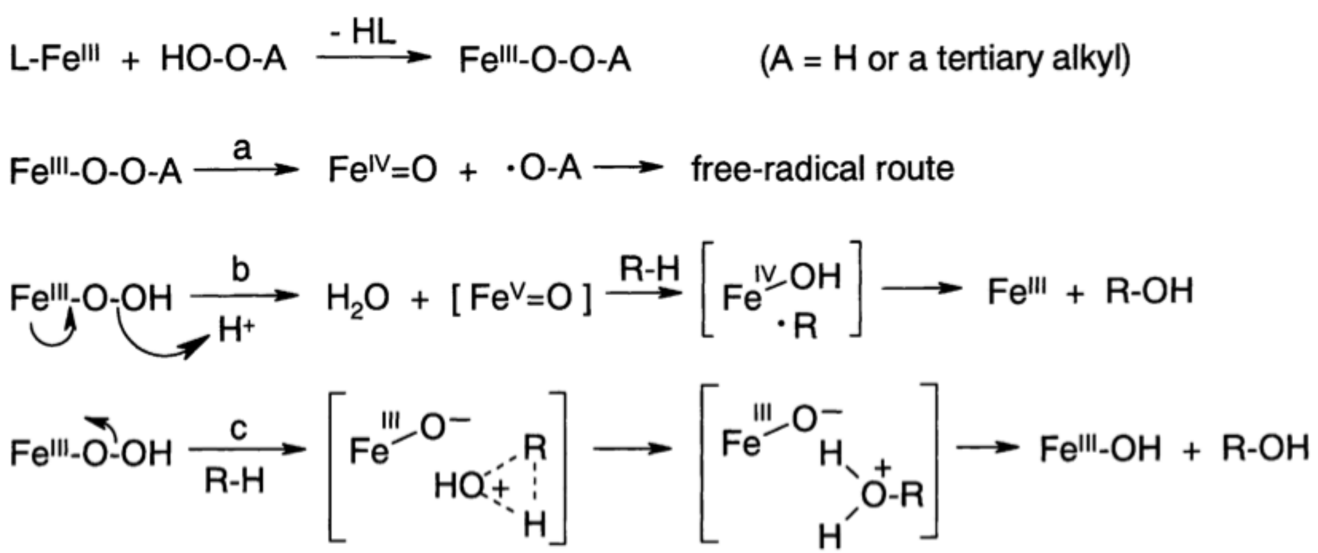


- **Pathway A**: Traditional, described by Fenton; in presence of saturated hydrocarbons; acidic solutions (pH 2)
- **Pathway B**: Aqueous media; strong chelators (e.g. EDTA, DETAPAC) or phosphate ions; Spin trapping showed dependence on the ligand
- **Pathway C**: Typically produced by ferro-porphyrines

F. Gozzo, *J. Mol. Cat. A. Chem.* **2001**, *171*, 1–22
I. Yamazaki, L. H. Piette, *J. Am. Chem. Soc.*, **1991**, *113*, 7588

Possible Mechanism V

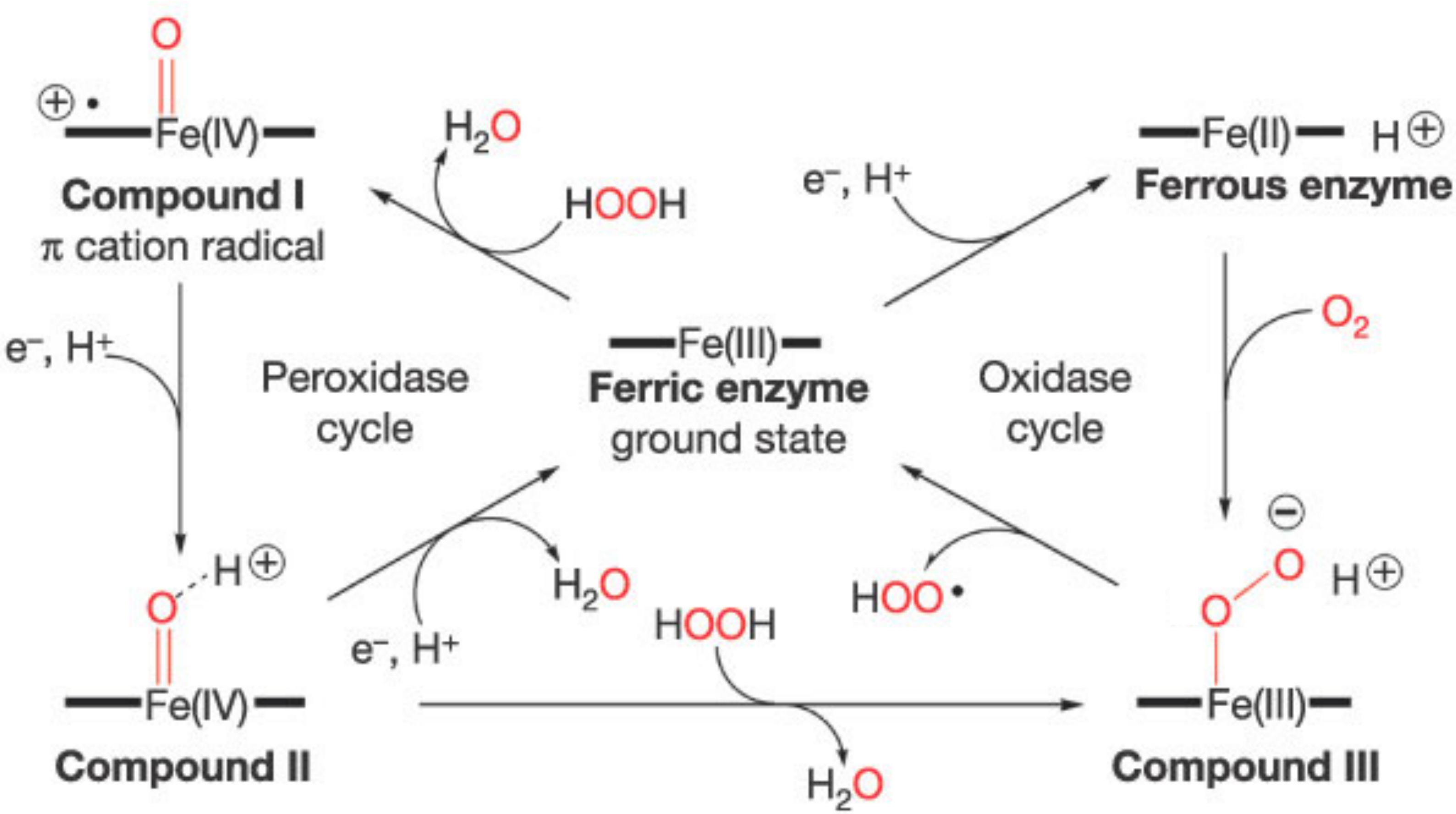
The Dualism: Fe(III)



- First step (Ligand displacement) generally accepted
- **Pathway a**: Free-radical route
- **Pathway b**: Oxenoid intermediate; “bound” alkyl radical
- **Pathway c**: Incipient species (^{+}OH)

F. Gozzo, *J. Mol. Cat. A. Chem.* **2001**, *171*, 1–22

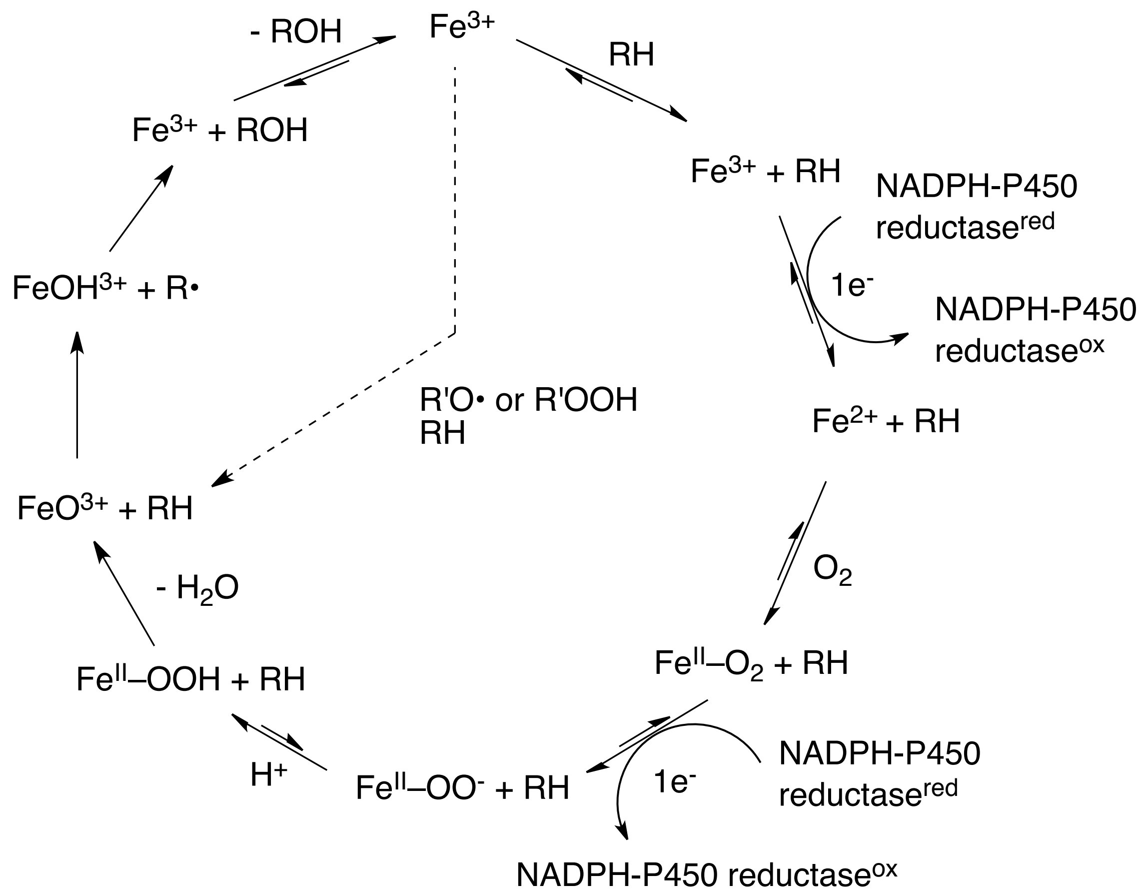
Fenton Chemistry in Biology and Medicine
Oxidase/Peroxidase
General Pathway



G. I. Berglund, G. H. Carlsson, A. T. Smith, H. Szöke, A. Henriksen, J. Hajdu, *Nature,* **2002**, *417*, 463–468

Fenton Chemistry in Biology and Medicine
Cytochromes P-450

- Family of monoxygenases
- Containing a heme cofactor (hemoproteins)
- Found in all domains of life (NOT *E. coli*)
- Oxidation of wide variety of drugs, carcinogens, steroids, pesticides and others



F. P. Guengerich, *J. Biochem. Mol. Tox.*, **2007**, *21*, 163–168

Fenton Chemistry in Biology and Medicine

Cytochromes P-450; Oxidations with FeO^{3+}

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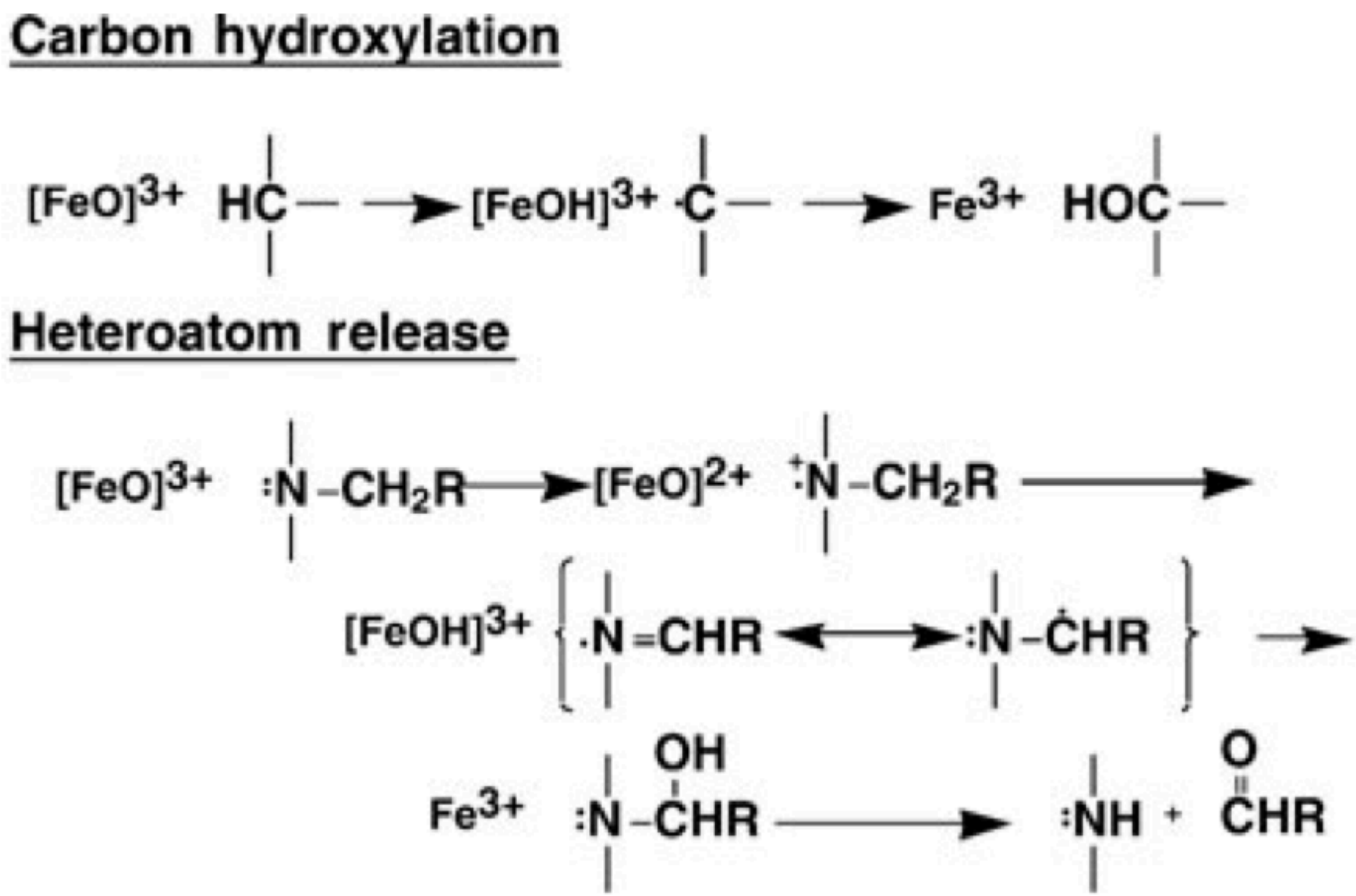
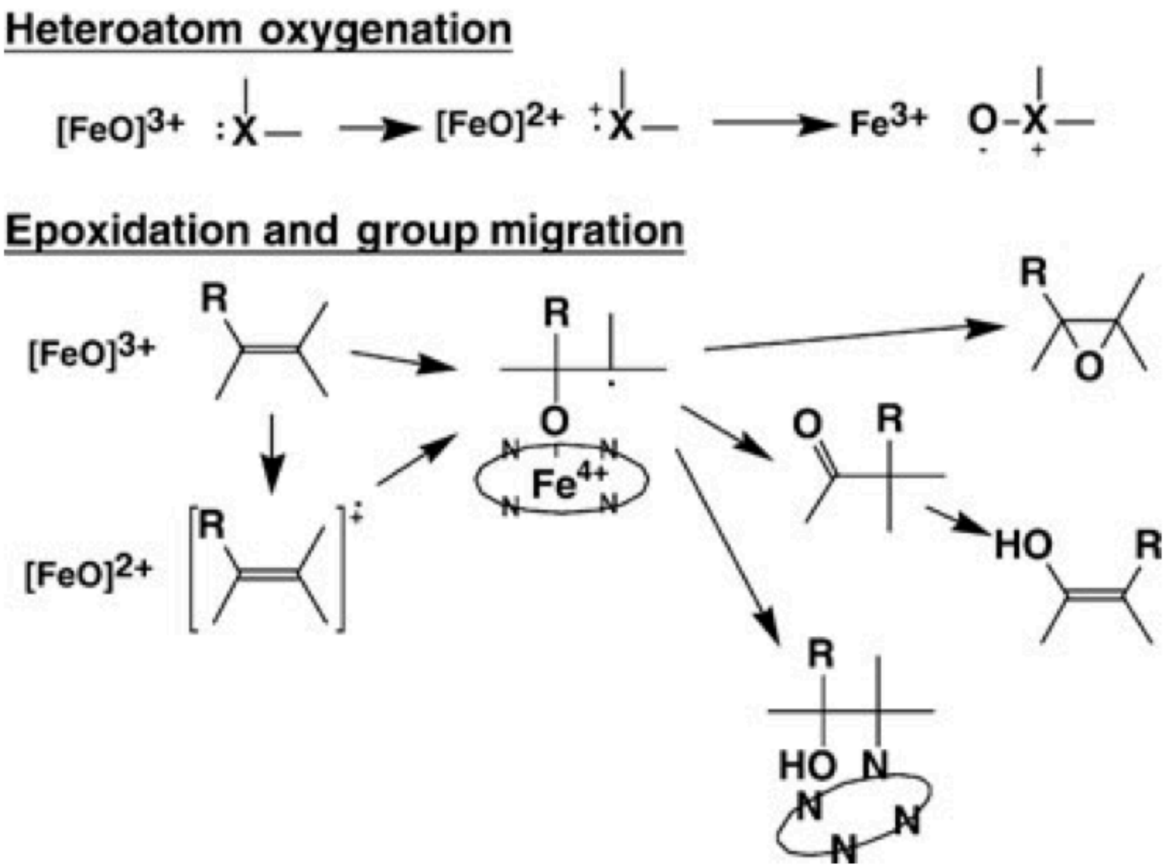


F. P. Guengerich, *J. Biochem. Mol. Tox.*, **2007**, *21*, 163–168

Fenton Chemistry in Biology and Medicine

Cytochromes P-450; Oxidations with FeO^{3+}



F. P. Guengerich, *J. Biochem. Mol. Tox.*, **2007**, *21*, 163–168

Fenton Chemistry in Biology and Medicine

Cytochromes P-450; Examples of oxidations with FeO^{3+}

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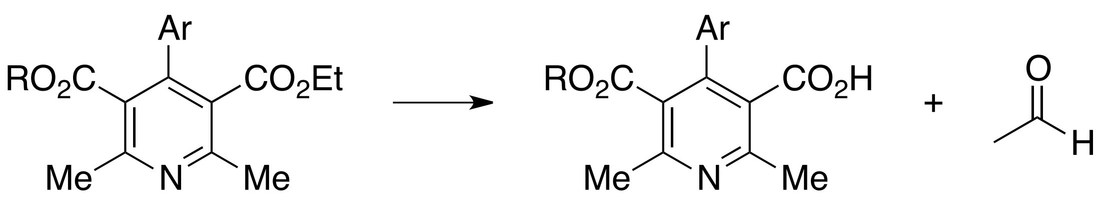
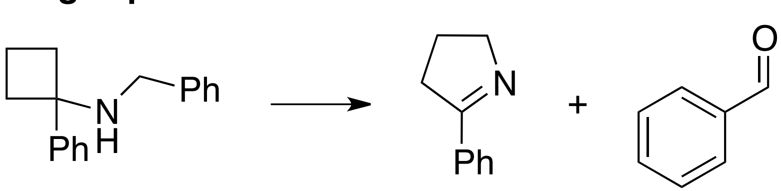
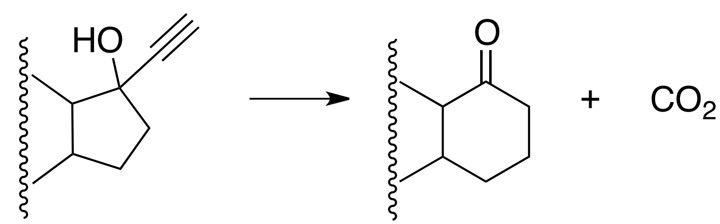
Oxidative ester cleavage

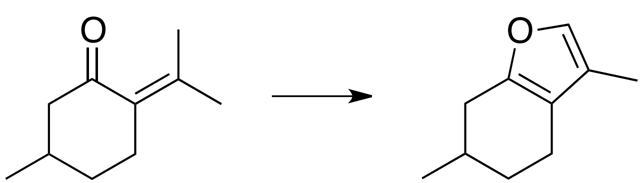


Ring expansion

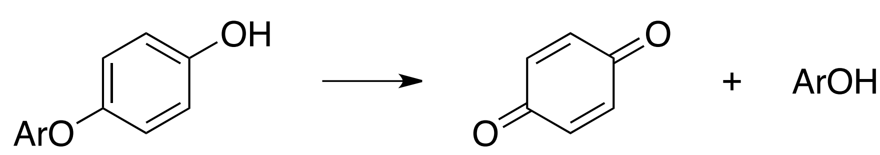




Ring formation



Dearylation

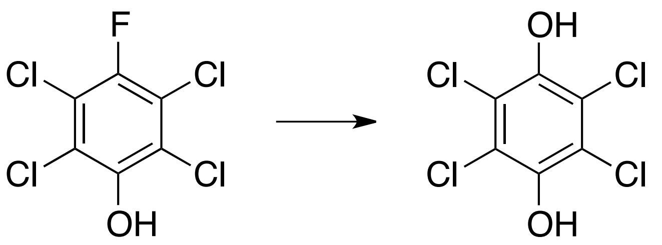


F. P. Guengerich, *J. Biochem. Mol. Tox.*, **2007**, *21*, 163–168
F. P. Guengerich, *Chem. Res. Tox.*, **2001**, *14*, 611–650

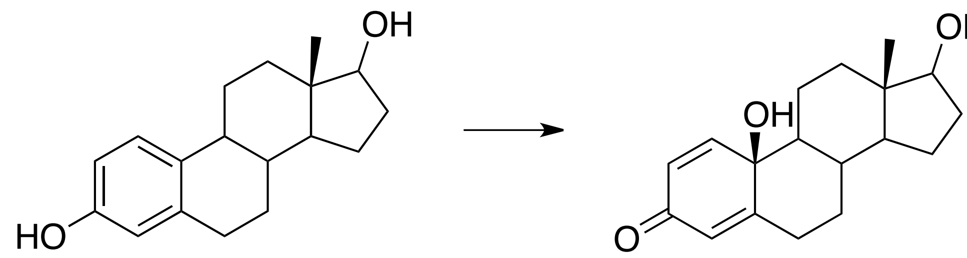
Fenton Chemistry in Biology and Medicine

Cytochromes P-450; Examples of oxidations with FeO^{3+}

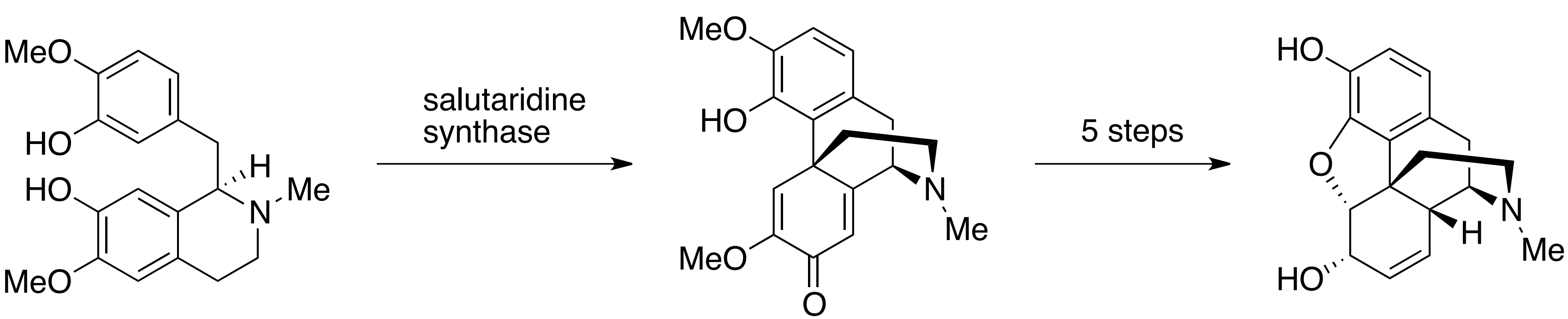
Aromatic dehalogenation via *ipso* attack



Dearomatisation and formation of a Michael acceptor



Oxidative ring coupling

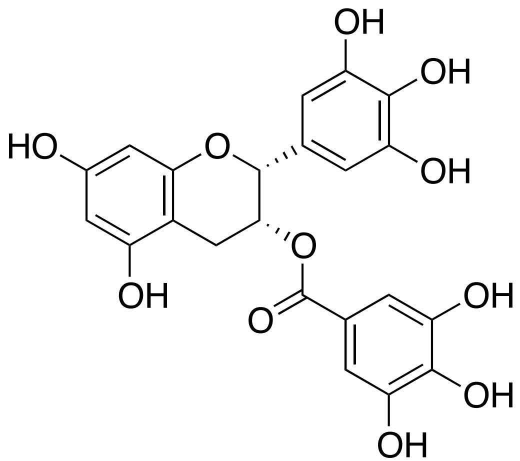


(*R*)-reticuline salutaridine (–)-morphine

F. P. Guengerich, *J. Biochem. Mol. Tox.*, **2007**, *21*, 163–168
F. P. Guengerich, *Chem. Res. Tox.*, **2001**, *14*, 611–650

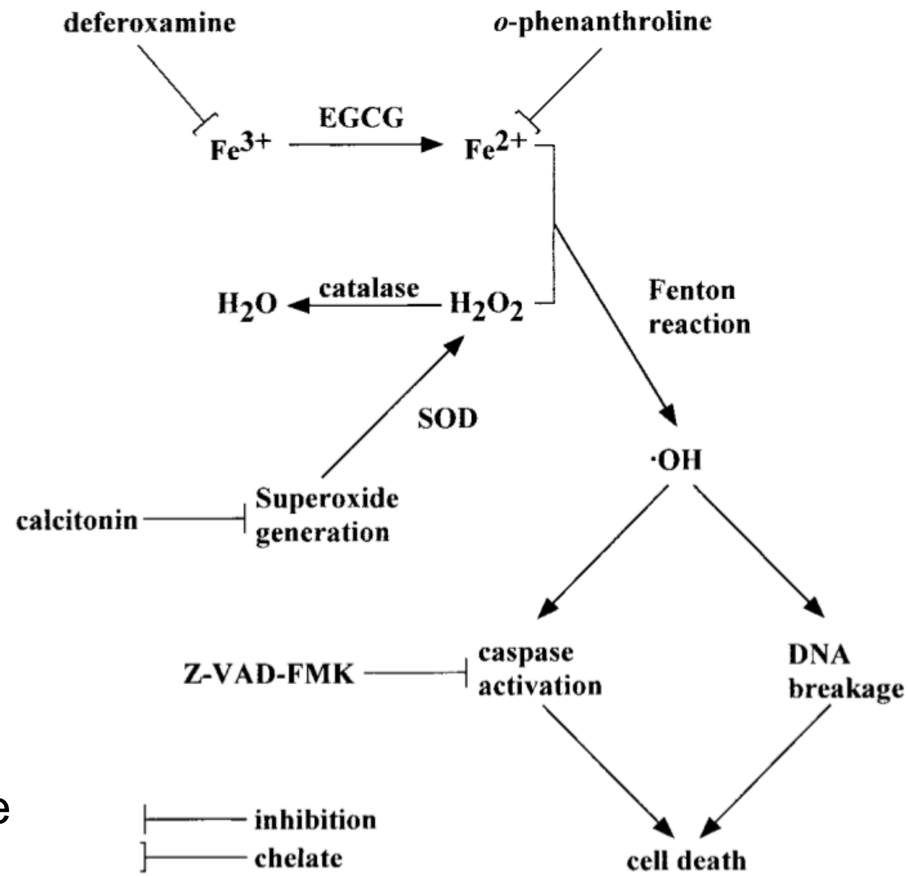
Fenton Chemistry in Biology and Medicine

Interactions; EGCG
Prevention/Treatment for Osteoporosis



Epigallocatechin gallate (EGCG)

- Found in (green) tea
- Potent antioxidant
- Class of flavonoids
- Cancer treatment and osteoporosis (triggers apoptosis in osteoclast like cells, osteoblasts not affected)



H. Nakagawa, M. Wachi, J.-T. Woo, M. Kato, S. Kasai, F. Takahashi, I.-S. Lee, K. Nagai, *Biochem. Biophys. Res. Comm.* **2002**, *292*, 94–101.

Fenton Chemistry in Ecology

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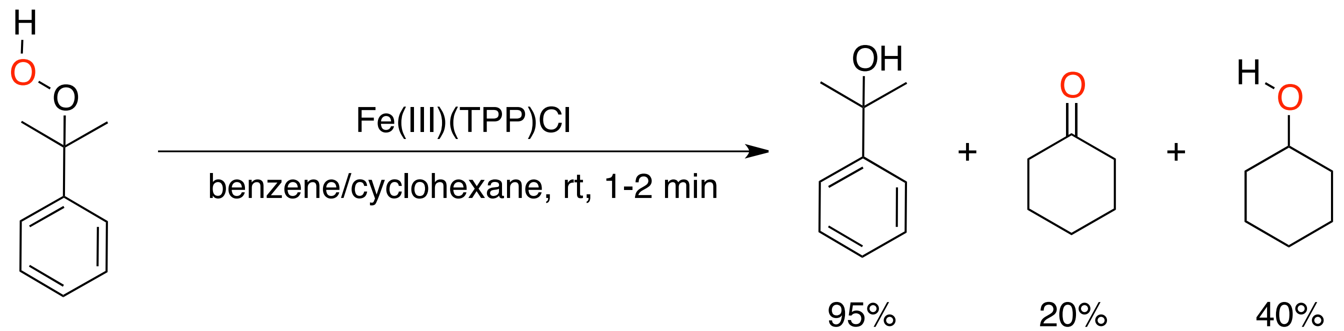
- The hydroxyl radicals could react rapidly and non-selectively with nearly all organic pollutants
- Waste-Water Treatment (Reducing Toxicity)
  - Aromatic amines
  - Dyes
  - Pesticides
  - Surfactants
  - Oxidation of As(III) to As(V)
  - ...
- Toxicity level is understood by the inhibited amount (%) of vital function of bioluminescent bacteria *Vibrio fischeri*

K. Barbusinski, *Pol. J. Environ. Stud.,* **2005**, *14*, 11–16

Fenton Chemistry in Organic Chemistry
Oxidation of inactivated C-H bonds

- Alkane oxidation under mild conditions (miming Cytochrome P-450)

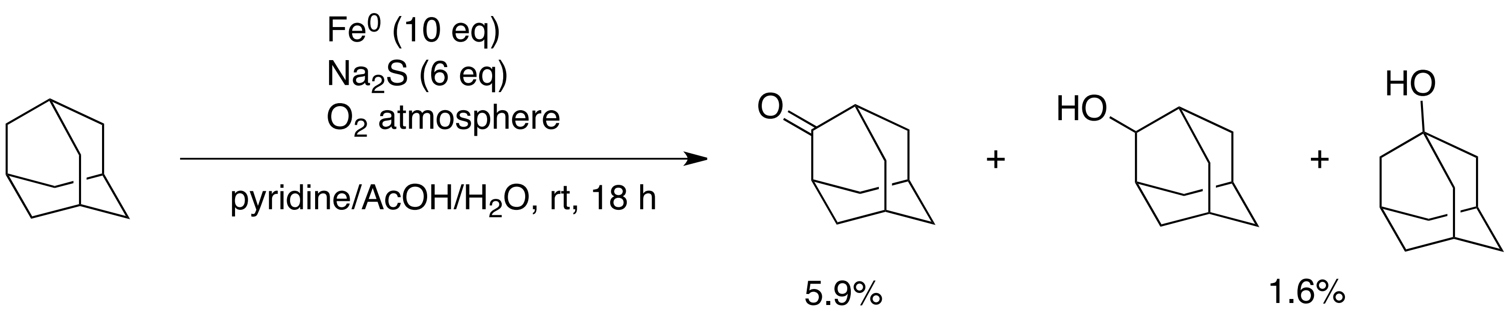


- Reaction works in aerobic and anaerobic conditions (slightly faster)
- Catalyst fully recovered (TPP = *tetra*-phenyl porphyrin)
- No reaction with only $TPPH_2$, $FeCl_3$ or $FeCl_2$
- Similar reactivity with *tert*-butyl hydroperoxide

D. Mansuy, J. F. Bartoli, J. C. Chottard, M. Lange, *Angew. Chem. Int. Ed.,* **1980**, *19*, 909–910

Fenton Chemistry in Organic Chemistry
Gif Chemistry (Barton)
Origin

- Considering pre-aerobic atmosphere
- Simpler mechanism for oxygen "detoxification" (non heme)



- Similar yield and selectivity with H_2S (Gif II) and FeS
- Statement: "Clearly our oxidation process is not in radical nature."
- Screening of carboxylic acids, sulphide compounds, solvents
- Temperature increase replaces sulfide

D. H. Barton, M. J. Gastiger, W. B. Motherwell, *J. Chem. Soc. Chem. Comm.,* **1983**, 41–43

Gif Chemistry

Free Radicals/Influence of Oxygen-Atmosphere

5) $Fe^{III} + R\text{-}O\text{-}OH \rightarrow Fe^{IV}{=}O + H^{+} + R\text{-}O\bullet$

6) $Fe^{IV}{=}O + R\text{-}O\text{-}OH \rightarrow Fe^{III} + OH^{-} + R\text{-}O\text{-}O\bullet$

Reaction	k (R = H)	k (R = *tert*-Bu)
7) $R\text{-}O\bullet + {>}CH_2 \rightarrow R\text{-}OH + {>}CH\bullet$	$> 10^{9}\ M^{-1}s^{-1}$	$> 10^{6}\ M^{-1}\ s^{-1}$
8) $R\text{-}O\bullet + R\text{-}O\text{-}OH \rightarrow R\text{-}OH + R\text{-}O\text{-}O\bullet$	$1\text{-}4.5\cdot10^{7}\ M^{-1}s^{-1}$	$8.7\cdot10^{6}\ M^{-1}\ s^{-1}$

9) $2\ R\text{-}O\text{-}O\bullet \rightarrow 2\ R\text{-}O\bullet + O_2$

10) ${>}CH\bullet + O_2 \rightarrow {>}CH\text{-}O\text{-}O\bullet$ $\quad k = 5\cdot10^{9}\ M^{-1}\ s^{-1}$

11) $2\ {>}CH\text{-}O\text{-}O\bullet \rightarrow {>}C{=}O + {>}CH\text{-}OH + O_2$

12) ${>}CH\bullet + R\text{-}O\text{-}O\bullet \rightarrow {>}CH\text{-}O\text{-}O\text{-}R \rightarrow {>}C{=}O + R\text{-}OH$

13) ${>}CH\text{-}O\text{-}O\bullet + R\text{-}O\text{-}O\bullet \rightarrow O_2 + {>}CH\text{-}O\text{-}O\text{-}R \rightarrow {>}C{=}O + R\text{-}OH$

F. Gozzo, *J. Mol. Catal. A,* **2001**, *171*, 1–22

Gif Chemistry
Nomenclature

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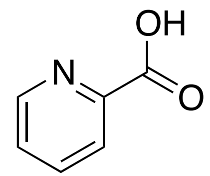
- Gif chemistry permits the conversion of saturated hydrocarbons into ketones at rt under nearly neutral conditions

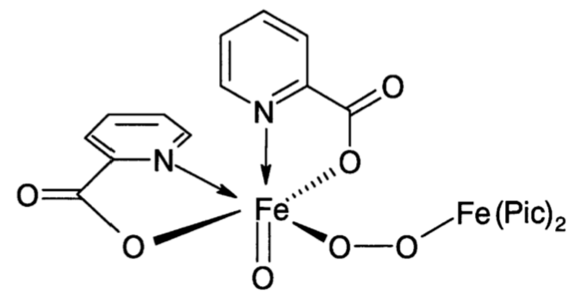
Table I. The Nomenclature of Gif Chemistry[a]

system	catalyst	electron source	oxidant
Gif^{III}	Fe(II)	Fe^0	O_2
Gif^{IV}	Fe(II)	Zn^0	O_2
GO	Fe(II)	cathode	O_2
$GoAgg^{I}$	Fe(II)	KO_2/Ar	
$GoAgg^{II}$	Fe(III)	H_2O_2	
$GoAgg^{III}$	Fe(III)/picolinic acid	H_2O_2	
GoChAgg	Cu(II)	H_2O_2	
Cu^0/O_2	Cu(I)?	Cu^0	O_2

[a] The nomenclature of the Gif systems is geographically based: G stands for Gif-sur-Yvette, O is for Orsay, Agg is for Aggieland, Texas A&M, and Ch is for Chernogolovka, Russia.



picolinic acid



- $GoAgg^{IV}$ TBHP and $GoAgg^{V}$ is ($GoAgg^{IV}$ + picolinic acid)

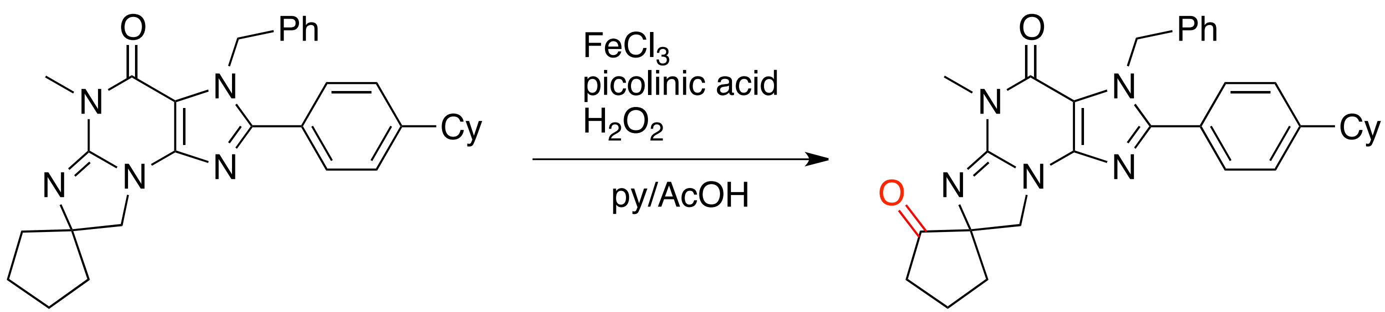
D. H. Barton, D. Doller, *Acc. Chem. Res.* **1992**, *25*, 504–512.

Gif Chemistry

Conclusions

Oxidation of a c-GMP phosphodiesterase inhibitor

- Studies concerning oxidation of various saturated hydrocarbons; but always very low conversion (typically below 10%)
- Oxidation takes place in presence of H_2S, Ph_2S, PPh_3, $P(OMe)_3$, PhSH, and PhSeH (normally easier to oxidise)
- "Iron species seems to be only activated in presence with C-H bond" (Barton) → Fe=C bond proposition
- $Fe^{V}=O$ preferentially inserted into secondary positions



SCH 57726

2% (95% sm recovery)

D. H. R. Barton, *Tetrahedron,* **1998**, *54*, 5805–5817

D. Doller, S. Chackalamannil, A. Stamford, B. McKittrick, M. Czarniecki, *Bioorg. Med. Chem. Lett.* **1997**, *7*, 1381–1386.

Applications of Fenton Chemistry

C-H bond oxidation

Benzylic oxidation and Oxidation of benzylic compounds

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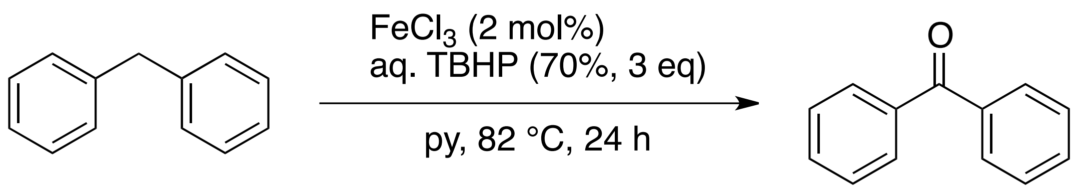
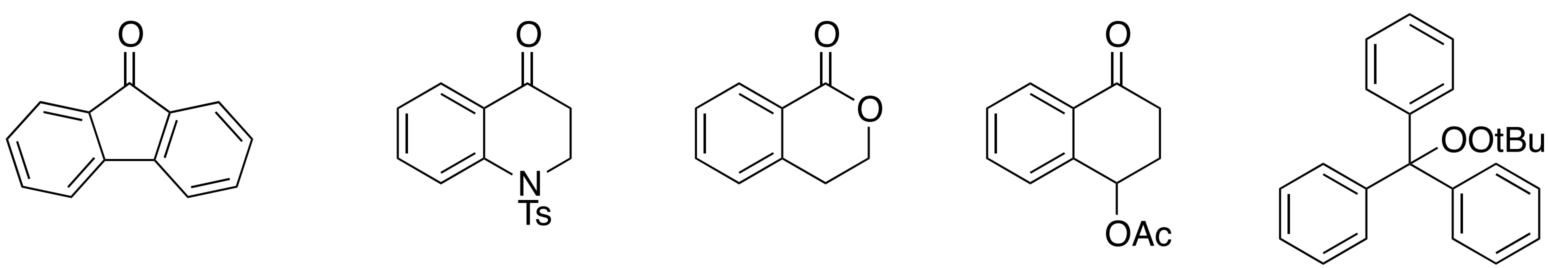


91%

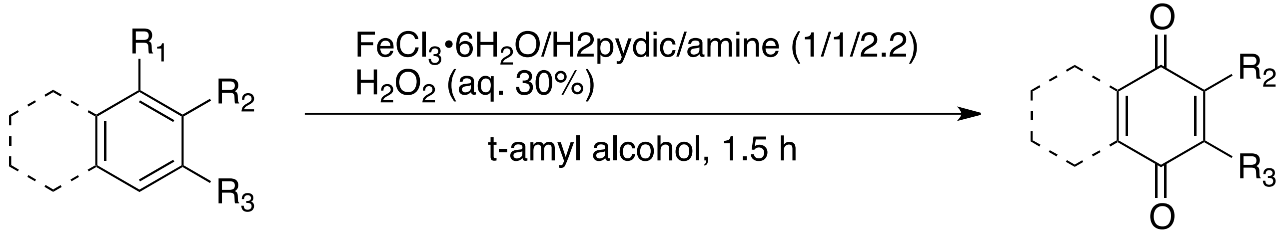
- Iron(III) chloride inexpensive; non-toxic; small quantities
- No ligand/acid needed (probably pyridine as coordinating agent)
- Slow addition is unnecessary



>99% 60% 74% 66% 91%

M. Nakanishi, C. Bolm, *Adv. Synth. Catal.* **2007**, *349*, 861 – 864

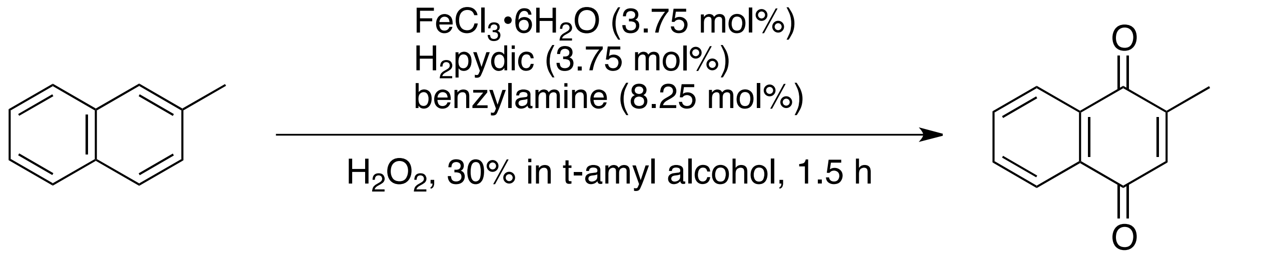
Applications of Fenton Chemistry
C-H bond oxidation
Oxidation of phenols/arenes



23-78%

R^1 = H or OH
R^2 = H or Me or OMe
R^3 = H or Me

H2pydic = pyridine-2,6-dicarboxylic acid
t-amyl alcohol = 2-methyl-2-butanol
amine = *N*-butylbenzylamine, benzylamine

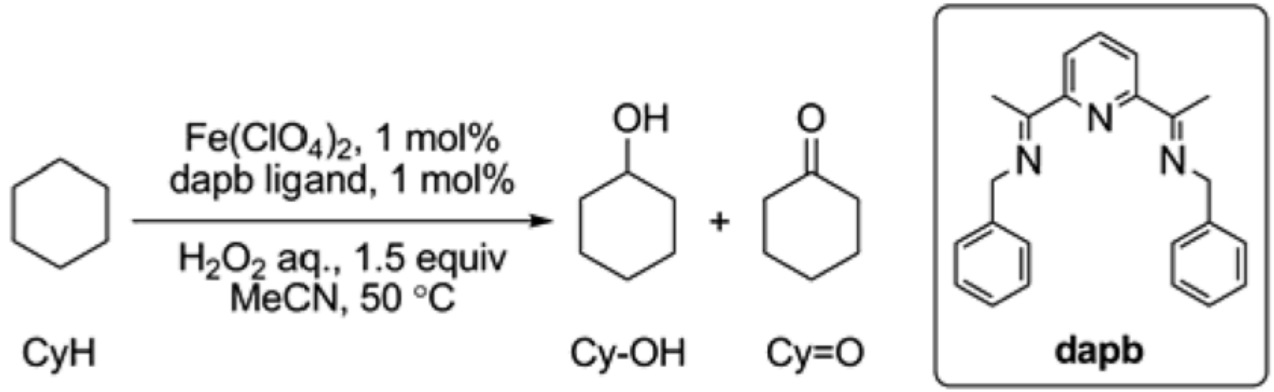


menadione (Vitamin K_3)
55%

K. Möller, G. Wienhöfer, K. Schröder, B. Join, K. Junge, M. Beller, *Chem. Eur. J.*, **2010**, *16*, 10300–10303

Applications of Fenton Chemistry

C-H bond oxidation
Ligands (Non-heme)

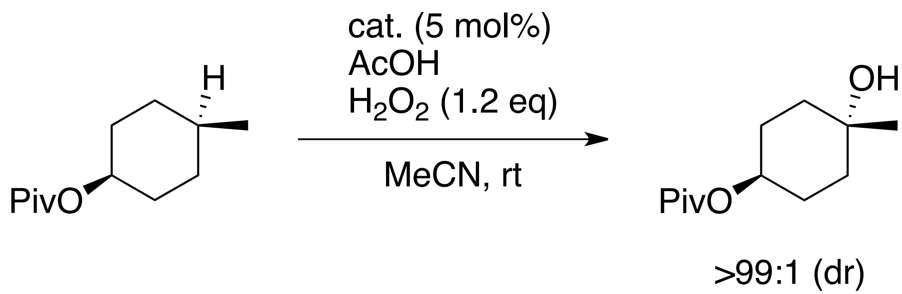


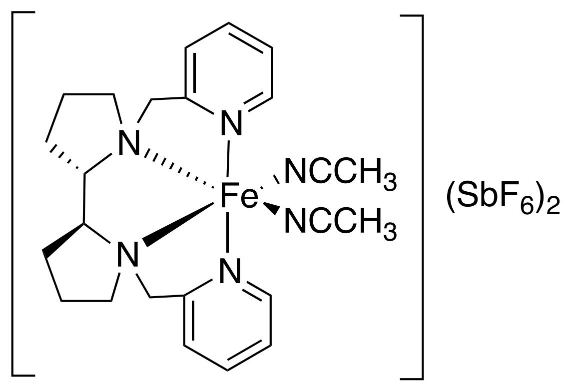
entry	catalyst	major products	desired product	by-products
1	with ligand	Cy-OH (37):Cy=O (54)	91%	9%
2	no ligand	Cy-OH (27):Cy=O (60)	87%	13%

- Oxidation occuring rapidly
- Counter-Anion did not play role in efficacy

B. Retcher, J. S. Costa, J. Tang, R. Hage, P. Gamez, J. Reedijk, *J. Mol. Catal. A*, **2008**, *286, 1–5*

Applications of Fenton Chemistry
C-H bond oxidation
Ligands (Non-heme); Selective oxidation I





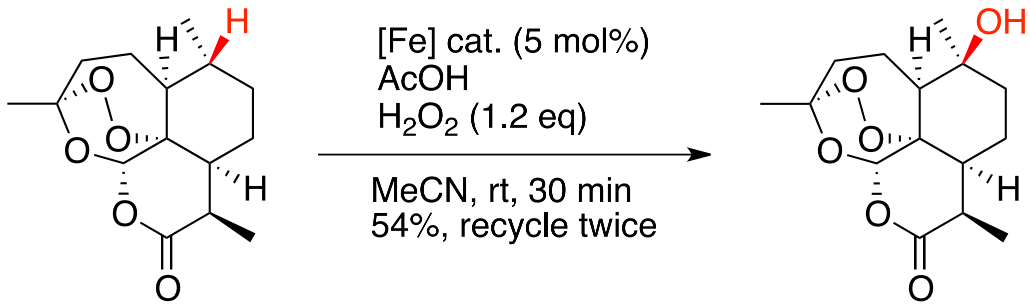
- Clean process (42% conversion of sm)
- Electronic feature more important than steric (CH more reactive than CH_2/CH_3)

M. S. Chen, M. C. White, *Science*, **2007**, *318*, 783

Applications of Fenton Chemistry

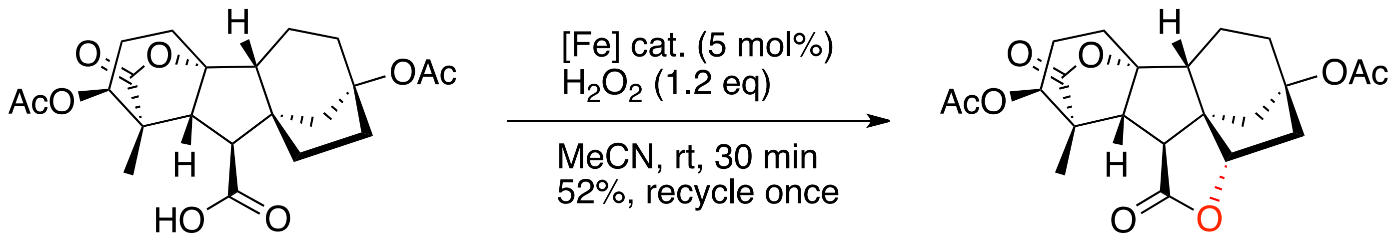
C-H bond oxidation

Ligands (Non-heme); Selective oxidation II



(+)-artemisinin

- Enzymatic reaction (*C. echinulata* = 4 days, 41%)



- Carboxylic acid moiety directs oxidation
- Slow addition of [Fe] → higher conversion no recycling

M. S. Chen, M. C. White, *Science*, **2007**, *318*, 783

Applications of Fenton Chemistry
C-H bond oxidation
Ligands (Non-heme); Selective oxidation III

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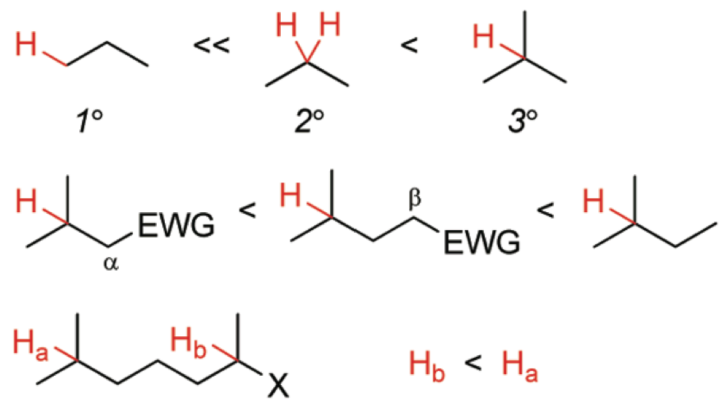
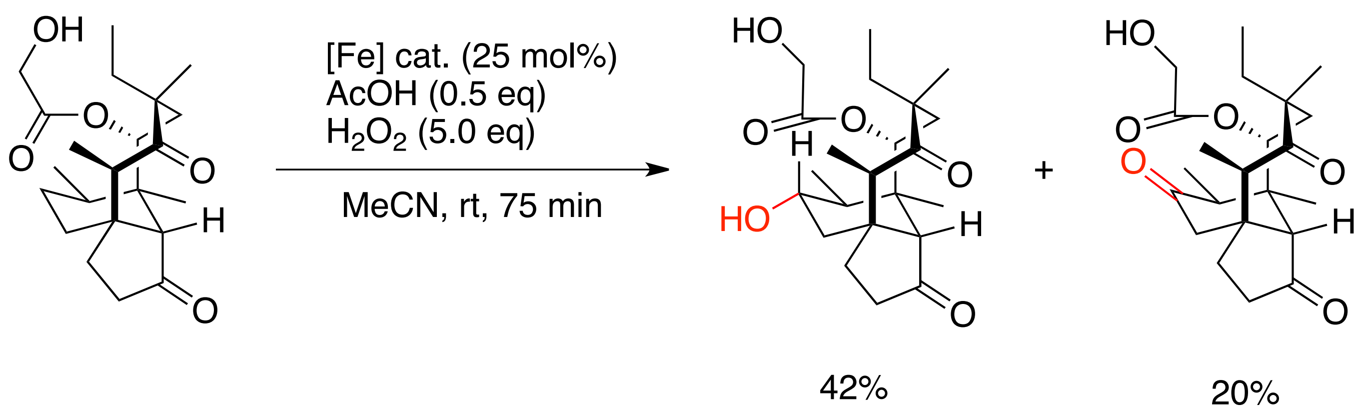


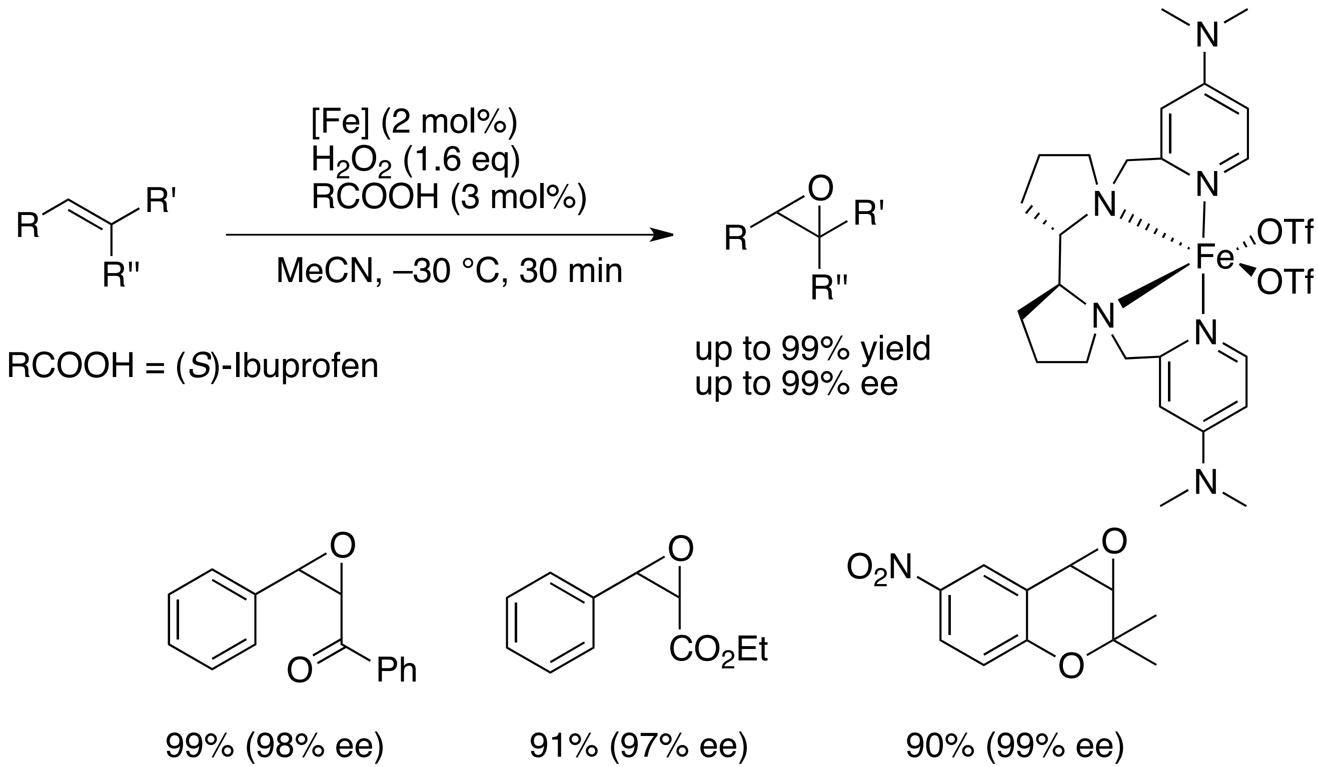
X = COMe, COOMe, CH_2Br, CH_2OAc,

- Reactivity also promoted by:
  - electronic
  - steric
  - stereoelectronic
  - functional group factors



M. S. Chen, M. C. White, *Science*, **2010**, *327*, 566

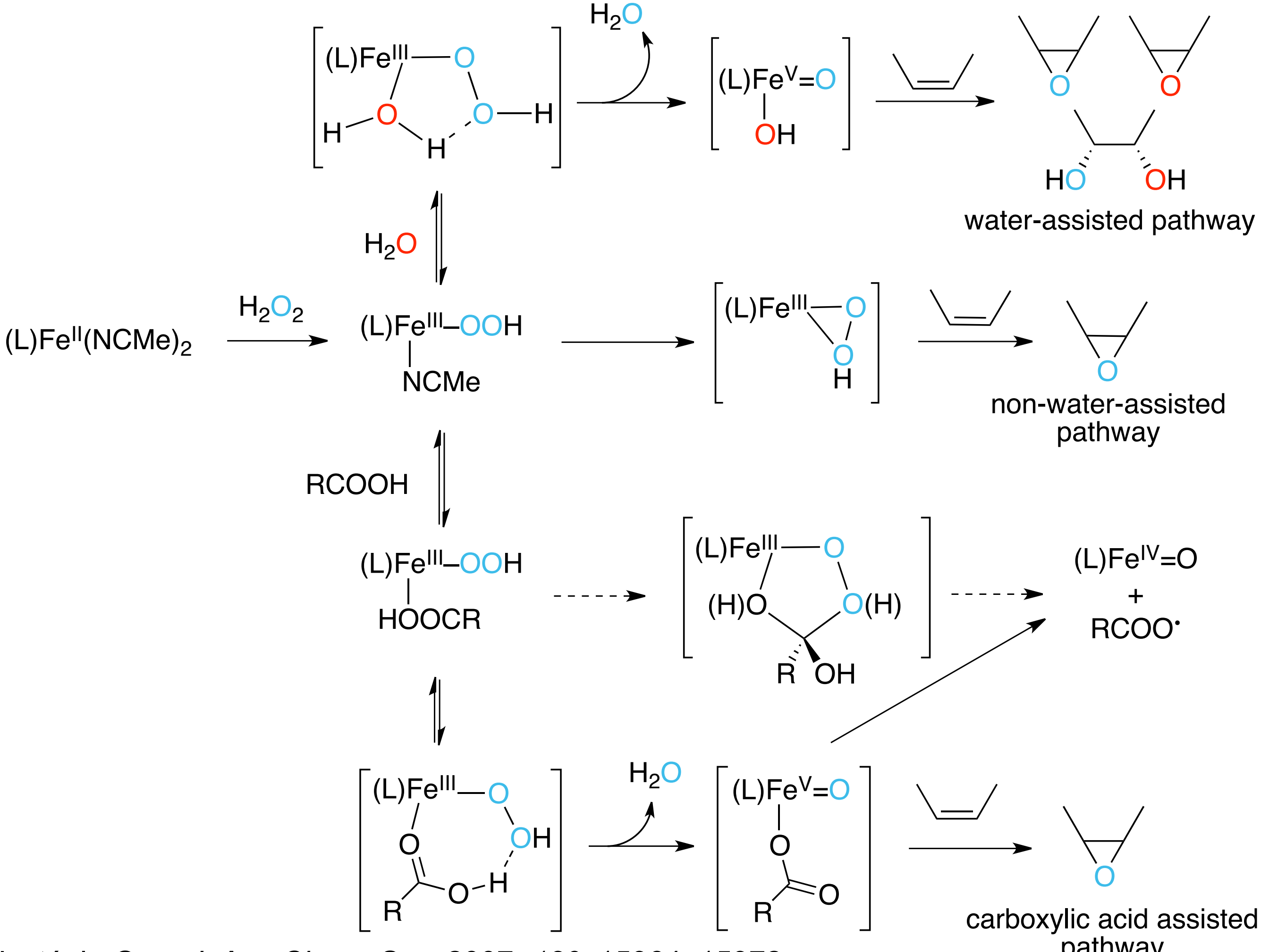
Applications of Fenton Chemistry
C=C bond oxidation
Ligands (Non-heme); Selective oxidation IV



- Using $H_2{}^{18}O$ and $H_2{}^{18}O_2$ for mechanistic analysis
- Ressemblance to "push-pull" effect in P-450

O. Cussó, I. Garcia-Bosch, X. Ribas, J. Lloret-Fillol, M. Costas, *J. Am. Chem. Soc.* **2013**, *135*, 14871–14878

Applications of Fenton Chemistry
C=C bond oxidation, Mechanism
Ligands (Non-heme); Selective oxidation IV

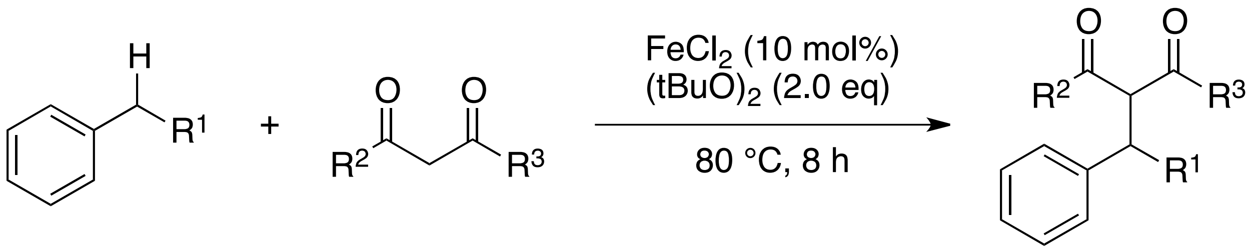


R. Mas-Ballesté, L. Que, *J. Am. Chem. Soc.* **2007**, *129*, 15964–15972

Applications of Fenton Chemistry

C-C bond formation

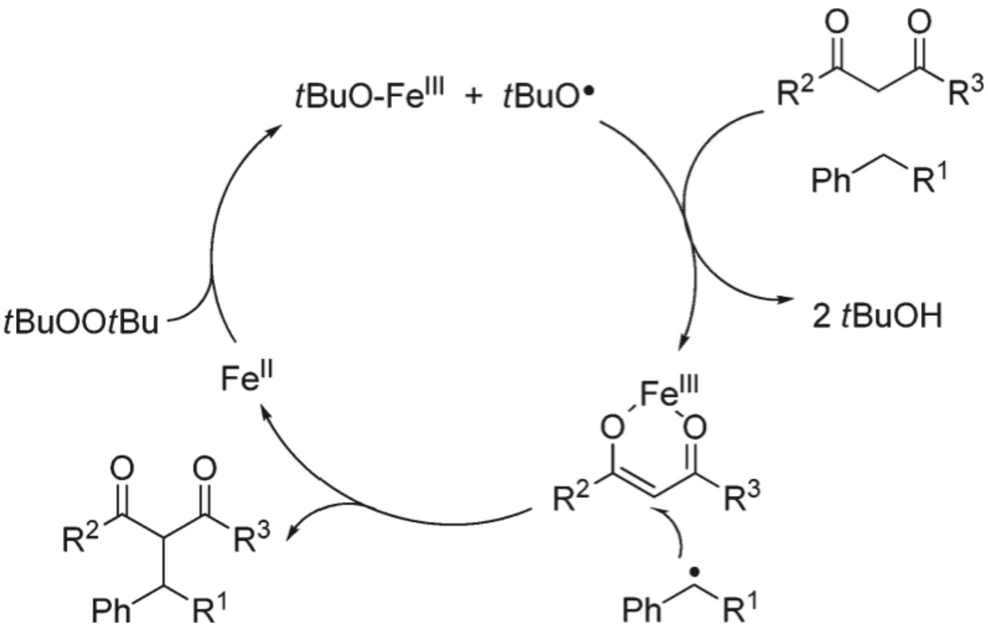
C(sp³)–C(sp³) Cross-Dehydrogenative-Coupling



R^1 = Ar, c-Alkyl; R^2 = Ph, OMe;
R^3 = Ph, OMe, OEt, NHPh, tBu, Me,

25-87%

- First example of Fe catalzsed CDC
- DTBP higher yield than TBHP

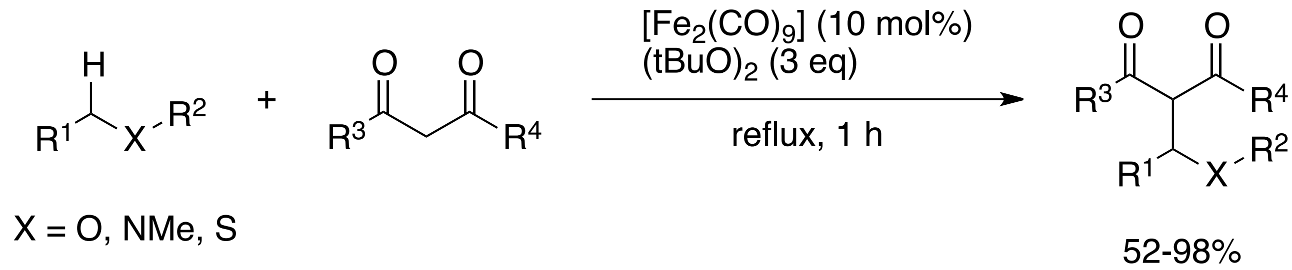


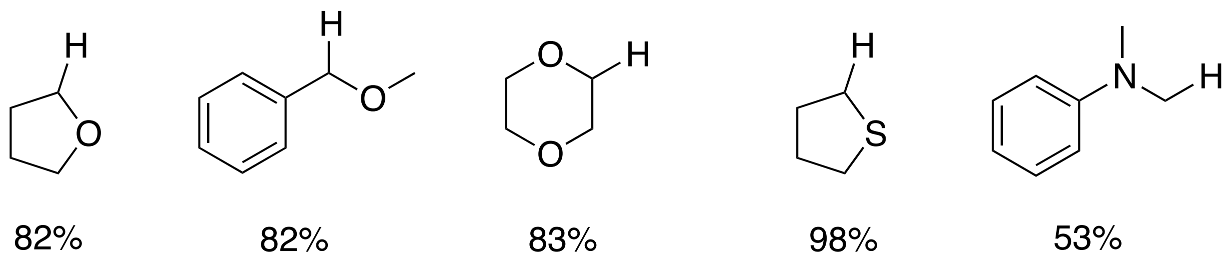
Z. Li, L. Cao, C.-J. Li, *Angew. Chem. Int. Ed. Engl.* **2007**, *46*, 6505–6507

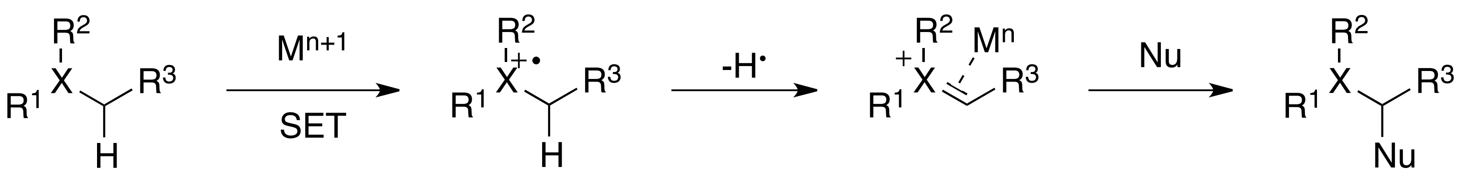
Applications of Fenton Chemistry

C-C bond formation

α-N/α-O C(sp³)–C(sp³) Cross-Dehydrogenative-Coupling



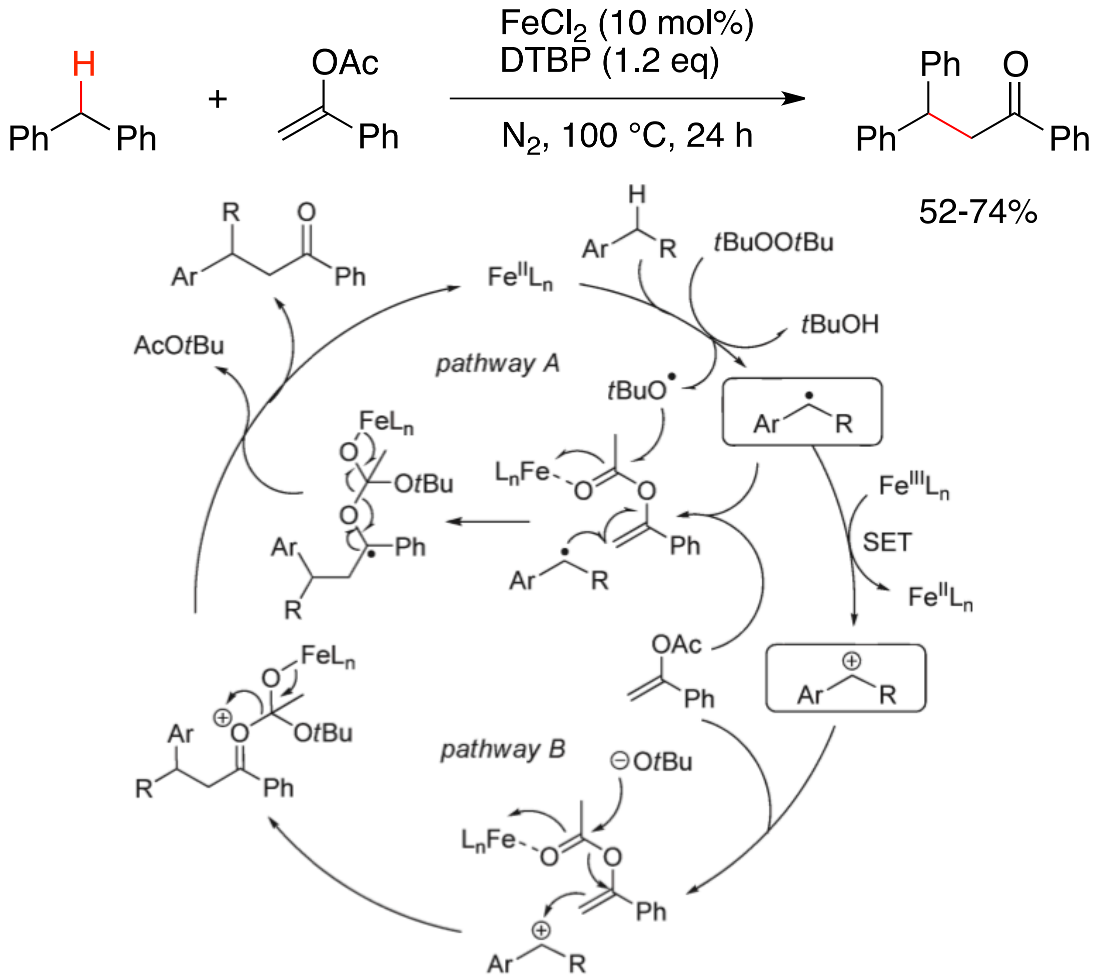




Z. Li, R. Yu, H. Li, *Angew. Chem. Int. Ed. Engl.* **2008**, *47*, 7497–7500.

Applications of Fenton Chemistry

C-C bond formation
C(sp^2)–C(sp^3) Cross-Dehydrogenative-Coupling

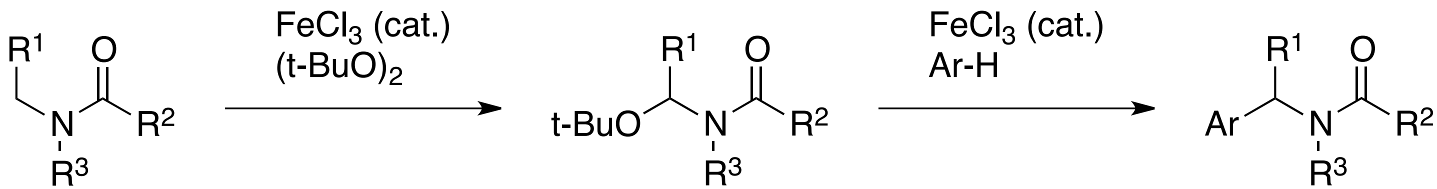


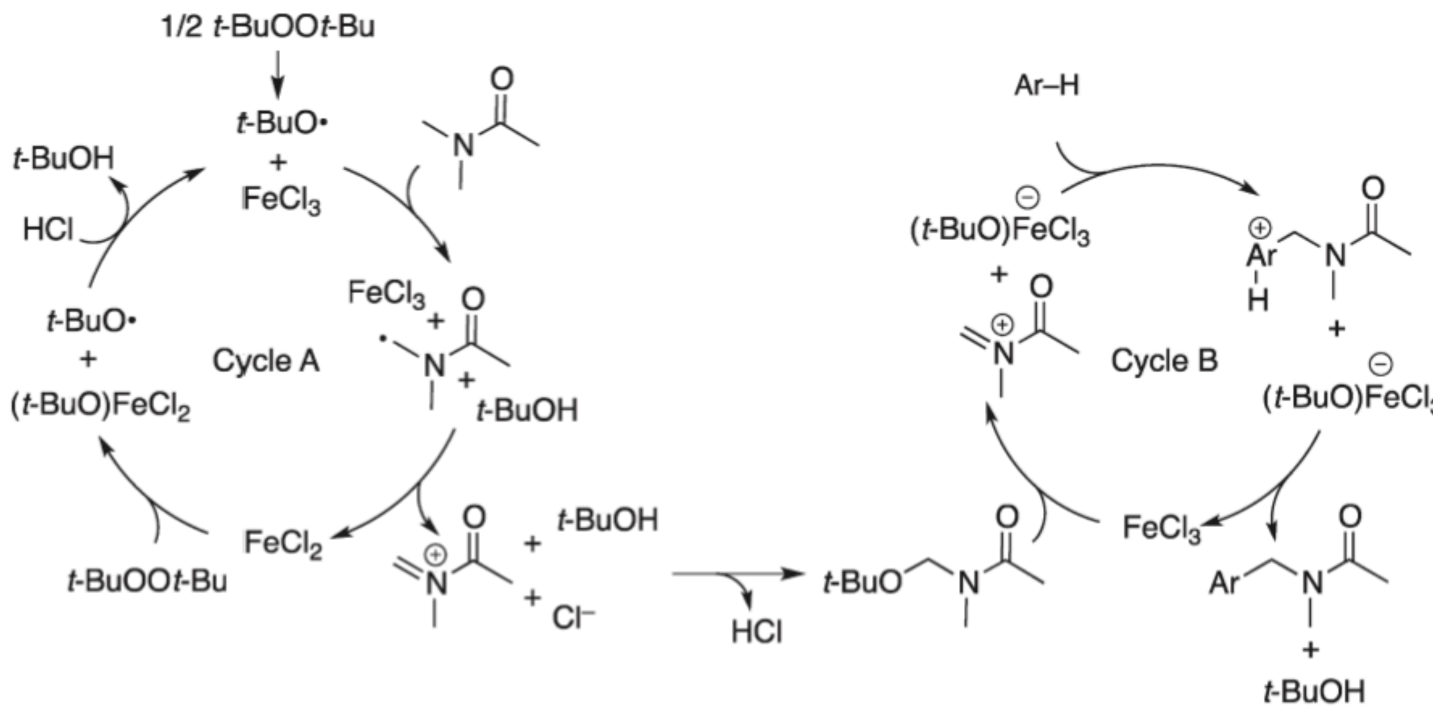
C.-X. Song, G.-X. Cai, T. R. Farrell, Z.-P. Jiang, H. Li, L.-B. Gan, Z.-J. Shi, *Chem. Commun.* **2009**, 6002–6004

Applications of Fenton Chemistry

C-C bond formation

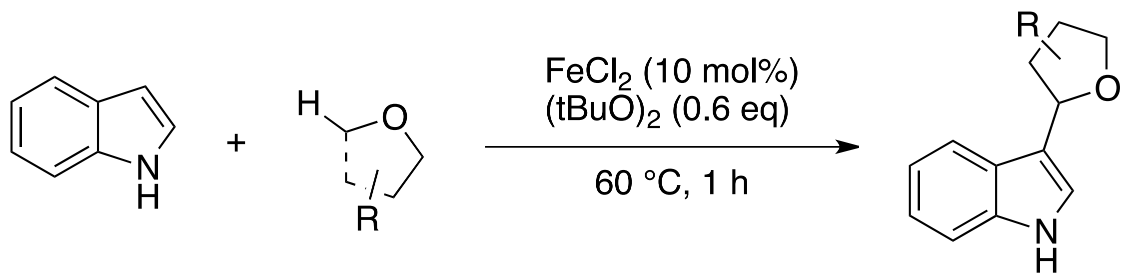
α-N-C(sp^3)–C(sp^2) Cross-Dehydrogenative-Coupling

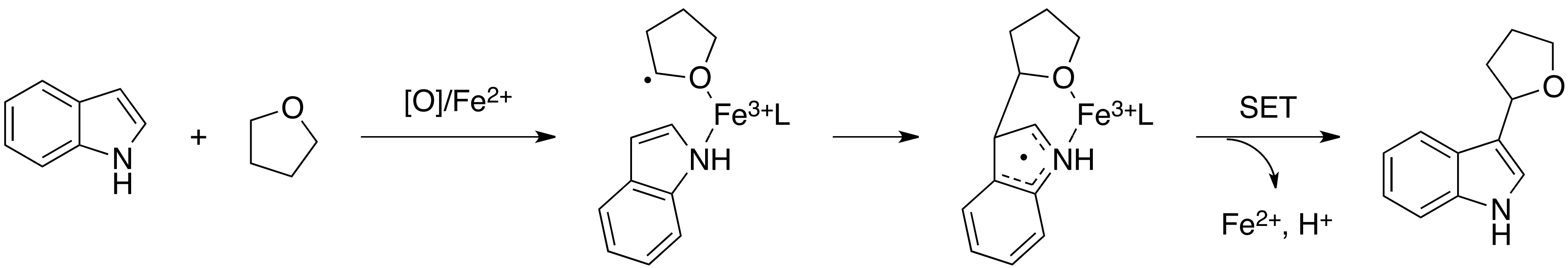


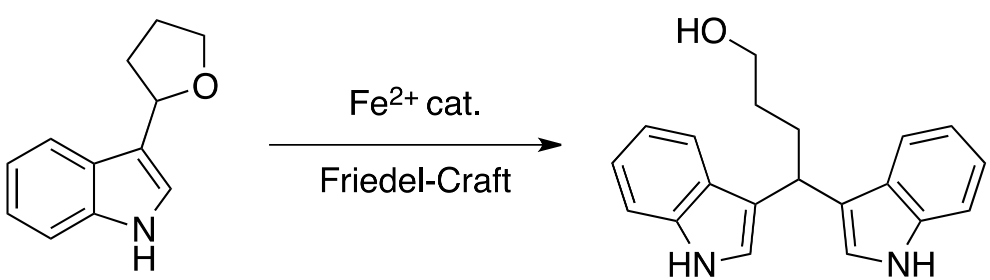


E. Shirakawa, N. Uchiyama, T. Hayashi, *J. Org. Chem.* **2011**, *76*, 25–34

Applications of Fenton Chemistry
C-C bond formation
C(sp^2)–C(sp^3) Cross-Dehydrogenative-Coupling







X. Guo, S. Pan, J. Liu, Z. Li, *J. Org. Chem.* **2009**, *74*, 8848–8851

Applications of Fenton Chemistry

C-C bond formation

C(sp^2)–C(sp^3) Cross-Dehydrogenative-Coupling

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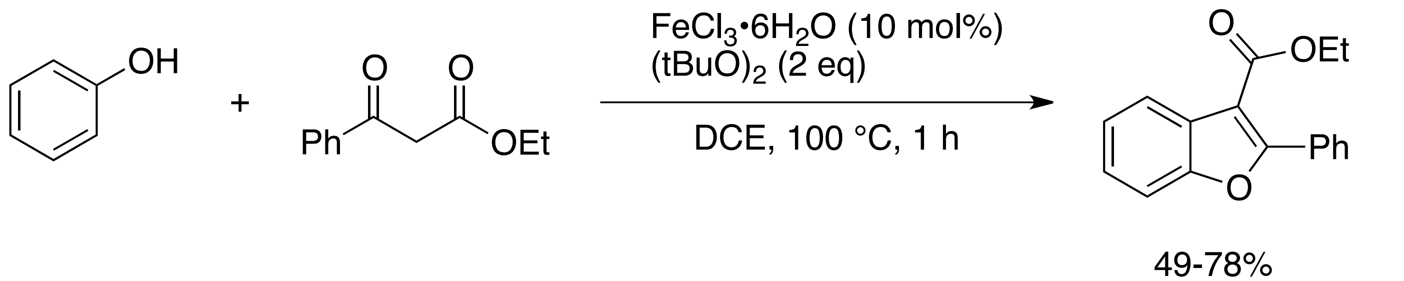
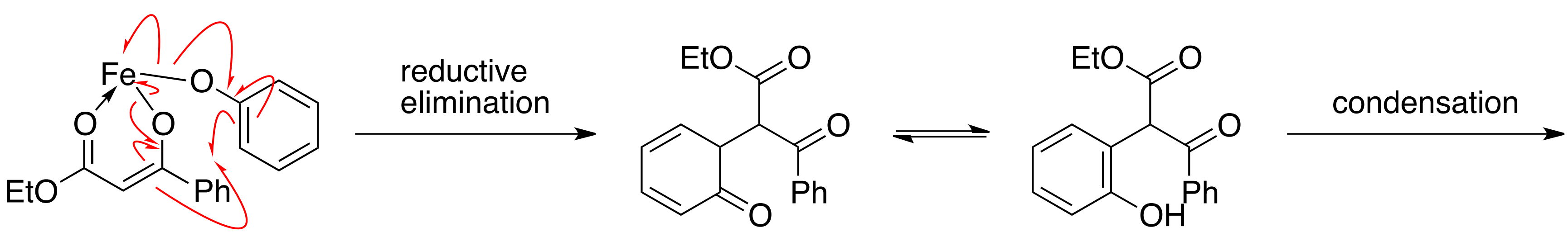


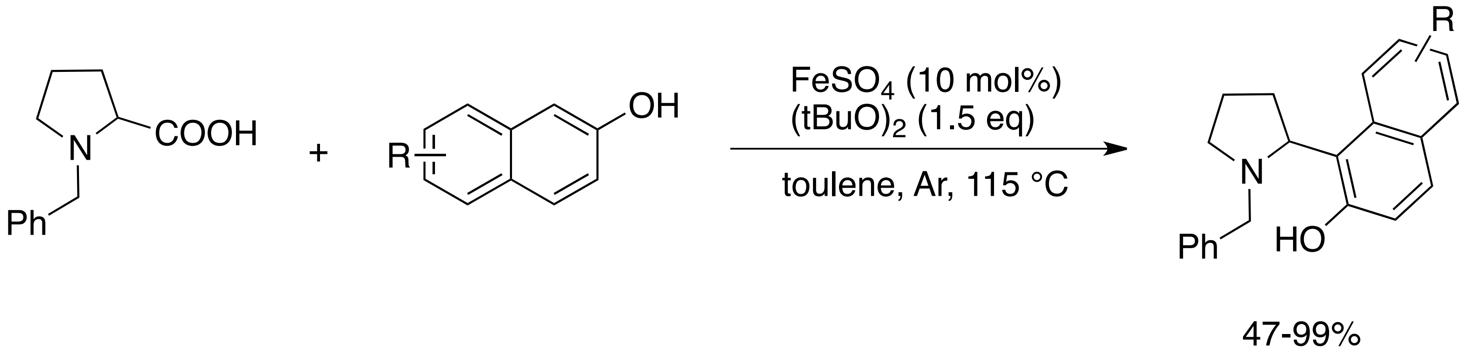
- Construction of polysubstituted benzofurans
- Water in catalyst promoted reaction greatly
- Other H$^+$ can be used such as AcOH, MeOH, EtOH, tBuOH
- Efficient pathway for biologically important 3-carboxylate benzofurans



X. Guo, R. Yu, H. Li, Z. Li, *J. Am. Chem. Soc.* **2009**, *131*, 17387–17393

Applications of Fenton Chemistry

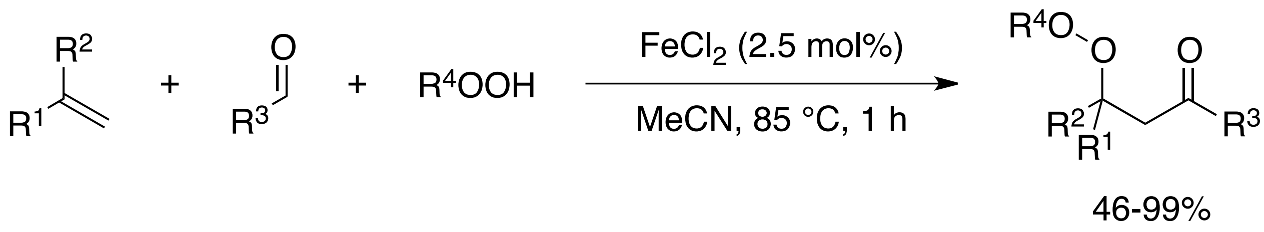
C-C bond formation
C(sp^2)–C(sp^3) Decarboxylative-Coupling

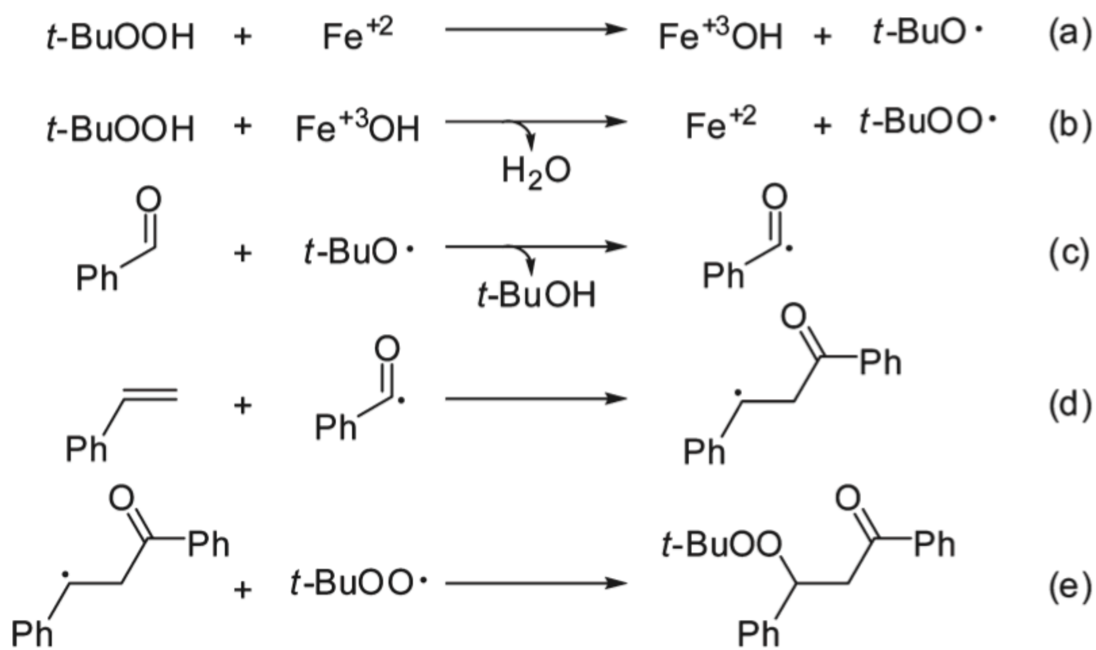


- Site-specific functionalisation
- Generation of novel amino-naphtol ligands (non-expensive)

H.-P. Bi, W.-W. Chen, Y.-M. Liang, C.-J. Li, *Org. Lett.* **2009**, *11*, 3246–3249.

Applications of Fenton Chemistry
C-C bond oxidation
C(sp^2)–C(sp^2) Cross-Dehydrogenative-Coupling





W. Liu, Y. Li, K. Liu, Z. Li, *J. Am. Chem. Soc.* **2011**, *133*, 10756–10759

Applications of Fenton Chemistry
C-C bond formation
C(sp)–C(sp^3) Cross-Dehydrogenative-Coupling

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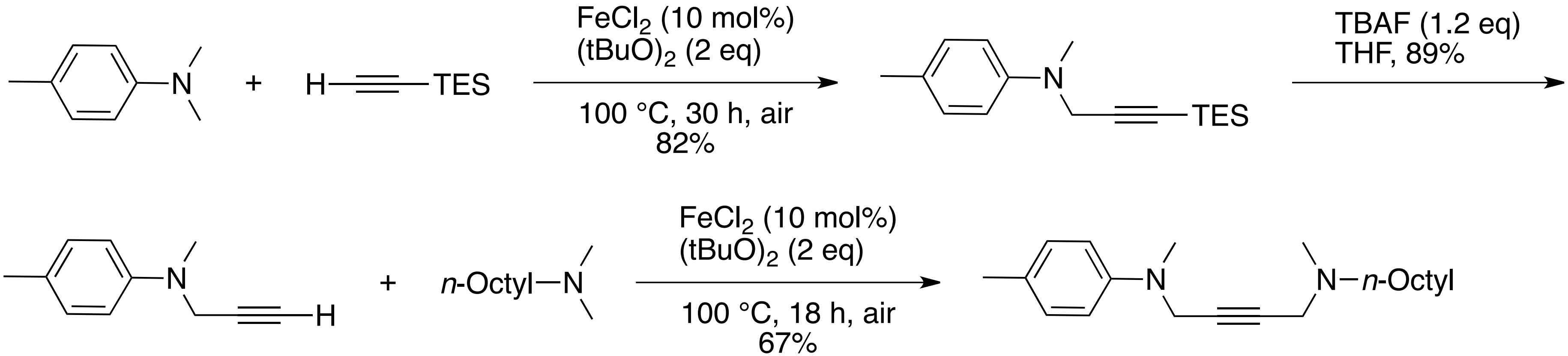
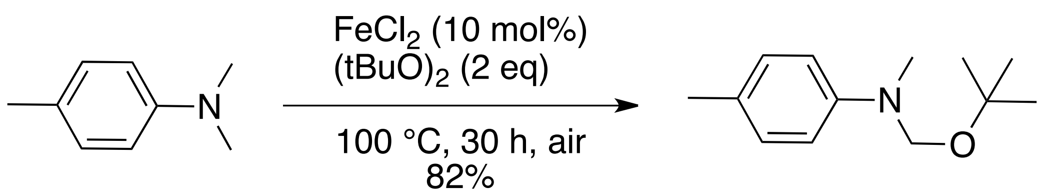


- no solvent
- Reaction *via* iminium ion and deprotonation of alkyne



in the absence of $FeCl_2$
not observed

C. M. R. Volla, P. Vogel, *Org. Lett.* **2009**, *11*, 1701–1704.

Applications of Fenton Chemistry
Other Metals

- Common are Mn(III), Cu(I+II), Co(II), Ti(III+IV), V(II), Cr(II+V), Ce(IV)
- Uranium (U(IV) and U(V))

$$U(IV) + H_2O_2 = U(V) + HO$$
$$U(V) + H_2O_2 = U(VI) + HO$$
$$U(IV) + HO = U(V) + H_2O$$
$$U(V) + HO = U(VI) + H_2O$$
$$HO + H_2O_2 = HO_2 + H_2O$$
$$U(IV) + HO_2 = U(V) + H_2O_2$$
$$U(V) + HO_2 = U(VI) + H_2O_2$$
$$HO + HO_2 = H_2O + O_2$$

- Neptunium (Np(VI))

$$H_2O_2 \underset{}{\overset{K}{\rightleftarrows}} H^+ + HO_2^-$$
$$HO_2^- + NpO_2^{+2} \xrightarrow{k_1} NpO_2^+ + HO_2$$
$$HO_2 + NpO_2^{+2} \xrightarrow{k_2} NpO_2^+ + O_2 + H^+$$
$$HO_2 + NpO_2^+ \xrightarrow{k_3} NpO_2^{+2} + HO_2^-$$
$$HO_2 + H_2O_2 \longrightarrow OH + H_2O + O_2$$
$$OH + H_2O_2 \longrightarrow HO_2 + H_2O$$
$$OH + NpO_2^+ \longrightarrow OH^- + NpO_2^{+2}$$

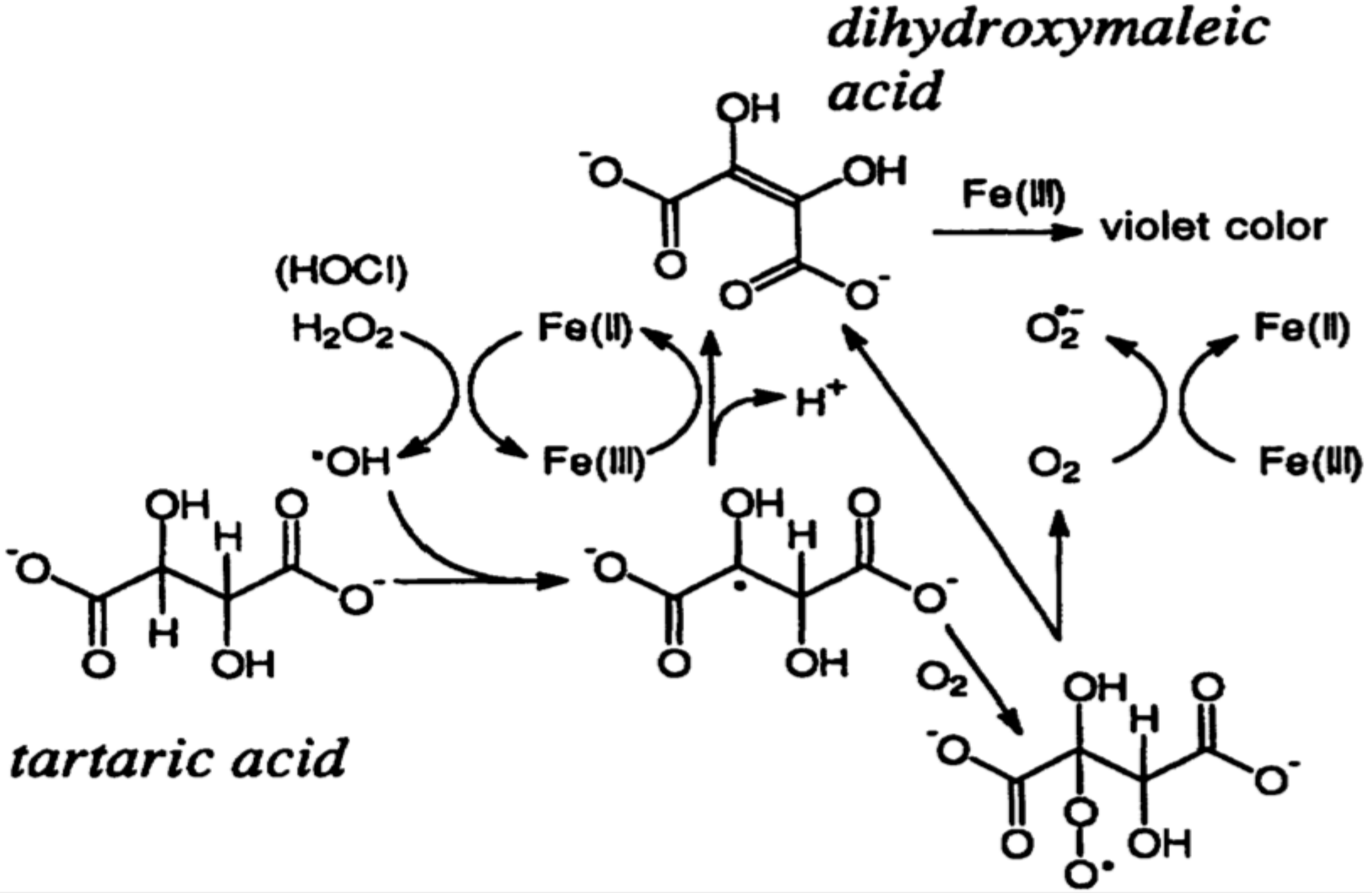
F. B. Baker, T. W. Newton, *J. Phys. Chem.,* **1961**, *65*, 1897–1899
A. J. Zielen, J. C. Sullivan, D. Cohen, J. C. Hindman, *J. Am. Chem. Soc.* **1958**, *80*, 5632–5635

Fenton Chemistry
Conclusion

- Fenton‘s Discovery
- The controversy concerning the mechanism (radical vs. non-radical pathway)
- Cytochromes P-450 and their unusual oxidations
- Ecological Fenton Chemistry
- Gif Chemistry and Oxidation of inactivated C-H bonds
- Various Applications of Fenton‘s Chemistry

Thank you for your attention

Fenton's Reaction
Mechanism



P-450 Oxidations I

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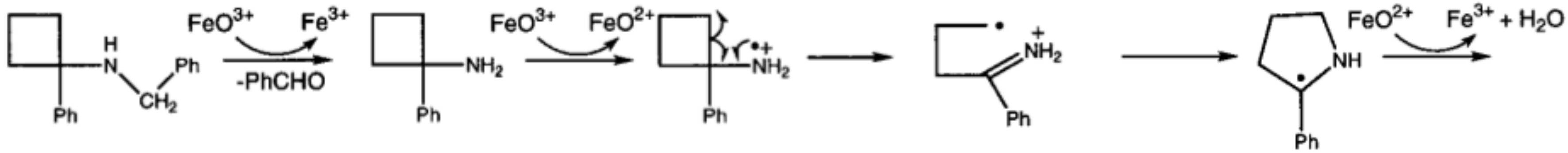
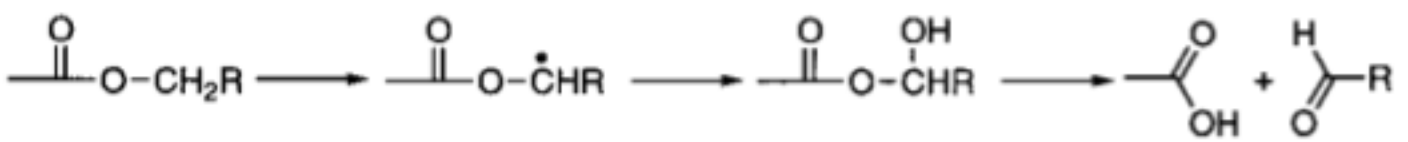
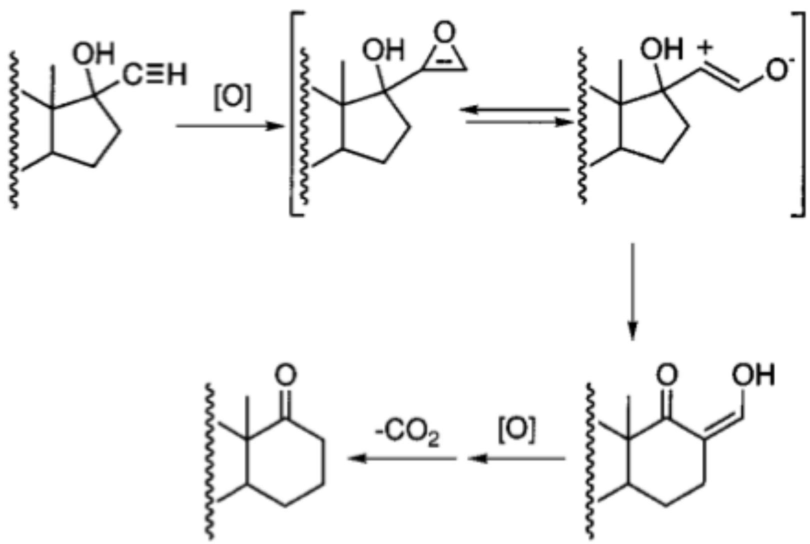
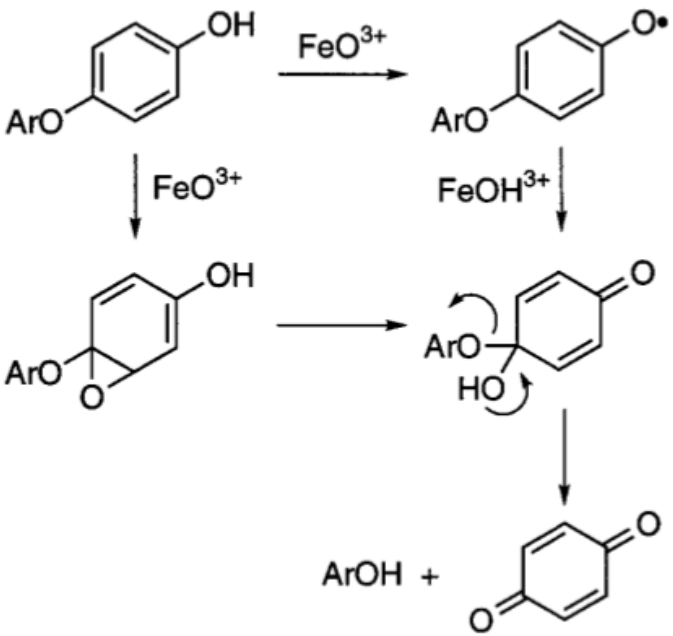
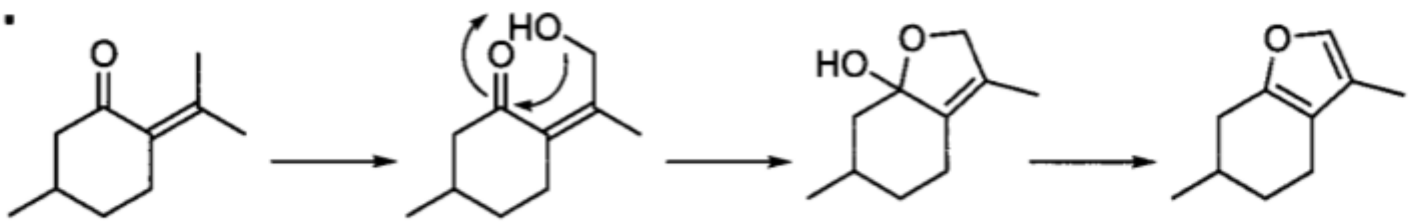






P-450 Oxidations II

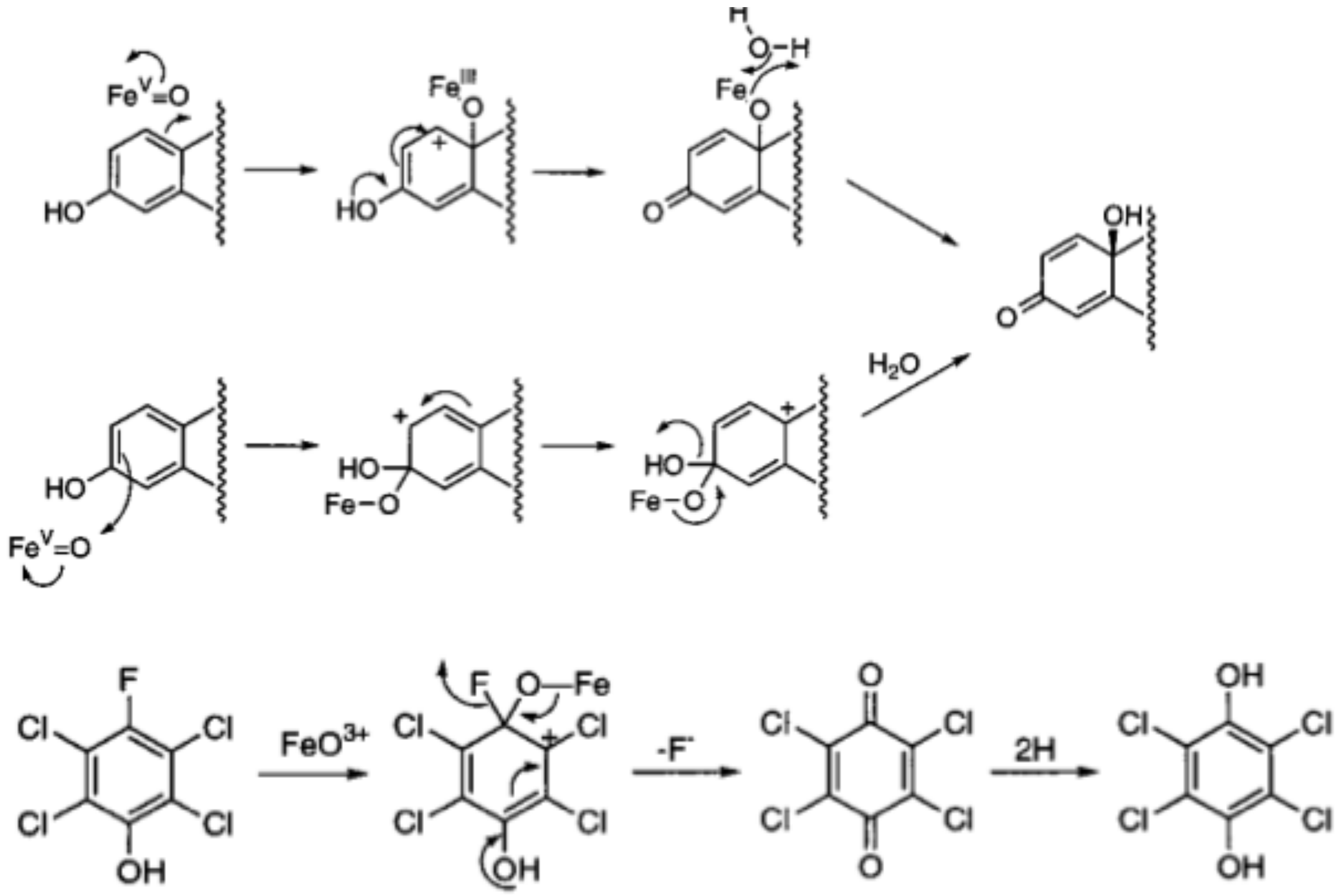




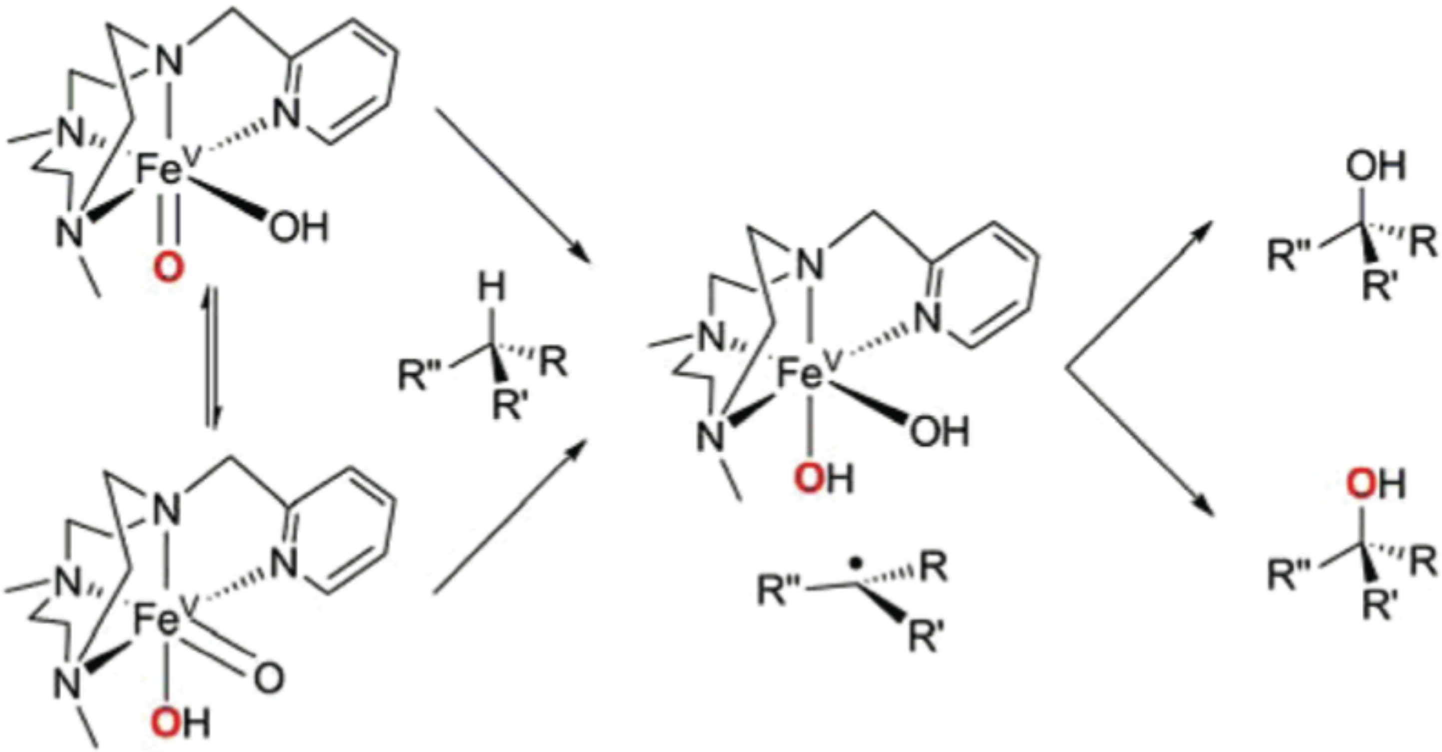
Pulegone

Menthofuran

P-450 Oxidations III



Ligand Effect



Three Component Coupling

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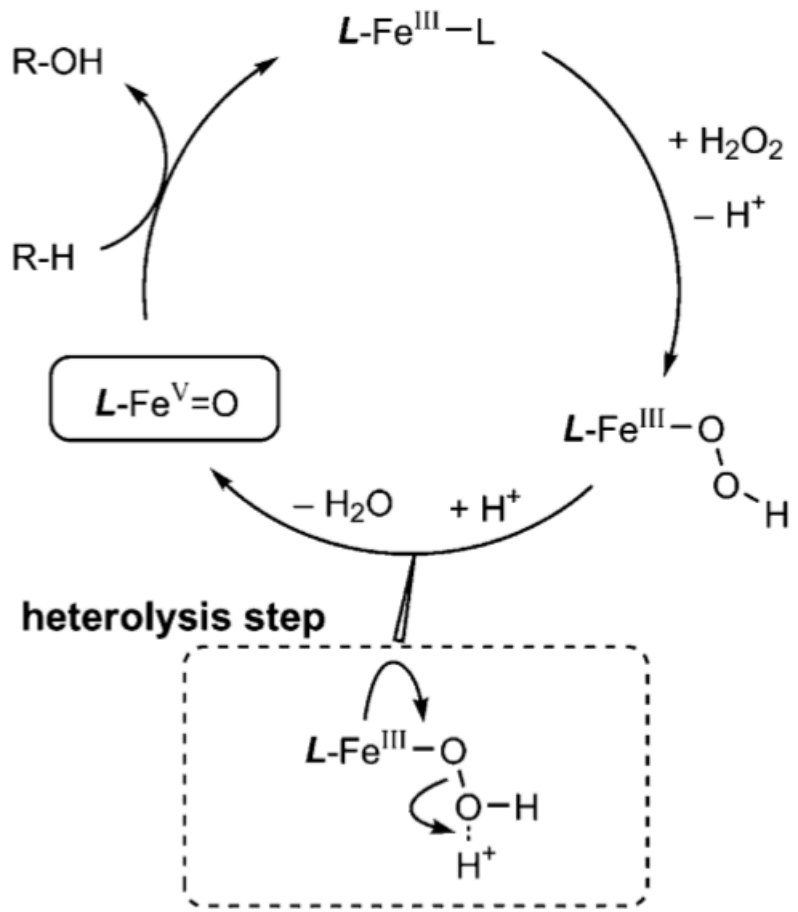

Proline Coupling, Decarboxylation

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