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A Comprehensive Review on the Oxidation of Inorganic Compounds by Cerium (IV): Kinetic and Mechanistic Perspectives

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Abstract : The oxidation studies with cerium(IV) is important because cerium(IV) is a significant single electron oxidant in aqueous acidic medium. This review paper investigated the kinetic and mechanistic perspective of oxidation of some inorganic compounds by cerium(IV) in aqueous medium. The present article highlights the different type of reactive species of cerium(IV) in various oxidation studies. The nature of these reactive species depends on the concentration and nature of medium used in the reaction. The order of reaction were identified with the respect of substrate, oxidants and metal ions i.e. catalyst.

Keywords: Oxidation. Inorganic Compounds, Cerium(IV), Kinetic, Mechanistic.

I. INTRODUCTION

The kinetics and mechanism of oxidation reactions from the kinetic data have been always the most intriguing subjects in chemistry. The kinetics of the oxidation of inorganic and organic compounds is highly useful in determining the different factors that influence the rate of reaction and in understanding the mechanism of chemical reactions. Oxidation of organic and inorganic compounds to give specific products by certain selective catalyst under certain conditions has received much attention from a chemical as well as an industrial point of view. The oxidation of organic compounds by Ce(IV) in general seems to proceed via the formation of an intermediate complex[**1-5]**. Among the inorganic substrates which serve as ligands for cerium (IV) are Cl^-, Br^- and hypophosphite [**6]**. Kinetics of oxidation of organic and inorganic compounds by a variety of oxidants such as chloramine-T[**7]**, H_2O_2**[8]**, potassium permanganate[**9]** and O_3[**10]**, quinolinium dichromate[**11]**, pyridiniumchlorochromate[**12],**sodium N-bromobenzenesul phonamide[**13]**, imidazolium dichromate[**14],**Cetyltrimethylammonium Dichromate[**15],**BenzimidazoliumFluorochromate[**16],** benzyltrimethylammoniumchlorobromate[**17],** iodine[**18],** chlorite[**19],** hypochlorous acid[**20],**ferrate (VI) and ferrate (V)[**21],** hypochlorite and hypochlorous acid[**22],** HCl[**23],**peroxymonosulfate[**24],** N-bromosuccinimide[**25],**Cerium (IV)[**26],** peroxomonosulphate[**27]**in acidic and basic media has been reported in literature. Out of these cerium(IV) and peroxymonosulphate oxidants are of particular importance due to their eco-friendly nature and their capability to oxidize a variety of organic and inorganic compounds catalyzed by different transition metal catalysts[**28-31]**.

Cerium (IV) as an oxidant

The accessibility of the $4f^0$ tetravalent oxidation state for molecular cerium complexes in solution is unique among the lanthanide elements. The thermodynamic basis for the isolation of molecular cerium(IV) compounds originates from the enhanced stabilityafforded by the [Xe] noble gas electronic configuration for the ion [**32**]. The existence of molecular cerium(IV) compounds and the ability to cycle between the +3 and +4 oxidation states plays a key role inthe separation of cerium from lanthanide-containing ore, and more generally in studies of valence control for nuclear fuel reprocessing [**33-35**]. Cerium is the only rare earth element with a stable +4 oxidation state in addition to +3 states. The colouredCe (IV) and the colourlessCe(III) in solution form a favourable one electron redox couple in aqueous acidic medium [**36**]. The redox potential of the Ce^{4+}/Ce^{3+} couple depends on the concentration and type of acid media used.

Cerium(IV) is a well-known oxidant in acid media [**37**] (the reduction potential [**38**] of the cerium(IV)/cerium(III) couple is 1.70 V) and is stable only in high acid concentrations.

In sulfuric acid and sulfate media cerium(IV) forms several sulfate complexes [39]. Similarly in perchloric acid solutions [**40-41**] cerium(IV) exists as Ce^{4+}, $[Ce(OH)]3^+$, $[Ce(OH_2)]^{2+}$, $[Ce–O–Ce]^{6+}$ and $[HOCe–O–CeOH]^{4+}$. A potentiometric study [**42**] of cerium(IV) hydrolysis indicates $[Ce(OH)]3^+$ to be the predominant active species of cerium(IV), in the concentration range 0.30–2.0 mol dm-3 $HClO_4$, but its role has not received much attention so far.

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Because of high stability, both kinetically and thermodynamically [**43, 44**] and easy availability, ceric sulfate has been widely used in synthetic [**45**] and mechanisticstudies [**46-50**] in aqueous H_2SO_4 media.

Actually, Ce(IV) is not so stable in aqueous $HClO_4$ andHNO_3 media [**43, 44**]. During the studies on kinetic andmechanistic aspects of Ce(IV) oxidation of differenttypes of both organic and inorganic substrates in aqueousH_2SO_4 media, the major problem arises from thestandpoint of identification of kinetically active Ce(IV)species [**46**]. Ce(IV) can exist as a mixture of differenttypes of sulfato species such as $Ce(SO_4)^{2+}$, $Ce(SO_4)^{2}$,$Ce(SO_4)_3^{2-}$ and $HCe(SO_4)_3^{-}$ in aqueous H_2SO_4 media.Generally, from the bisulfate dependence of rate atfixed $[H^+]$, attempts [**51-52**] have been made to identifythe kinetically active Ce(IV) species. Because ofso many interdependent equilibria involving Ce(IV),SO_4^{2-} ,HSO_4^- , and $[H^+]$, e.g. Ce(IV)–HSO_4^-–H+; SO_4^{2-} –HSO_4^- –H+, it is very very difficult [**53**] to computethe actual concentration of $[HSO_4^-]$ and [H+] underthe experimental conditions. In fact, so many approximationsare required to compute $[HSO_4^-]$ and $[H^+]$ and it leads to the uncertainty to identify the kineticallyactive Ce(IV) species from the dependence ofrate on $[HSO_4^-]$ and $[H^+]$. In fact, several conflicting results are presented in the literature. Hintz and Johnson [**54**] have reported the reactivity order,$Ce(SO_4)^{2+}$ _ $Ce(SO_4)_2$ – $Ce(SO_4)_3^{2-}$ in the oxidationof cyclic alcohols. In the oxidation of reducingsugars by Ce(IV) in aqueous sulfuric acid media, thereis also a conflict among the workers [**55,56**] regardingthe kinetically active Ce(IV) species.

In sulphuric acid and sulphate media Ce(IV) forms several sulphate complexes, but their role has not received much attention so far. The slow cerium(IV) oxidation reaction of many inorganic compounds are facilitated by a minute amount of metal ions in aqueous sulphuric acid medium. [**57]** .In this process the redox potential of different couples involved in the oxidation reaction in acidic medium make the metal ions catalysis for oxidation of inorganic compounds by cerium (IV) appear feasible.

II. REACTIVE SPECIES OF CERIUM (IV)

According to Hardwick and Robertson**[58],** Cerium(IV) exists in various forms insulphuric acid solution (2 M) and constant ionic strength, the following equilibrium exists.

$$Ce^{4+} + HSO_4^- \overset{\beta_1}{\rightleftharpoons} Ce(SO_4)^{2+} + H^+ \qquad \beta_1 = 3500$$

$$Ce(SO_4)^{2+} + HSO_4^- \overset{\beta_2}{\rightleftharpoons} Ce(SO_4)_2 + H^+ \qquad \beta_2 = 200$$

$$Ce(SO_4)_2 + HSO_4^- \overset{\beta_3}{\rightleftharpoons} HCe(SO_4)_3^- \qquad \beta_3 = 20$$

The value of equilibrium constants are $Q_3 = 3.5\times10^3$ (25°C), $Q_2 = 2\times10^2$ (25°C) and Q_3= 0.6 (at=20^{0}C). Without keeping the ionic strength constant, Buganko and KaunLin[59] investigated the nature of cerium(IV) spectrophotometrically in 0.1 to 1.76 M sulphuric acid. The equilibrium at 20±2°C is shown below.

$$Ce(SO_4)_2 + HSO_4^- \overset{k'}{\rightleftharpoons} HCe(SO_4)_3^- \qquad k' = 0.6 \pm 1$$

$$HCe(SO_4)_3^- + HSO_4^- \overset{k''}{\rightleftharpoons} HCe(SO_4)_3^- \qquad k'' = 2.0 \pm 1$$

They have suggested that $Ce(SO_4)_2$ and $HCe(SO_4)_3^-$ are the predominant species up to 2M and $H_3Ce(SO_4)_4$ exists above 2M H_2SO_4.Hargreves and Sutcliffhave**[60]** suggested in addition the following equilibrium to explain mechanism, of certain reactions.

$$Ce(SO_4)_4^{4-} + H^+ \overset{K_1}{\rightleftharpoons} HCe(SO_4)_4^{3-}$$

$$HCe(SO_4)_4^{3-} + H^+ \overset{K_2}{\rightleftharpoons} H_2Ce(SO_4)_4^{2-}$$

$$H_2Ce(SO_4)_4^{2-} + H^+ \overset{K_3}{\rightleftharpoons} H_3Ce(SO_4)_4^-$$

$$H_3Ce(SO_4)_4^- + H^+ \overset{K_4}{\rightleftharpoons} H_4Ce(SO_4)_4$$

But the equilibrium constants were not calculated, though they may be useful in some mechanisms. In addition to the above equilibrium, the following were also proposed**[61]**

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$$Ce(SO_4)_2 + H_2SO_4 \underset{}{\overset{K_5}{\rightleftharpoons}} H_2Ce(SO_4)_3$$

$$H_2Ce(SO_4)_3 + H_2SO_4 \overset{K_6}{\rightleftharpoons} H4Ce(SO_4)_4$$

ZhaiYoung quin et al.**[62]**and Bai W. S. and Coworkers**[63]** have reported $Ce(SO_4)_2$as the kinetically active species of cerium(IV) in sulphuric acid medium in the oxidation of ethanol amine and ethylene glycol respectively. On the other hand Tripathy, S. N., Prasad, R. K. [**64]** and Kushwaha, U., & Singh, A. K.[65]have suggested reactive species as $Ce(SO_4)_3^{2-}$. While S. K. Mishra and Y. K. Gupta[66] have shown that $CeSO_4^{2+}$is the active species of cerium(IV), Hinz, H. L., and Johnsonr**[67]**also suggested $CeSO_4^{2+}$ as the reactive species in the oxidation study of cyclic alcohol by cerium(IV) in mixed pecrchloric and sulphuric acid medium.

III. OXIDATION STUDIES OF INORGANIC COMPOUNDS WITH CERIUM (IV)

To understand the mechanistic aspects of reduction of Ce(IV) to Ce(III), several kinetic studies of Ce(IV) sulphateoxidation of different types of organic substrates have been carried out by different workers **[68-70].**A number of inorganic ions are oxidised by Ce(IV), but none of them serves as ligand to Ce +4 and it was to investigate this possibility that hypophosphite was employed for the reduction of Ce(IV). Bernhart**[71]** has reported that hypophosphite can be quantitatively oxidised by Ce(IV).

The Kinetic study of oxidation of antimony (III) by cerium(IV) in presence of micro amounts of iodide and chloride in an aqueous sulphuric acid medium have been studied spectrophotometrically at 25°C and I=3.0mol dm^{-3}**[72].** The stoichiometry of the reaction is 1:2, which shows that one mole of antimony (III) requires two moles of cerium(IV). In the presence of chloride the order of reaction was unity with respect to cerium(IV) and halide, whereas in the presence of iodide the order with respect to cerium(IV) was unity and with respect to iodide was more than one. The order with respect to antimony (III) is less than unity in presence of both halide ions. Increased sulphuric acid concentration increased the rate of reaction. The order with respect to $[H^+]$ was less than unity. The active species of cerium (IV) was understood to be $H_3Ce(SO_4)_4^-$,whereas in the presence of chloride the active species of reductant was SbCl3 and in the case of iodide the reactive species was SbI_2.

The oxidation of antimony(III) by cerium(IV) in the presence of bromide in sulphuric acid medium has been studied**[73].** The active species of cerium(IV) is identified as $H_3Ce(SO_4)_4^-$, whereas that of reluctant as $SbBr_2^+$.

The oxidation of antimony(III) by cerium(IV) has been studied spectrophotometrically in the presence of a minute amount of Mn(II) in aqueous sulphuric acid medium**[74].** The stoichiometry of the reaction showed that one mole of antimony(III) required two mole of cerium(IV). The reaction was first order with respect to Ce(IV) and Mn(II). The order with respect to antimony (III) was less than unity. The rate of reaction decreased with increased concentration of sulphuric acid. The added product sulphate and bisulphate decreases the rate. The active species of Ce(IV), Sb(III) and Mn(II) was $Ce(SO_4)_2$, $[Sb(OH)(HSO_4)]^+$ and $[Mn(H_2O)_4]^{2+,}$ respectively.

The kinetics of oxidation of antimony(III) by cerium(IV) in perchloric acid medium has been reported**[75].** The varying of H^+ concentration the rate first increases, reaches maximum at $[H^+]$ = 0.15 M then decreases as the concentration of hydrogen ion increases. the active species of oxidant and reductant in perchloric acid medium are assumed to be the hydrolytic species of Ce(IV) and SbO^+ based on the effect of H^+ concentration and spectrophotometric measurements.

The oxidation of arsenic by cerium (IV) catalysed by chromium(III) has been studied in aqueous sulphuric acid medium**[76].** The rate of reaction was increase with increasing the concentration of sulphuric acid in both catalysed and uncatalysed condition. The active species of cerium(IV) is understood to be the $H_3Ce(SO_4)_4^-$.

S. M. Tuwar and coworkers**[77]** has been reported the oxidation of mercury(I)catalysed by Pd(II) in aqueousperchloric acid medium. The stoichiometry resultsshowed that one mole of Ce(IV) required one mole of Hg(I). The active species of oxidant and catalyst are found to be $HCe(SO_4)_3^-$ and $PdCl^+$respectively.

The Kinetic study of oxidation of selenium(IV) by cerium(IV) in perchloric acid medium has been reported by Dikshituluand coworkers**[78].** They observed that the added product cerium(III) retards the reaction. In the presence of cerium(III), the order with respect to cerium(IV) found to be between one and two, while in the absence of cerium(III) the order with respect to cerium(IV) was first order and with respect to selenium(IV) was fractional order.

Ru(III) catalysed oxidation of selenium(IV) by Ce(IV) in perchloric acid medium also reported by Dikshitulu and coworkers**[79].** In the another study of oxidation of tellurium (IV) by cerium(IV) in perchloric**[80],** nitric and sulphuric acid medium**[81],** The added product cerium(III) retard the reaction while other product tellurium(VI) accelerated the reaction. The order of reaction with respect to cerium(IV) was first order and with respect totellerium (IV) was fractional.

K. A Thabajat. al**[82]** reported the oxidation of palladium(II) by cerium(IV)in perchloric acid medium at 40°C. The reaction was reported to be first order each inCe(IV) and Pd(II). The order with respect to [H^+] was fractional. The active species of oxidant and reluctant are respectively$H_3Ce(SO_4)_4$ and [$PdCl_4$].

S. A.Chimatdar andcoworkers**[83]** studied the Ru(III) catalyzed oxidation of thallium(I) by cerium(IV) in aqueous sulphuric acid medium. The reaction was reported to be first order in cerium(IV) as well as in Ru(III) and the order in thallium(I) varies from zero to unity. Increase in acid concentration accelerates the reaction rate.The added productscerium(III) and thallium (III) has a negligible effect on the reaction rate.

B. P.Sinha and H. P.Mathurreported**[84]** the Os(VIII) catalysed oxidation thallousion by ceric ion in sulfuric acid medium. The order with respect to both ceric ion and thalluous ion noticed unity. The rate of reaction directly proportional to the concentration of catalyst.The HSO_4^- inhibits the reaction and the rate of reaction varies inversely as the square of bisulphate ion concentration. Thallic and cerious ion does not have any effect on the rate.Another reaction of oxidation of Ti(I) by cerium(IV) in presence of Os(VIII) catalyst in perchloric acid medium reported by B. P.Sinha and H. P.Mathur**[85].**In this case the order of the reaction is found to be a function of both Ti(I) and Ce(IV). Hydrogen ion inhibits the reaction rate.

Oxidation of hyphophosporous acid by cerium(IV) in aqueous nitric**[86]**medium has been reported. The kinetic studies of oxidation of Fe(II)[**87],** chloride**[88],** bromide**[89],** hypophosphite**[90],**bromous acid[91], Cr(III)[92], As(III)[93] (in presence of iodide) by cerium(IV) have been reported.

IV. CONCLUSION

Cerium (IV) in redox reactions reacts as an outer sphere or inner sphere mechanism in agreement with the type of the reluctant. This review study has correlated the different factors to control the mechanistic paths of catalysis in the title oxidation reactions. Several kinetically active cerium(IV) species like $Ce(SO_4)_2$, $CeSO_4^{2+}$, $Ce(SO_4)_3^{2-}$, $HCe(SO_4)_3^-$, $H_3Ce(SO_4)_4$ were involved in the oxidation reaction of different type of inorganic and organic compounds. The effect of substrates, oxidants, metal ions (as catalyst) and added products on the rate of reaction have been reported and observed that reaction rate varies with varying the concentration of these reagents.

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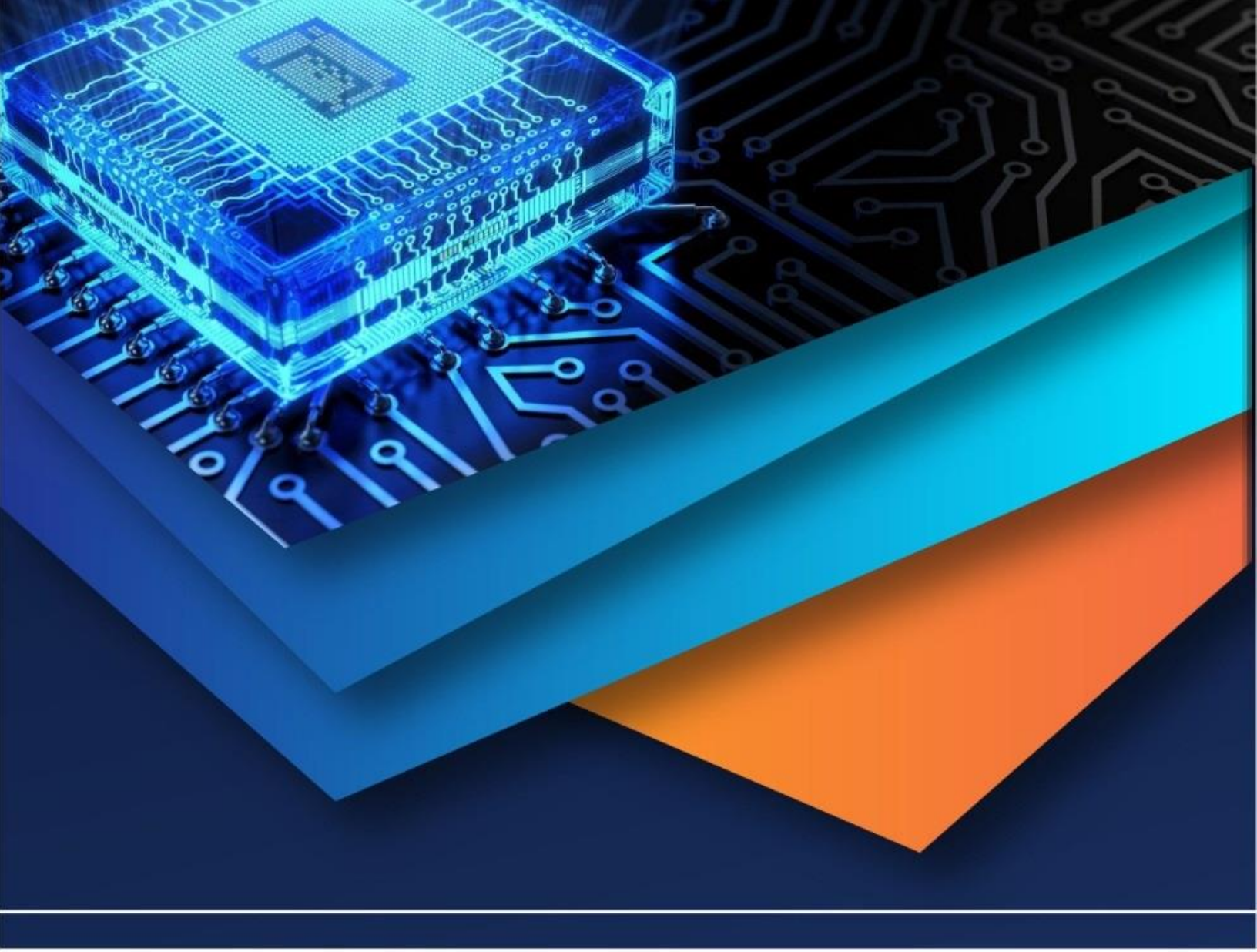

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