

## Oxidative degradation of antibacterial drugs in effluents of hospital wastes: A mechanistic Approach

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### **Abstract:**

This study provides an overview of chemical kinetics, discusses various oxidant such as Hexacyanoferrate (III), Manganese dioxide, Diperoxidocuprate(III), Chloramine-T, Potassium permanganate, and their oxidizing qualities, and briefly touches on antibacterial, which are potent antibiotics used in both human and veterinary medicine. Environmental impact problems are raised by the fate of antibiotic parent and metabolite chemicals that enter environmental ecosystems through several mechanisms. The goal of the current study was to calculate the reaction kinetics and elucidate the reaction pathways involved in oxidative degradation of the environmentally relevant antibacterial drugs in an acidic/alkaline aqueous media. this work will be useful in treating wastewater in areas where fluoroquinolone antibacterial drugs have caused pollution. This study also provides a comparative discussion about oxidizing nature of various oxidants.

### **Introduction:**

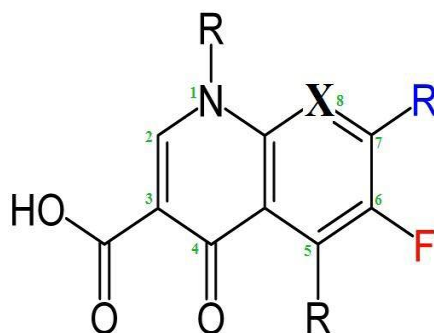
Chemical kinetics, the study of chemical process rates, encompasses the development of mathematical models that can characterize the properties of a chemical reaction as well as the exploration of how various experimental conditions can affect a chemical reaction's rate and provide insight into the mechanism and transition states of the reaction. Chemical engineers and chemists can better comprehend and describe chemical processes and the intricate chemistry of living systems by using the mathematical models that describe chemical reaction kinetics. Chemical reactors can be designed or modified using these models to optimize product yield, segregate products more effectively, and get elimination of by-products that are hazardous for the environment. For each reaction, an appropriate mechanism is suggested based on observations made within the experimental conditions. Reaction mechanism is the process by which molecules breakdown and atoms reorganize in accordance with the order of a reaction with regard to different reactive species. However, all of the chemical events in a chemical reaction cannot be explained by the order of the reaction with respect to the reactants and thermodynamic parameters. This is mostly because the reactions rarely proceed in a single step. Numerous basic reactions take place in the ongoing conversion of the reactants to the products and such types of reactions are either complex or

composite in nature. tion of the reactants and catalyst, the stereochemistry of the reactants and products, and the quantities of each must all be explained in a comprehensive mechanism. Further details regarding the specific stages that could be a part of the reaction mechanism are provided by the rate law. Setting up a kinetics experiment for the chemical reaction is the first step towards determining the rate law. To find the change in a species' concentration over time, a kinetics experiment is methodically designed to allow measurements to be taken at specific intervals.

## Antibacterial drugs:

Every reactive intermediate, activated complex, transition state, broken and created bonds, and so on are all described by the reaction mechanism. Antibiotics, including tetracycline,  $\beta$ -lactum, microcline, sulphonamide, lincosamide, and fluoroquinolone, are among the different kinds of antibiotics that fall under the category of synthetic broad-spectrum antibiotics. Among these antibiotics, fluoroquinolones (FQs) are widely used antibacterial agents in a variety of applications, including the treatment of bacterial infections, urinary tract infections, treatment of STDs, infections of the bones and joints, typhoid fever, and tuberculosis, among others [1].

Gram-positive and gram-negative aerobic and anaerobic organisms are among the expanded spectrum of bacteria that the more recent FQs can inhibit clinically [2]. Originating as a by-product of the manufacture of the antimalarial drug chloroquine [3], nalidixic acid was the first member of the quinolone class of antimicrobials to be reported in 1962 [4]. The first 4-quinolone to be marketed for clinical usage was nalidixic acid. It has pharmacokinetic qualities that made it useful for treating urinary tract infections and was effective against some Gram-negative bacteria. The discovery of adding a fluorine atom and a piperazine ring to the basic quinolone nucleus at positions 6 and 7 marked a significant advancement for this class of antibacterial drugs. The "pharmacore" or fundamental fluoroquinolone molecule is depicted in (Fig. 1). These replacements lowered toxicities, enhanced absorption, and enhanced antibacterial action. Many FQs were patented after that and are still in use today. As a result, ongoing efforts have been made to replace the simpler fluoroquinolone pharmacophore with a complex one.



*Fig. 1.1: General structure of 4-Quinolones. Quinolones: X = CH or C-R8; Naphthyridones: X = N.*

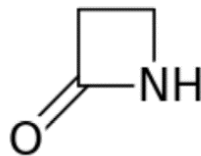
## Classification of Antibacterial Drugs

Bacteriostatic substances are those that prevent bacterial development, whereas bactericidal substances kill germs [5]. The term "antibiotic" is further classified as "antivirals," "antifungals," and "antibacterials" depending on the type of microorganisms they oppose [6].

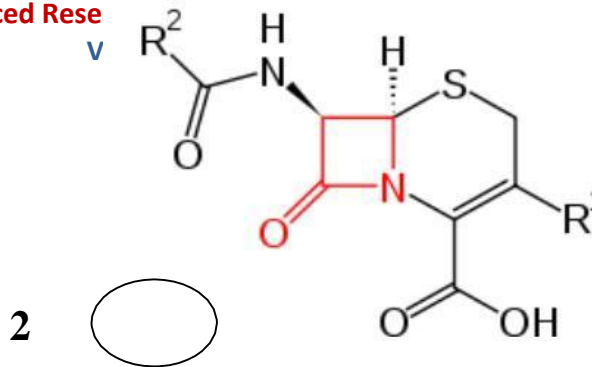
There are several different kinds of antibiotics and they can be classified based on their chemical structure, mechanism of action, action spectrum and the route of administration [7]. Out of these classifications the most popular one is their mechanism of action and based on it the most common groups are  $\beta$ -lactam, sulfonamides monobactams, carbapenems, amino glycosides, glycopeptides, lincomycin, macrolides, polypeptides, polyenes, rifamycin, tetracyclines, chloramphenicol, quinolones, and fluoroquinolones, Nitro-derivatives Fusidanos, Pleuromulins, Diamino pyridines oxazolidinones, phosphonate's etc. [8].

### 1.3.1 Beta-lactams

This class of antibiotics' members have a 3-carbon and a single, highly reactive nitrogen ring (**Fig.2 and 3**). They obstruct the proteins necessary for the creation of bacterial cell wall, and thus either eliminates or prevents them from growing. Antibiotics have the ability to attach to this Penicillin-binding protein (PBP)-enzymes, and as a result, they obstruct the peptidoglycan production leading to cell death [9]. The most well-known beta-lactam class's representatives are Monobactams, Cephalosporins, Penicillins, and carbapenems.



*Fig. 1.2: Chemical structure of Beta-lactam ring*



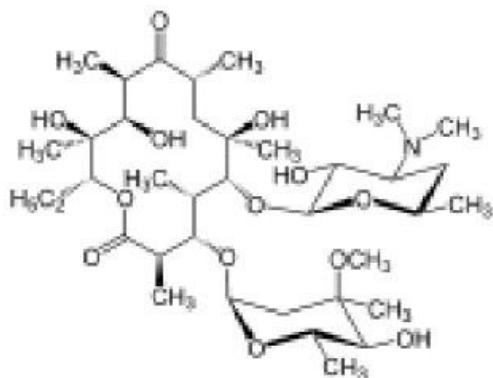
*Fig. 1.3 Chemical structure of beta lactam structure. Core structure of Penicillins (1) and Cephalosporins (2)*

### 1.3.2 Macrolides

As a metabolic byproduct of the fungus *Saccharopolyspora erythraea*, which inhabits soil, J. M. McGuire identified and isolated the first antibiotic in this class in 1951 [10].

The distinctive deoxy polysaccharides L-cladinose and D-desosamine are connected to 14-, 15-, or 16-membered macrocyclic lactose rings, which are the hallmark of macrolides (**Fig. 4**). Macrolides typically have a broad spectrum of action, While certain bacterial species, like *Streptococcus pneumoniae*, are resistant to

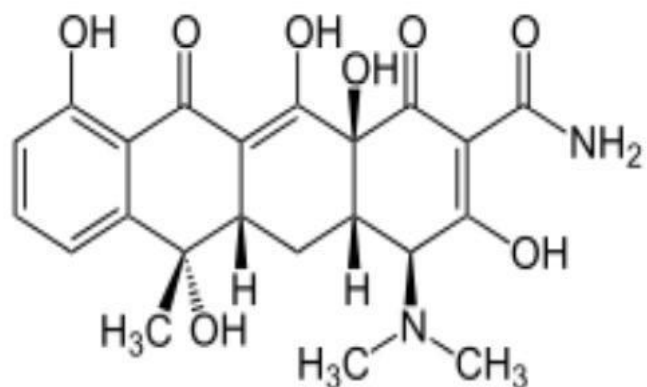
the antibiotics. Erythromycin, azithromycin, and clarithromycin are a few examples of members [11]



**Fig. 1.4: Structure of Macrolide**

### 1.3.3 Tetracyclines

Benjamin Diggar isolated tetracycline in 1945 from a soil bacterium belonging to the genus *Streptomyces* [12]. Aureomycin, also known as chlorotetracycline, was the original member of this class. Members of this class are identified by their names ending in "-cycline" and possess four (4) hydrocarbon rings (**Fig. 1.5**). This class of antibiotics has been divided into several generations according on how they were synthesized. Tetracycline, Chlortetracycline, Oxytetracycline, Demeclocycline, Doxycycline, Lymecycline, Meclocycline, Methacycline, Minocycline, Rolitetracycline tige cycline belongs to various class of tetracyclins [13].



**Fig. 1.5 Structure of Tetracycline**

### ***1.3.4 Quinolones***

Scientists searching for antimalarial medications initially found this class of antibiotics as nalidixic acid. Two important families of drugs are quinolones and naphthyridones. These groups of compounds include nalidixic acid, enoxacin, ciproxacin, temafloxacin, sparfloxacin, cinoxacin, norfloxacin, and ofloxacin [14]. Numerous changes to its parent structure since its discovery gives many other anti bacteria which boosted their bioavailability, spectrum of activity, and potency, improving their effectiveness in treating a variety of diseases, including respiratory tract, systemic, and urinary tract infections [15].

### ***1.3.5 Aminoglycosides***

Streptomycin, which was initially identified in 1943, was the first medication to be found within this class of antibiotics [16]. The range of antibiotic action exhibited by aminoglycosides is extensive. By attaching to a ribosomal subunit, they can prevent bacteria from synthesizing proteins [17]. They work well against aerobic Gram-negative rods and some Gram-positive bacteria. According to Talaro and Chess [18], streptomycin is used to treat tuberculosis, tularemia, and the bubonic plague on several times.

### ***1.3.6 Sulfonamides***

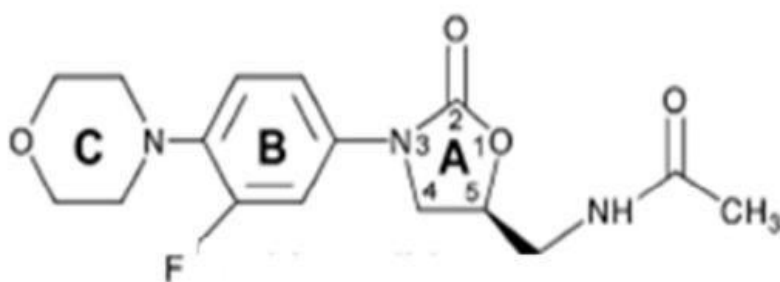
Sulfonamides are widely used to treat a variety of infections, such as tonsillitis, septicemia, meningococcal meningitis, bacillary dysentery, and some urinary tract infections. They inhibit both Gram-positive and Gram-negative bacteria, including *Nocardia*, *E. coli*, *Klebsiella*, *Salmonella*, *Shigella*, and *Enterobacter*, as well as some protozoa [19]. Research has demonstrated that sulfonamides can also obstruct chemicals that cause cancer [20]. Synthetic antimicrobial drugs containing the sulphonamide group are known as the original antibacterial sulfonamide [21]. Due to their toxicity and adverse effects—some of which include hemolytic anemia, porphyria, urinary tract disorders, and hypersensitivity reactions—they should be used with caution [22].

### ***1.3.7 Glycopeptides***

Since the last 20 years, semi-synthetic derivatives of glycopeptide antibiotics, also known as GPAs, have emerged with better pharmacokinetic and activity characteristics [23]. GPAs are often shortened forms of natural drugs. Glycopeptides are naturally composed of a cyclic peptide consisting of seven amino acids and two sugars attached to it. Yim and Researchers [24] presents the structures of several types of glycopeptides in an effective manner. Additionally, a lipophilic side chain extends the half- life of glycopeptides and has antibacterial activity[25].

### 1.3.8 Oxazolidinones

A class of synthetic antibiotics called oxazolidinones have only recently been permitted for usage. The first component to be synthesized, linezolid (Figure 6), was only allowed for use in clinical settings in 2000. By attaching to the P site of the ribosomal 50S subunit, oxazolidinones prevent the synthesis of new proteins[26]. According to Moellering [27], linezolid is used to treat skin infections and respiratory tract infections. While following the recommended standard procedures for administering linezolid is generally safe, extended treatment might lead to adverse effects such as myelosuppression, which can cause anemia and thrombocytopenia [28].



*Fig. 1.6 Structure of Linezolid [17]*

## 1.4. Oxidation of Antibacterial Drugs

It is quite concerning that antibiotics are present in the aquatic environment. Ongoing research aims to effectively eliminate or convert antibiotics into species with a reduced risk. An efficient method for the goal is the transformation of antibiotic by oxidation [29]. The oxidizing ingredient in water treatment combines with the impurities to produce less toxic molecules. Enhancing the efficacy of antibiotics removal in natural waterways requires several types of chemical techniques and advanced oxidation processes (AOPs). These consist of sonolysis [30], chlorination [31], chloramination [32], and oxidation of chlorine dioxide [33], ozonation [34], photocatalytic and photolytic degradation [35], persulfate (PS) [36], peroxomonosulfate (PMS) techniques [37], Fenton reaction processes [38], permanganate [39], and ferrate(VI) [40]. Reactive oxygen species (ROS), reactive sulfur species (RSS), reactive halogen species, and high-valent transition metal species were used in the majority of these technologies to eliminate antibiotics [41]. The order of oxidants' reactivity in basic solution is as follows:  $\text{SO}_4^{\cdot-} > \cdot\text{OH} > \text{O}_3 > \text{ClO}_2 > \text{H}_2\text{O}_2 > \text{ClO}^- > \text{Fe (VI)} > \text{Mn (VII)}$ . The in-situ creation of highly reactive radicals, like  $\cdot\text{OH}$  or  $\text{SO}_4^{\cdot-}$ , is used as a substitute or addition to traditional wastewater treatment techniques for this reason [42]. Furthermore, two high-

valent metal oxidants, Mn (VII) and Fe (VI), have been developed [43,44] as promising technologies for the oxidative treatment of several pharmaceuticals.

#### **1.4.1 Metal-based catalytic ozonation**

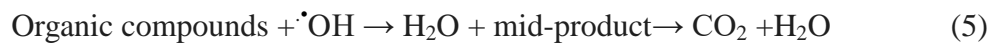
Ozonation has been widely used in drinking water treatment as well as wastewater treatment over the past few decades to effectively remove a variety of micro-pollutants [45]. The aqueous breakdown of organic compounds by ozonation generally happens via two chemical pathways: indirect assault with  $\cdot\text{OH}$  generated by  $\text{O}_3$  decomposition in water and direct oxidation, also known as ozonolysis [46]. In the process of aqueous ozonation,  $\text{O}_3$  targets specific organic molecules' nucleophilic moieties, like unsaturated bonds, aromatic, and amino groups, using an electrophilic mechanism, while  $\cdot\text{OH}$  interacts much more broadly with the majority of organic compounds through hydrogen atom transfer, adduct formation and single electron transfer routes [47]. The removal and mineralization of CIP by ozonation were effectively increased by the addition of Manganese oxides supported by carbon nanotubes ( $\text{MnO}_x/\text{MWCNT}$ ), according to Sui et al. [48]. Application with  $\text{MnO}_x/\text{MWCNT}$  enhanced the elimination of CIP in actual water matrix when compared to ozonation. The faster  $\cdot\text{OH}$  generation in the  $\text{MnO}_x/\text{MWCNT}$  catalytic ozonation system is thought to be caused by three factors: (i)  $\text{O}_3$  decomposition initially began by  $\text{OH}^-$ , (ii)  $\text{O}_3$  improvement by MWCNTs to produce  $\cdot\text{OH}$ , and (iii) electron transfer between  $\text{MnO}_x$  and  $\text{O}_3$ . [49].

#### **1.4.2 The Fenton Reaction System**

Fenton oxidation, which uses  $\text{Fe}^{2+}$  and  $\text{H}_2\text{O}_2$  in an acidic medium (pH 2-4), is a commonly used treatment for the removal of micro-pollutants from industrial wastewater and surface water [50]. This metal-catalyzed oxidation is a straightforward and adaptable process that can produce reactive  $\cdot\text{OH}$  in homogeneous or heterogeneous systems via free radical chain reactions, all without the need for specialized equipment or reactants [51].  $\cdot\text{OH}$  has a high redox potential and is a non-selective oxidizing species that can remove numerous organic micropollutants at once.  $\text{Fe}^{2+}$  functions as a catalyst and quickly combines with the oxidant  $\text{H}_2\text{O}_2$  to form  $\text{Fe}^{3+}$  and  $\cdot\text{OH}$ , which have potent oxidizing abilities.  $\text{Fe}^{3+}$  combines with  $\text{H}_2\text{O}_2$  in a solution to form  $\text{Fe}^{2+}$  when  $\text{Fe}^{3+}$  is present. The  $\text{Fe}^{2+}$  cycle can produce more  $\cdot\text{OH}$  if there is enough  $\text{H}_2\text{O}_2$  present [52].







By using UV radiation, H<sub>2</sub>O<sub>2</sub>, and ultrasound in combination with a unique marine sediment/titanate, Tavasol et al. were able to remove cefadroxil from aqueous media [53]. The rate of Cephalexin removal was as high as 94.71% with increased titanium loading on the sediment, which had a high specific surface area and photocatalytic activity (Fig.7).

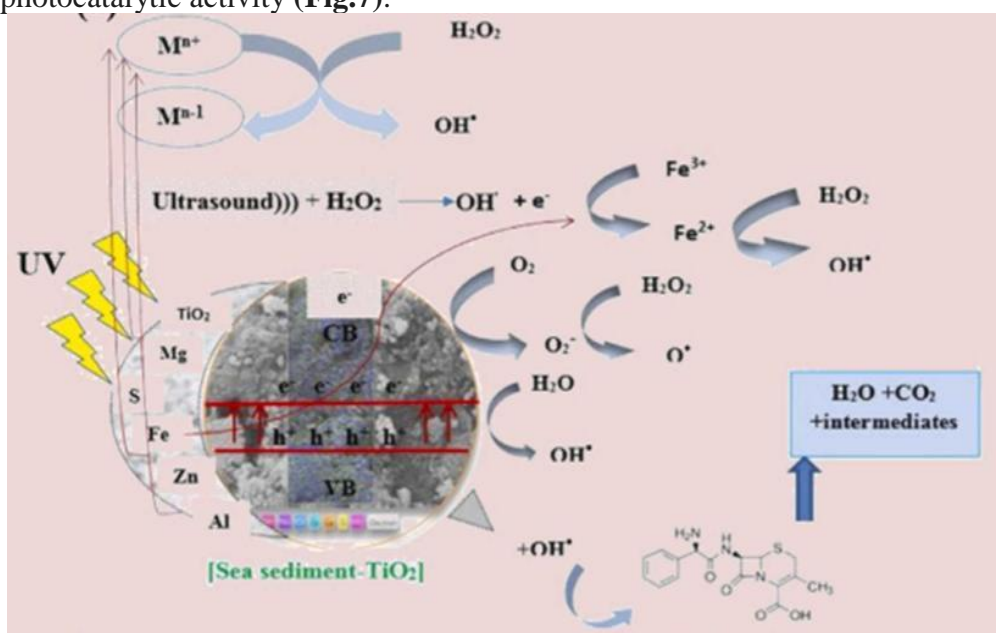


Fig.1.7: Process suggested for the "sea sediment/titanium-H<sub>2</sub>O<sub>2</sub>-UV-ultrasonic" system's eliminating cephalexin [53].

### 1.4.3 Oxidation systems based on SO<sub>4</sub><sup>-</sup> Radicals

Research in activated PS/PMS technology for wastewater and water treatment is growing [54,55]. PS/PMS can be efficiently activated by energy inputs like heat [56] or UV [57], but a more practical way for its practical application is single- electron transfer activation by transition metals like iron, copper, and cobalt.

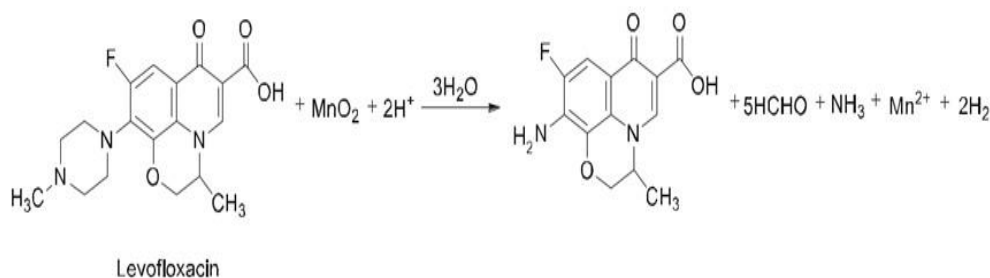
Fe<sup>2+</sup> in the aqueous phase or on the surface of the catalyst may generally activate PS to produce reactive SO<sub>4</sub><sup>-</sup> species quickly, which speeds up the early- stage breakdown of these FQs [58]. According to Sturini et al. [59], the functional groups oxidized during the photodegradation of FQs followed

this order: piperazinyl ring > benzo ring > pyridine ring.

#### 1.4.4 Manganese dioxide

The oxidation state of manganese varies from +7 to +2. The most stable oxidation states among them are +2, +4, and +7. In the aquatic environment, Mn(IV) oxide/hydroxide colloidal particles, plays a crucial role in initiating chemical transformations of organic molecules [60]. Due to its changeable valence, it has been widely studied and employed for electrochemical capacitor, oxidation, and catalysis applications [61]. The preparation of water-soluble colloidal manganese dioxide from the reduction of aqueous  $\text{KMnO}_4$  by  $\text{Na}_2\text{S}_2\text{O}_3$  under neutral conditions is made possible for the first time quantitatively by Perez- Benito et al. [62]. In an aqueous acidic solution at  $25^\circ\text{C}$  temperature, the kinetics of levofloxacin

oxidation by water-soluble manganese dioxide have been investigated [63]. According to the reaction's stoichiometry, 1 mol of manganese dioxide is required for the oxidation of 1 mol of levofloxacin [Fig. 1.8]. In various permanganate processes, colloidal  $\text{MnO}_2$  has been employed as an auto catalyst [64]. Mn(III) and Mn(IV) oxides are two of the major manganese oxides that act as oxidizing agents for different organic molecules.



**Fig. 1.8. Stoichiometry of reaction between Levofloxacin & Manganese dioxide**

#### 1.4.5. Hexacyanoferrate (III)

Environmentally friendly oxidants, iron in its various oxidation states from +3 to +6, have drawn a lot of interest [65]. Iron (III) complexes are receiving significant attention as oxidants because of their affordable availability, low evaluation challenge, and ability to react in both acidic and alkaline media [66]. Hexacyanoferrate(III){HCF(III)} is an environmentally benign oxidant, and employed as a coagulant [67]. Their redox potential is the only factor that affects their oxidation capacity [68]. It is a single-electron oxidant with a pair of  $[\text{Fe}(\text{CN})_6]^{3-} / \text{Fe}(\text{CN})_6^{2-}$  redox potential. In acidic media,  $4-$  is +0.36 V, while in basic media, it is +0.45 V. [69,70].

In alkaline conditions, HCF(III) has been frequently utilised to oxidise a wide range of organic and inorganic compounds [71]. Using HCF(III), the absorbance at 420 nm was measured to explore the kinetics of the oxidation of fluoroquinolones, such as ciprofloxacin (CIP), norfloxacin (NOR), enrofloxacin (ENR), and nalidixic acid (NAL). Based on the results of this experiment utilizing

electrochemical oxidation methods, it can be concluded that the piperazine ring is the active site for the oxidation of fluoroquinolones by HCF(III) [72]

Oxidative degradation of Cefuroxime (CFA) by Hexacyanoferrate (III) [HCF (III)] in aqueous alkaline medium at 40°C temperature. The stoichiometry of the reaction indicates that the oxidation of one mole of cefuroxime requires two moles of Hexacyanoferrate (III) [73].



**Fig 1.9 : Stoichiometry of CEF / HCF III reaction**

#### 1.4.6. Diperoxidocuprate (III)

Due to its limited solubility and stability in aqueous medium, DPC [74], a flexible one-electron oxidant, is insufficient or scarce in oxidation reactions [75]. It is also employed as a reagent for analysis [76]. Malatesta [77] achieved the first chemical synthesis of Diperoxidocuprate(III) almost fifty years ago. Since then, a significant amount of study on the synthesis, structural identification, and analytical uses of this complex have been described [78].

In kinetic studies of the oxidation of different inorganic and organic substrates, Diperoxidocuprate (III) (DPC) is employed [79-82]. DPC was found to oxidize MF in an aqueous alkaline media is a fractional order reaction.

The kinetics and mechanism of oxidation of nalidixic acid (NA) by diperoxidocuprate (III)(DPC) in aqueous alkaline medium has been studied spectrophotometrically at 303 K. The reaction exhibits first order with respect to oxidant but substrate dependence is complex. The stoichiometry of the reaction has been observed to two moles of the oxidant for a mole of the substrate. According to spectroscopic evidence, 1-ethyl-2-hydroxy-1, 4-dihydro-7-methyl-4-oxo-1, and 8-naphthyridine-3-carboxylic acid are the oxidation products of the substrate[83].

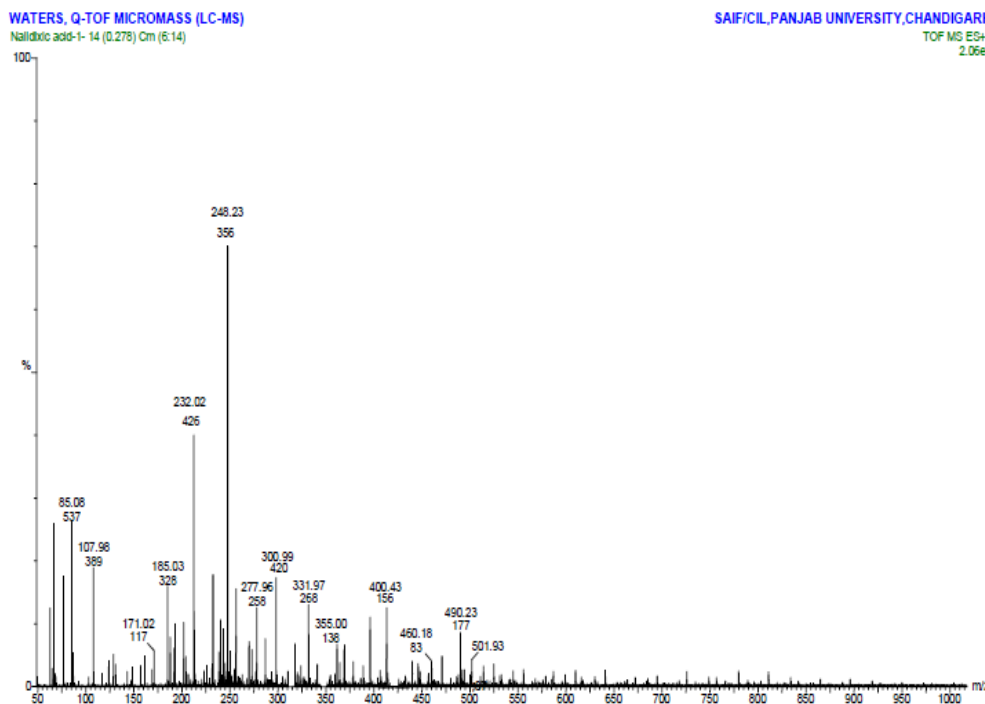


Fig1.4: LC-MS spectra of oxidation product of NA/DPC III reaction

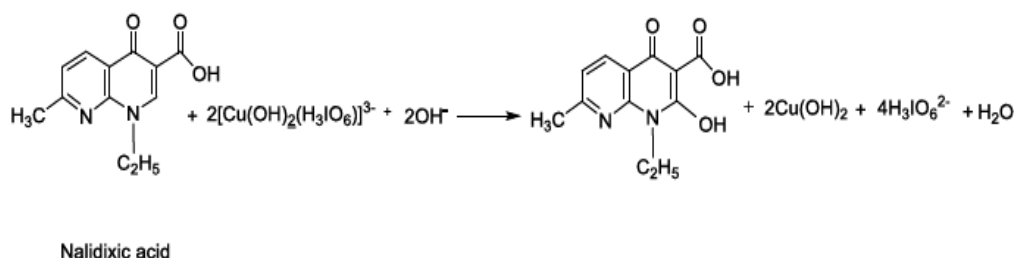


Fig. 1.10 : Stoichiometry of NA/ DPC III reaction

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