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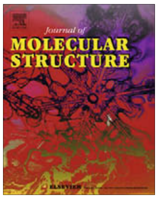

# Oxidative study of gabapentin by alkaline hexacyanoferrate(III) in room temperature in presence of catalytic amount of Ru(III) – a mechanistic approach

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ABSTRACT

The kinetics of oxidation of gabapentin by hexacyanoferrate(III) in aqueous alkaline medium at a constant ionic strength of 0.5 mol $dm^{-3}$ was studied spectrophotometrically. The reaction is of first order in [HCF(III)] and of less than unit order in [alkali]. The reaction rate is independent upon [gabapentin]. Effects of added products, ionic strength and dielectric constant of the reaction medium have been investigated. Oxidative product of gabapentin was identified. A suitable mechanism has been proposed. The reaction constants involved in the different steps of mechanism are calculated. The activation parameters of the mechanism are computed and discussed.

## 1. Introduction

Hexacyanoferrate(III) is a weak oxidant; has been widely used to oxidize numerous organic and inorganic compounds in alkaline media. The authors [1,2] have suggested that alkaline hexacyanoferrate(III) ($[Fe(CN)_6]^{3-}$) ion simply acts as an electron-abstracting reagent in redox reactions. However, Speakman and Water [3] have suggested different paths of oxidation of aldehydes, ketones and nitroparaffines. Singh and co-workers [4,5], while discussing the oxidations of formaldehydes, acetones and ethyl methyl ketone have suggested that the oxidation takes place *via* an electron transfer process resulting in the formation of a free radical intermediate. The polarizable cyano ligands around the central atom in ferricyanide facilitate electron transfer from electron rich substrates like gabapentin to the iron.

1-(aminomethyl)cyclohexaneaceticacid, commercially called gabapentin (GAP), an amino acid is an anticonvulsant that is chemically unrelated to any other anticonvulsant [6] or mood regulating medication. It is also widely used to treat individuals suffering from many kinds of pain problems, tremors, restless legs syndrome, hot flashes associated with menopause, and various psychiatric disorders. Gabapentin seems to be effective in some people with bipolar mood disorders that have not responded to lithium or other mood-stabilizers [7].

The study of neuroleptic drugs becomes important because of their biological significance and selectivity towards the oxidant to yield different products. Gabapentin is prescribed – usually in combination with other medications for the prevention of seizure in people suffering from seizure disorders. It is sometimes prescribed for the management of neuralgia (nerve pain). Its oxidative study is reported [8,9] using DPC and DPN where in the results were entirely different. The oxidation of many organic compounds using coordination compounds is also well documented [10,11].

Ruthenium(III) is known to be an efficient catalyst in several redox reactions particularly in alkaline medium [12,13]. The mechanism of catalysis can be quite complicated due to the formation of different intermediate complexes, free radicals and different oxidation states of ruthenium. A micro amount of ruthenium(III) is sufficient to catalyze the reaction in alkaline medium and a variety of mechanisms are possible. Thus, to explore the mechanism of oxidation by hexacyanoferrate(III) in aqueous alkaline medium and to check the selectivity of gabapentin towards hexacyanoferrate(III) in catalyzed system, we have selected ruthenium(III) as a catalyst. Hence, we have undertaken a detailed study of the title reaction is to arrive at a plausible mechanism.

## 2. Experimental

### 2.1. Materials and reagents

Reagent grade chemicals and double distilled $H_2O$ were used throughout this work. The stock solution of gabapentin (s.d. finechem.) was prepared by dissolving a known amount of the sample in distilled water. A solution of $[Fe(CN)_6]^{3-}$ was prepared by dissolving $K_3[Fe(CN)_6]$ (BDH) in $H_2O$ and standardized [14a] by adding 10% KI solution in presence of zinc sulphate and the iodine

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liberated was titrated with standard sodium thiosulphate solution. The product, $[Fe(CN)_6]^{4-}$ solution was prepared by dissolving pure sample of $K_4[(Fe(CN)_6]$ and its concentration was checked [14b] volumetrically by titrating against standard ceric ammonium sulphate solution in sulphuric acid medium; *N*-phenylantranilic acid was used as an indicator. Sodium hydroxide and sodium perchlorate (BDH, AnalaR) were employed to maintain the required alkalinity and ionic strength respectively in reaction solutions. The ruthenium(III) solution was prepared by dissolving a known weight of $RuCl_3$ (s.d. fine-chem.) in 0.20 mol dm$^{-3}$ HCl. Mercury was added to the ruthenium(III) solution to reduce any ruthenium(IV) formed during the preparation of the ruthenium(III) stock solution and was used after a day. The ruthenium(III) concentration was assayed [15] by EDTA titration.

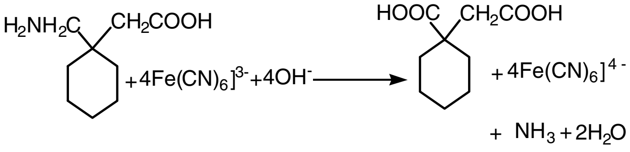
### 2.2. Kinetics measurements

The oxidation of gabapentin by hexacyanoferrate(III) was followed under pseudo-first order conditions where [GAP] was in excess over $[Fe(CN)_6]^{3-}$ at 25 ± 0.1 °C unless and other wise stated. The reaction was initiated by mixing the required quantities of previously thermostatted solution of $[Fe(CN)_6]^{3-}$ and gabapentin which also contained known quantities of ruthenium(III), sodium hydroxide and sodium perchlorate used to maintain the required concentrations of catalyst, alkalinity and ionic strength respectively. Progress of the reaction was followed by measuring the absorbance of unreacted hexacyanoferrate(III) spectrophotometrically at 420 nm ($\varepsilon$ = 1060 dm$^3$ mol$^{-1}$ cm$^{-3}$); other constituents of the reaction mixture do not absorb significantly at this wavelength. Application of Beer's law under the reaction conditions had been verified earlier between $1.0 \times 10^{-4}$ and $1.0 \times 10^{-3}$ mol dm$^{-3}$ of $[Fe(CN)_6{}^{3-}]$ ($\varepsilon = 1060 \pm 20$ dm$^3$ mol$^{-1}$ cm$^{-1}$). The first order rate constants, ($k_{obs}$) were obtained from the plots of log $[Fe(CN)_6{}^{3-}]$ *versus* time. The rate constants were reproducible within ±5%.

## 3. Results and discussions

### 3.1. Stoichiometry and product analysis

Different sets of reaction mixtures with different concentrations of reactants were kept for 6 h at 25 ± 0.1 °C under nitrogen atmosphere in a closed vessel. The remaining $[Fe(CN)_6]^{3-}$ was assayed spectrophotometrically by measuring the absorbance at 420 nm. The product $[Fe(CN)_6]^{4-}$ was analyzed by titrating against Ce(IV) solution [14b]. The results indicated that four moles of hexacyanoferrate(III) were reacted with one mole of gabapentin as in equation (1)



(1)

The main oxidative products were submitted to spot tests. The main reaction product was identified as 1-(carboxymethyl) cyclohexanecarboxylic acid by spot test [16] for free dicarboxyl groups. The product was extracted with ether and purified. The IR spectrum of the compound revealed the presence of two carboxylic groups, one aliphatic and other cyclo alkane. Another product, ammonia was identified [14c] by spot test. It was observed that, the products do not undergo further oxidation under prevailing kinetic conditions.

### 3.2. Reaction orders

The order with respect to [GAP], [Ru(III)] and [alkali] were found from the graph of log $k_{obs}$ *versus* log(concentration) plots.

The concentration of hexacyanoferrate(III) was varied in the range, $8.0 \times 10^{-5}$ to $8.0 \times 10^{-4}$ mol dm$^{-3}$ at constant [GAP], [Ru(III)], [OH$^-$] and ionic strength. The non-variation in pseudo-first order rate constants at various concentrations of hexacyanoferrate(III) indicates, the order in $[Fe(CN)_6]^{3-}$ as unity. The linearity in the plot of log(concentration) *versus* time also supports the order in $[Fe(CN)_6]^{3-}$ (Table 1).

The substrate, gabapentin was varied in the range of $1.0 \times 10^{-3}$ to $1.0 \times 10^{-2}$ mol dm$^{-3}$ at 25 °C while keeping all other reactants concentrations and conditions constant. The order of reaction was found to be independent on [GAP]. Hence, its order is zero (Table 1).

The effect of [alkali] on the rate of reaction was studied at constant concentrations of gabapentin, ruthenium(III) and hexacyanoferrate(III) at a constant ionic strength of 0.50 mol dm$^{-3}$ (Table 2). The rate constants were increased with the increase in [alkali] and found that an apparent less than unit orders dependence on [alkali] (≈0.6).

The catalyst, ruthenium(III), was varied in the concentration range of $5.0 \times 10^{-7}$ to $5.0 \times 10^{-6}$ mol dm$^{-3}$ at constant [GAP], $[Fe(CN)_6]^{3-}$ and [OH$^-$] at ionic strength of 0.50 mol dm$^{-3}$ (Table 2). The order with respect to [Ru(III)] was found to be unity.

**Table 1**
Effect of variation of $[Fe(CN)_6]^{3-}$ and [gabapentin] on ruthenium(III) catalyzed oxidation of gabapentin by hexacyanoferrate(III) in aqueous alkaline medium at 25 °C

| $[Fe(CN)_6]^{3-} \times 10^4$ (mol dm$^{-3}$) | [GAP] $\times 10^3$ (mol dm$^{-3}$) | $k_{obs} \times 10^3$ (s$^{-1}$) |
|---|---|---|
| 0.8 | 4.0 | 2.89 |
| 1.0 | 4.0 | 2.84 |
| 2.0 | 4.0 | 2.88 |
| 4.0 | 4.0 | 2.90 |
| 6.0 | 4.0 | 2.81 |
| 8.0 | 4.0 | 2.84 |
| 4.0 | 1.0 | 2.65 |
| 4.0 | 2.0 | 2.89 |
| 4.0 | 4.0 | 2.90 |
| 4.0 | 6.0 | 2.85 |
| 4.0 | 8.0 | 2.70 |
| 4.0 | 10 | 2.78 |

[OH$^-$] = 0.10; [Ru(III)] = $1.0 \times 10^{-6}$; $I$ = 0.50/mol dm$^{-3}$.

**Table 2**
Effect of variation of [Ru(III)] and [OH$^-$] on ruthenium(III) catalyzed oxidation of gabapentin by hexacyanoferrate(III) in aqueous alkaline medium at 25 °C

| [Ru(III)] $\times 10^6$ (mol dm$^{-3}$) | [OH$^-$] (mol dm$^{-3}$) | $k_{obs} \times 10^3$ (s$^{-1}$) | |
|---|---|---|---|
| | | Found | Calculated* |
| 0.5 | 0.10 | 1.76 | 1.50 |
| 0.8 | 0.10 | 2.51 | 2.38 |
| 1.0 | 0.10 | 2.89 | 2.97 |
| 2.0 | 0.10 | 5.90 | 5.95 |
| 4.0 | 0.10 | 11.9 | 11.90 |
| 5.0 | 0.10 | 15.0 | 14.90 |
| 1.0 | 0.05 | 1.87 | 1.83 |
| 1.0 | 0.08 | 2.56 | 2.57 |
| 1.0 | 0.10 | 2.89 | 2.97 |
| 1.0 | 0.20 | 4.21 | 4.20 |
| 1.0 | 0.40 | 5.84 | 5.61 |
| 1.0 | 0.50 | 6.20 | 6.00 |

[Fe(CN)6]$^{3-}$ = $4.0 \times 10^{-4}$; [GAP] = $4.0 \times 10^{-3}$; $I$ = 0.50/mol dm$^{-3}$.
*'$k_{obs}$' are calculated using rate law (6) and '$k$' and '$K$' were $7.94 \times 10^3$ dm$^3$ mol$^{-1}$ s$^{-1}$ and 6.0 dm$^3$ mol$^{-1}$, respectively.

### 3.3. Effect of $[Cl^-]$

Since, ruthenium chloride was used as the catalyst, the effect of $Cl^-$ on rate of reaction was studied. The concentration of KCl was varied in the range of $1.0 \times 10^{-5}$ to $1.0 \times 10^{-4}$ mol dm$^{-3}$ keeping all other reactants concentration and conditions constant. It was found that the added chloride did not affect the rate of reaction.

### 3.4. Effect of ionic strength and dielectric constant

The effect of ionic strength was studied by varying the sodium perchlorate concentration from 0.50 to 1.50 mol dm$^{-3}$ at constant concentrations of hexacyanoferrate(III), gabapentin, ruthenium(III) and alkali. It was found that the rate constants were increased with increase in concentration of $NaClO_4$ and the plot of $\log k_{obs}$ *versus* $\sqrt{I}$ was linear with positive slope (0.6) (Fig. 1) ($r > 0.9888$, $S < 0.017$).

The dielectric constant ($D$) effect was studied by varying the *t*-butyl alcohol–water content in the reaction mixture with all other conditions being maintained constant. The dielectric constants were computed from the values of pure liquids [17,18]. The solvent did not react alone with oxidant and catalyst or in the mixture of both under the experimental conditions. The rate constants, $k_{obs}$ were increased with decrease in the dielectric constant of the medium. The plot of log $k_{obs}$ *versus* $1/D$ was linear with positive slope (Fig. 1) ($r > 0.9983$, $S < 0.0341$).

### 3.5. Effect of initially added products

The initially added product such as hexacyanoferrate(II) was studied in the concentration range from $4.0 \times 10^{-5}$ to $4.0 \times 10^{-4}$ mol dm$^{-3}$ keeping all other reactant concentrations constant. It was found that added product had a negligible effect on the rate of reaction.

### 3.6. Polymerization study

The reaction mixture was mixed with acrylonitrile, a monomer and kept for 2 h in an inert atmosphere. On diluting with methanol there was no precipitation formed, indicating the intervention of free radicals in the reaction was absent.

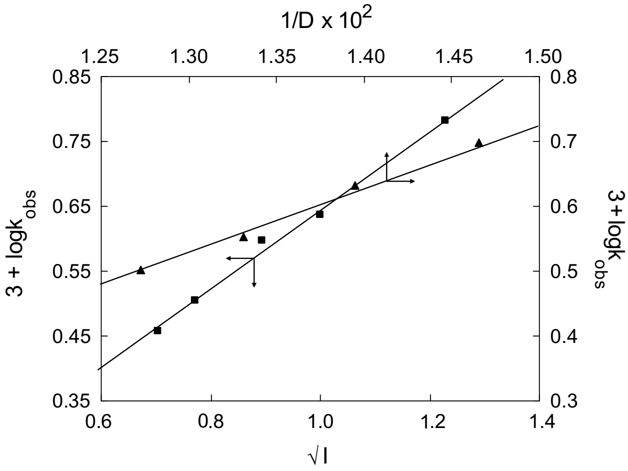


**Fig. 1.** Effect of variation of ionic strength (I) and dielectric constant on the ruthenium(III) catalyzed oxidation of gabapentin by hexacyanoferrate(III) in aqueous alkaline medium at 25 °C.

### 3.7. Effect of temperature

The rate of reaction was measured at different temperatures at 30, 35, 40 and 45 °C with $[Fe(CN)_6]^{3-} = 4.0 \times 10^{-4}$; $[GAP] = 4.0 \times 10^{-3}$; $[Ru(III)] = 1.0 \times 10^{-6}$; $[OH^-] = 0.10$ and $I = 0.50$/mol dm$^{-3}$. The rate was found to increase with increase in temperature. The first order rate constants $k_{obs} \times 10^3\ s^{-1}$ were obtained as 2.89, 3.33, 3.90 and 4.58 at 30, 35, 40 and 45 °C, respectively. These values lead to activation parameters as $E_a = 23.8 \pm 0.2$ kJ mol$^{-1}$, $\Delta H^{\#} = 21.4 \pm 0.2$ kJ mol$^{-1}$, $\Delta S^{\#} = -221 \pm 10$ JK$^{-1}$ mol$^{-1}$, $\Delta G^{\#} = 88 \pm 4$ kJ mol$^{-1}$ and $\log A = 1.63 \pm 0.2$.

Ruthenium(III) is known [19,20] to catalyze many oxidation reactions both in acid and alkaline media. Its catalytic ability is due to the exhibition [21] of multivalent states as −2, −1, 0, +1, +2, +3, +4, +5, +6, +7 and +8. Nevertheless, ruthenium compounds are widely used as $RuCl_3$. Its aqueous solutions are normally prepared in HCl and hence, Ru(III) is expected to be existed in its chloride complexes [21] viz., $[Ru(H_2O)_5Cl]^{2+}$, $[Ru(H_2O)_4Cl^2]^+$, $[Ru(H_2O)_2Cl_4]^-$ and $[RuCl_6]^{3-}$ in addition to its aqueous species as $[Ru(H2O)_6]^{3+}$. The chloride complexes are possible [22] only when on heating or aging in presence of higher concentration of HCl. In the present study the fresh solutions are used and solutions are prepared in low concentration of HCl. Hence, the chloride complexes are excluded. Alternatively it may be in the form of $[Ru(H_2O)_6]^{3+}$. This has been supported by its UV–vis spectrum which was identical with reported study [21] for $[Ru(H_2O)_6]^{3+}$.

However, in alkaline medium, ruthenium(III) is known [23] to exist as its hydroxylated species with general formula $[Ru(OH)_x(H_2O)_{6-x}]^{3-x}$, where $x < 6$ and variable depending upon the $[OH^-]$ used. Under the conditions $[OH^-] > [Ru(III)]$, it is mainly present as $[Ru(H_2O)_5(OH)]^{2+}$ as shown in equation (2),

$$[Ru(H_2O)_6]^{3+} + OH^- \overset{K}{\rightleftharpoons} [Ru(H_2O)_5(OH)]^{2+} \tag{2}$$

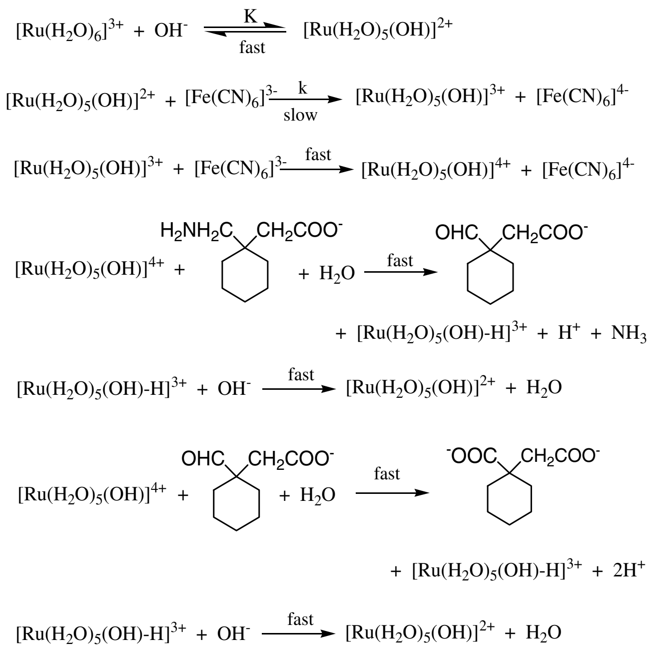
Hence, active species of ruthenium(III) has been considered as $[Ru(H_2O)_5(OH)]^{2+}$, which was supported by the positive fractional order in $[OH^-]$. The zero order dependency on [GAP], first order each on $[Fe(CN)_6]^{3-}$ and [Ru(III)] and fractional order in $[OH^-]$ can be accommodated in the following scheme.

Mechanism written as in scheme 1 is in accordance with the fact that ruthenium is known to be better catalyst in which hydride ion [24] is during oxidation of substrate.

The results of absence of intervention of free radical also indicate that the interaction between substrate and one equivalent oxidant is ruled out. The zero order dependency on [substrate] also supports the reaction between substrate and catalyst in fast steps. The interaction between Ru(IV) and GAP is very remote because of the fact that such interaction leads intervention of free radical, which is obscure. The similar [15] mechanism has been proposed for Ru(III) catalyzed oxidation of DMSO by $[Fe(CN)_6]^{3-}$. Since, the order with respect to catalyst and oxidant is one each and increase in rate with increase in ionic strength explains the interaction between Ru(III) and $[Fe(CN)_6]^{3-}$ through inner sphere mechanism wherein the electrons are transferred through a common bridging ligand $CN^-$ from Ru(III) to Fe(III).

The product, $[Fe(CN)_6]^{4-}$ is generally known to retard the rate of reaction. However, in Ru(III) catalysis its effect has not been observed which may be due to the formation of highly reactive intermediates Ru(IV) or Ru(V). The unaltered in the rate by adding $Cl^-$ can also be explained on the basis of formation Ru(III)-chloride complexes which are very remote as they are formed [22] only at high concentration of $Cl^-$.

Increasing the dielectric constant ($D$) of the reaction medium decrease in rate was observed. This might be due to the stabilization of activated complex at high dielectric constant of the medium. The large negative value of $\Delta S^{\#}$ and large positive value



**Scheme 1.**

of $\Delta G^{\#}$ support the loss of degree of freedom. The small value of the frequency factor supports the inner sphere interaction of species.

The rate law for scheme 1 can be derived as follows.

$$\frac{-\mathrm{d}[\mathrm{Fe(CN)_6}]^{3-}}{\mathrm{d}t} = \mathrm{rate} = k[\mathrm{Fe(CN)_6}]_{\mathrm{f}}^{3-}[\mathrm{Ru(H_2O)_5(OH)}]_{\mathrm{f}}^{2+} \quad (3)$$

$$k_{\mathrm{obs}} = k[\mathrm{Ru(H_2O)_5(OH)}]_{\mathrm{f}}^{2+} \quad (4)$$

After calculating the total concentration of Ru(III), $[\mathrm{Ru(III)}]_{\mathrm{T}}$ and total concentration $[\mathrm{OH^-}]_{\mathrm{T}}$ using the first equilibrium as in the scheme, $k_{obs}$ becomes

$$k_{\mathrm{obs}} = \frac{kK[\mathrm{Ru(III)}]_{\mathrm{T}}[\mathrm{OH}]_{\mathrm{T}}}{1 + K[\mathrm{OH^-}]} \quad (5)$$

The denominator of the above equation also contains the term $(1+K[\mathrm{Ru(III)}]_{\mathrm{T}})$ but it is neglected as $[\mathrm{Ru(III)}]_{\mathrm{T}}$ is very small and hence becomes unity.

The rate constant '*k*' and equilibrium constant '*K*' can be evaluated using the equation (5) after omitting the subscript T in the above equation, the equation (6) follows.

$$\frac{1}{k_{\mathrm{obs}}} = \frac{1}{kK[\mathrm{Ru(III)}][\mathrm{OH^-}]} + \frac{1}{k[\mathrm{Ru(III)}]} \quad (6)$$

The plot of $1/k_{\mathrm{obs}}$ *versus* 1/[Ru(III)] should be a straight line with zero intercept; and the $1/k_{\mathrm{obs}}$ *versus* $1/[\mathrm{OH^-}]$ plot should be non-zero intercept and found to be so in Fig. 2. From the slope and intercept of such graph, the '*k*' and '*K*' are evaluated as $k = 7.937 \times 10^3\ \mathrm{dm^3\ mol^{-1}\ s^{-1}}$ and $K = 6.0\ \mathrm{dm^3\ mol^{-1}}$. The value of '*K*' is very close to the literature value [12,13]. Hence, the mechanism is proved. The '*k*' and '*K*' derived as above are used to regenerate the $k_{\mathrm{obs}}$ values for different experimental conditions and are found to be neighborhood of experimental values (Table 2).

## 4. Summary

The title reaction is zero order dependency in substrate and first order each in oxidant and catalyst. Hence, the results suggest that oxidation of substrate occurred in the fast step. The interaction between oxidant and catalyst was proposed in the rate determining step to give Ru(IV) and further oxidizes to Ru(V) followed by oxidation of substrate; the interaction between oxidant and substrate is of inner sphere complex type which was supported by the small value of frequency factor and large value of second order rate constant ($7.9 \times 10^3\ \mathrm{dm^3\ mol^{-1}\ s^{-1}}$). The non-intervention of free radical during the reaction revealed that oxidation of substrate occurs in the complementary reaction of two equivalent change in intermediate, Ru(V) and a substrate in a single step. The catalyst was acted as hydride ion abstracting agent.

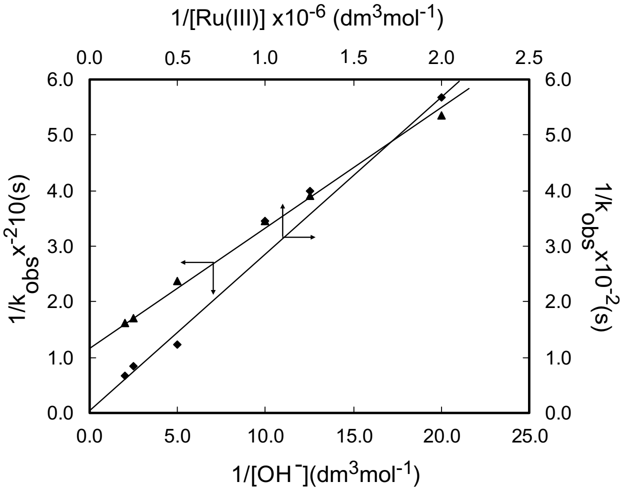


**Fig. 2.** Verification of rate law (6) of ruthenium(III) catalyzed oxidation of gabapentin by hexacyanoferrate(III) in aqueous alkaline medium at 25 °C (conditions as in Table 2).

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