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The Kinetic Study of the Solvent Effect of Polyhydric Alcohol on the Thermodynamic Extensive properties of the Catalysed Solvolysis of propionate ester

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ABSTRACT

The changes observed and evaluated due to the solvent effect of a dipolar protic or dipolar aprotic solvent on the thermodynamic extensive properties of solvolysis of the propionate ester have been found responsible for changes in its biochemical and medicinal properties.With a view to highlight the effect of a polyhydric alcohol (dipolar protic solvent) on the extensive thermodynamic properties of propionate ester, the kinetics of alkali catalysed hydrolysis of ethylpropionate was studied in aquo-glycerol reaction media.

From the enhancement observed in ΔG^* values with simultaneous decrease in the values of ΔH and ΔS^* of the reaction, it is inferred that the organic co-solvent glycerol acts as entropy controller and enthalpy stimulator solvent for alkali catalysed hydrolysis of ethyl propionate.

The numerical value of Iso-kinetic temperature of the reaction which comes to be nearly 286.0 (below 300) indicates that there is weak but considerable solvent-solute interaction in the aquo-glycerol reaction media.

Key words: Extensive properties, Trihydric alcohol, Dielectric effect Solvation and desolvation, Weak solvent -solute interacion

Introduction : Though different kinecists[1-2] of the kinetic field have reported the effect of different solvents on the rates, mechanism and the thermodynamic properties of hydrolysis of simple esters, but, a little attention has been paid towards the studies of solvent effect of polyhydric alcohol on the rate, mechanism and extensive thermodynamic properties and solvent-solute interaction for alkali catalysed solvolysis of ethyl propionate.In order to highlight the above noted facts, it has been proposed to study the kinetics of alkali catalysed hydrolysis of ethyl propionate in aquo-glycerol reaction media.

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Experimental & Calculation :

The kinetics of alkali catalysed hydrolysis of ethyl propionate was carried out separately in the different aquo-organic co-solvent media (aquo-glycerol) prepared by adding different volumes of glycerol (20 to 80%). The strength of the solution was kept 0.1 M with respect of NaOH and 0.05 M with respect to the ester. The reaction was found to follow the second order kinetic equation and the evaluated values of specific rate constants have been recorded in Table - I. For studying the effect of change of concentration of organic component (glycerol), the variation of log k values of the reaction with its mol % in the reaction media has been enlisted in Table - II.Using Arrhenius equation, the iso-composition and iso-dielectric activation energies values of the reaction were evaluated and are recorded respectively in Table - III & IV. The thermodynamic activation parameters such as ΔH^*, ΔG and ΔS^* have been evaluated using Wynne-Jones and Eyring[3] equation and their consolidated values have been shown in Table - V.For studying the mechanism of the reaction, the evaluated number of water molecules associated with the transition state of the reaction at different temperatures have been depicted in Table - VI.

Results and Discussion :

Effect of Solvent on the Specific Rate Constants of the Reaction:

In order to highlight the effect of the solvent on the specific rate constant values of the reaction, the log k values were plotted against the mole % of the organic co-solvent glycerol (their values from Table - II) as shown in Fig. - 1, and were found to follow decreasing trends. However, the depletion found in the rate with increasing mole % of the organic co-solvent (glycerol) at all the temperatures follow smooth path following two intersecting straight lines at about 16.25 mol % of glycerol having different numerical values of the slope (of similar nature) before and after the point of intersection (at about 16.25 mol % of glycerol in the reaction media). From the plots, it was found that with increasing the temperature of the reaction, the degree of depletion in the rate constants of the reaction becomes steeper.

However, the possible rate depleting factors in the rate can be listed as follows:

(i) decrease in the bulk dielectric constant value of the medium,

(ii) decrease in the polarity of the reaction media on adding less polar glycerol to it

The above noted two depleting factors are quite in operation and this is quite in agreement with the theory of Hughes and Ingold[4] that the rate ought to decrease with decreasing dielectric constant of the reaction media. Such decrease in rate constant with increasing proportion of the organic co-solvent like glycerol has been reported earlier by Laidler and Landskroener[5], Akanksha & Singh et al.[6] and Singh & Hafizee et al.[7]. In recent years, Singh, R.T.[8] and Kumar & Singh et al.[9] have also reported similar observations and inferences on the effect of solvent on the specific rate constants of catalysed solvolysis reactions. However, the decrease observed in the specific rate constant values with different numerical values of slopes may be attributed partly due to the dielectric effects of the reaction media and partly due to solvation changes taking place in it (aquo-glycerol reaction media).

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Effect of Solvent on the Iso-composition Activation Energy (E_C) of the Reaction :

On perusal of the data mentioned in Table - III, it is observed that the value of iso-composition activation energy of the reaction go on decreasing from 80.45 kJ/mol to 49.19 kJ/mol with increasing concentration of glycerol from 20 to 80% (v/v), in the reaction media.

The depletion E_C values of the reaction in aquo-glycerol media may be due to either of the following three causes :

(i) The transition state is solvated and the initial state is desolvated,

(ii) The transition state is less desolvated than the initial state, and

(iii) The transition state is more solvated than the initial state.

Among these three factors, the first factor seems to be operative in this case as from the values of thermodynamic activation parameters of the reaction in Table - V, both ΔH^* and ΔS^* values of the reaction are found to decrease with increasing proportion of glycerol in the reaction media (ΔH^* values decreases from 77.70 kJ/mol to 46.81 kJ/mol and ΔS^* values decreases from –35.71 J/K/mol to – 144.69 J/K/mol at 30°C).

Regarding effect of solvent on the E_C value of reaction, similar findings and their interpretations have been reported earlier by Singh & Parween et al.[10], Pathak & Singh et al.[11] and also recently by Choubey & Singh et al.[12].

Solvent Effect on the Iso-dielectric Activation energy (E_D) of the reaction :

From the values recorded in Table - IV, it appears that E_D values of the reaction go on increasing with increasing dielectric constant values of the aquo-glycerol reaction media. The E_D value is 61.85 kJ/mol at D value 50 and increases to 80.67 kJ/mol at D value 65. The enhancement in the E_D values with increase in D values of the reaction media is in accordance with depletion in E_C or E_{exp} values of the reaction with increasing mol % of glycerol in the reaction media. These findings and conclusions have been found in support of the earlier reports of Wolford[13], Priyanka and Singh et al.[14] and recent reports of Singh & Hafizee et al.[15].

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Solvent Effect on Thermodynamic Activation Parameters of the Reaction :

From Table - V, on perusal of the values of thermodynamic activation parameters, namely ΔG*, ΔH* and ΔS*, it is observed that ΔG* values (free energy of activation) of the reaction increases with simultaneous decrease in its ΔH* and ΔS* values. At 30°C, ΔG* values have been observed increasing from 88.52 k cal/mol to 90.65 k cal/mol with increasing concentration of glycerol from 20 to 80% (v/v) in the reaction media. Though this enhancement is not very large, however, it is quite considerable and acceptable too. In order to highlight the effect of changing concentration of the organic content (glycerol) in the reaction media, ΔH*, ΔG* and ΔS* values have been plotted against the changing mol % of glycerol in the reaction media and their plots are shown in Fig. - 2, 3 and 4 respectively.

From the plots of ΔG* values against mol % of glycerol as shown in Fig.-3, it is found that ΔG* values go on increasing non-linearly with gradual addition of glycerol in the reaction media. This finding is indicative of desolvation of reactants as explained by Elsemongy et a1.[16]

So far as the variations in ΔH* and ΔS* are concerned on observing their values from Table - V and their plots against mol % of glycerol as shown in Fig. - 2 and 4, it is interestingly found that both of them decrease linearly with gradual addition of glycerol in the reaction media.

From the thermodynamic relation:

$$\Delta G^* = \Delta H^* - T\Delta S^*$$

it can be easily concluded that increase in ΔG* values with simultaneous decrease in both of ΔH* and ΔS* values is only possible when ΔS* values decreases more than ΔH* value. From such findings, it is inferred that in presence of glycerol in the reaction media, the alkali catalysed hydrolysis of ethyl propionate becomes entropy controlled and enthalpy stimulated reaction.

Moreover, non-linear variation in ΔH* and ΔS* values with increasing mol % of glycerol as shown in Fig. - 2 and 4 respectively, gives information of the fact that specific solvation is taking place in aquo-glycerol solvent systems similar to that as reported in the past by Saville et al.[17]. Similar solvent effect on thermodynamic activation parameters and their explanations have been found in support of the earlier reports of Singh & Priyanka et al.[18], Singh & Navendu et al.[19] and also with the recent reports of Kumar & Singh et al.[20].

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Solvent Effect on Iso-kinetic Temperature and Solvent-Solute Interaction in aquo-glycerol Media :

The value of the iso-kinetic temperature of the reaction was evaluated by using Barcley- Butler[21] relationship which is expressed as

$$dm\,(\Delta H^*) = b\ dm\,(\Delta S^*)$$

It is a straight line equation representing the relationship between enthalpy and entropy of activation values of the reaction. 'b' is known as iso-kinetic temperature. From the values of ΔH^* and ΔS^* values available in Table - V, the plots of ΔH^* versus ΔS^* at 30^oC were made which is shown in Fig. - 5. From the slope of the straight line of the plots, the value of the kinetic temperature was evaluated to be $286.17 \approx 286.0$ (below 300). Thus, in the light of Leffler's guidelines[22], from the numerical values of the iso-kinetic temperature (which is below 300), it can safely be inferred that there is appreciable change in the structure of the reactant or in the solvent or in both the reactant and solvent due to weak but considerable interaction between solvent and solute present in the aquo-glycerol reaction media in the similar way as reported earlier by Singh & Singh et al.[23], Pathak & Singh et al.[24] and recently also by Kumari & Singh et al.[25] and Singh & Singh et al.[26].

Effect of number of water molecules of the reaction media on the Mechanism of the Reaction:

The number of water molecules associated with the formation of transition state or activated complex has been evaluated by plotting log k values of the reaction against log $[H_2O]$ values as shown in Fig.- 6 in the light of relation proposed by Robertson.[27]

$$\log k = \log k_0 + n \log [H_2O]$$

From Fig. - 6, it is apparent that at each temperature two straight lines intersecting each other at log $[H_2O]$ value approximately 1.485 having different values of slopes are obtained.

From the slopes of the plots of log k versus log $[H_2O]$, the evaluated values of number of water molecules associated with the activated complex of the reaction are noted in Table - VI. From Fig. - 6 as well as from Table - VI, it is clear that the number water molecules associated with the transition state increases from 0.246 to 0.757 with increase in temperature above 20 to 40^oC befor log $[H_2O]$, value 1.485 which corresponds to 55.0% of water concentration in the reaction media.Similarly, in case of above 55.0 % of water concentration in the reaction media, the number of water molecules associated with the transition state in its formation increases from 0.601 to 1.218 with rise of temperature from 20 to 40^oC. Overall, the number of water molecules involved in the formation of the activated complex of the reaction increases from 0.246 to 1.218 with rise of temperature From 20 to 40^oC.

According to observation and findings of Robertson et al.[24] it has been suggested that number of water molecules associated with the transition state is fairly high for unimolecular reaction while that for bimolecular reaction will be very low.

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Hence in the light of principle of Robrtson et al.[28], it may be inferred that with rise in temperature of the reaction, the mechanistic path of the reaction changes from bimolecular to unimolecular in aquo-glycerol media. From above noted findings about the increasing number of water molecules associated with activated complex, it may be inferred that in presence of glycerol in the reaction media and with rise of temperature of the reaction, the structure of water is changed from its dense form to bulky form at equilibrium.

$$(H_2O)_d \quad _____ \quad (H_2O)_b$$

Earlier Kumari & Singh et al.[29], Singh & Hafizee et al.[30] and recently Singh & Lal et a.[31] and Sharma& Singh et al.[32] have also reported similar findings and their interpretations for effect of solvent on the mechanism of the catalysed solvolysis reactions in different aquo-organic co-solvent reaction media

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Table – I

Specific rate constant values of Alkali Catalysed hydrolysis of Ethyl propionate in water – Glycerol media.
K x 10^3 in $(dm)^3$ mol^{-1} min^{-1}

Temp in °C	% of Glycerol (v/v)						
	20%	30%	40%	50%	60%	70%	80%
20°C	71.37	65.07	60.06	57.39	53.96	50.10	45.57
25°C	120.67	109.72	99.59	92.92	82.66	74.17	64.24
30°C	208.64	185.18	163.38	149.18	127.29	109.62	89.85
35°C	346.66	301.86	257.81	220.50	190.50	147.86	120.59
40°C	579.83	492.85	419.08	338.17	283.53	225.48	164.74

Table – II
Variation of log k values of the reaction at different temperatures with mol % of glycol in water- Glycerol media

% of Glycerol (v/v)	Mol % of Glycerol	3 + log k values				
		20°C	25°C	30°C	35°C	40°C
20%	05.81	1.8535	2.0816	2.3194	2.5399	2.7633
30%	9.56	1.8134	2.0403	2.2676	2.4798	2.6927
40%	14.12	1.7786	1.9982	2.2132	2.4113	2.623
50%	19.79	1.7588	1.9681	2.1589	2.3434	2.5299
60%	27.01	1.7321	1.9130	2.1048	2.2799	2.4526
70%	36.54	1.6998	1.8702	2.0399	2.1699	2.3531
80%	49.67	1.6587	1.8078	1.9535	2.0813	2.2168

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Table – III

Evaluated values of Iso-composition Activation Energy (E_C or E_{exp}) of the reaction in water- Glycerol media.

% of Glycerol (v/v)	20%	30%	40%	50%	60%	70%	80%
E_C value in KJ/ mol	80.45	77.14	73.78	67.92	63.79	56.38	49.16

Table – IV

Evaluated values of Iso – Dielectric Activation Energy (E_D) of the reaction at different desired 'D' values of the water – Glycerol media.

D values	D = 50	D = 52.5	D = 55	D = 57.5	D = 60	D = 62.5	D =65
E_D values in kJ/mol	61.85	65.60	67.70	71.14	73.93	77.73	80.67

Table – VI

Values of the slopes of the plots of log k versus log [H_2O] at different temperatures.

Temperature in°C	Slope – l When log [H_2O] values is below 1.485	Slope – ll When log [H_2O] values is below 1.485
20°C	0.246	0.601
25°C	0.371	0.710
30°C	0.505	0.931
35°C	0.694	1.032
40°C	0.757	1.218

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Table - V

Consolidated Values of Activation parameters (ΔH*, ΔG* and ΔS*) of the reaction in water-Glycerol system at different temperatures.

ΔH* and ΔG* in kJ/mol, ΔS* in J/K/mol

% of Glycerol (v/v)	Mole % of Glycerol	Δ H* in kJ/mol	20⁰ C		25⁰ C		30⁰ C		35⁰ C		40⁰ C	
			Δ G*	Δ S*	Δ G*	Δ S*	Δ G*	Δ S*	Δ G*	Δ S*	Δ G*	Δ S*
20 %	05.81	77.70	88.13	-35.60	88.38	-35.84	88.52	-35.71	88.73	-35.81	88.87	-35.69
30%	9.56	76.39	88.36	-41.85	88.61	-41.71	88.82	-41.02	89.08	-41.20	89.29	-41.21
40%	14.12	72.11	88.55	-56.11	88.85	-56.17	89.14	-56.20	89.48	-56.40	89.71	-56.23
50%	19.79	66.40	88.66	-75.97	89.03	-76.98	89.45	-75.05	89.88	-76.23	90.26	-76.23
60%	27.01	60.25	88.82	-97.51	89.34	-97.62	89.77	-97.43	90.26	-97.44	90.73	-97.38
70%	36.54	55.48	89.00	-114.40	89.58	-114.43	90.14	-114.39	91.01	-115.36	91.33	-114.54
80%	49.67	46.81	89.23	-144.78	89.94	-144.73	90.65	-144.69	91.43	-144.87	92.14	-144.82

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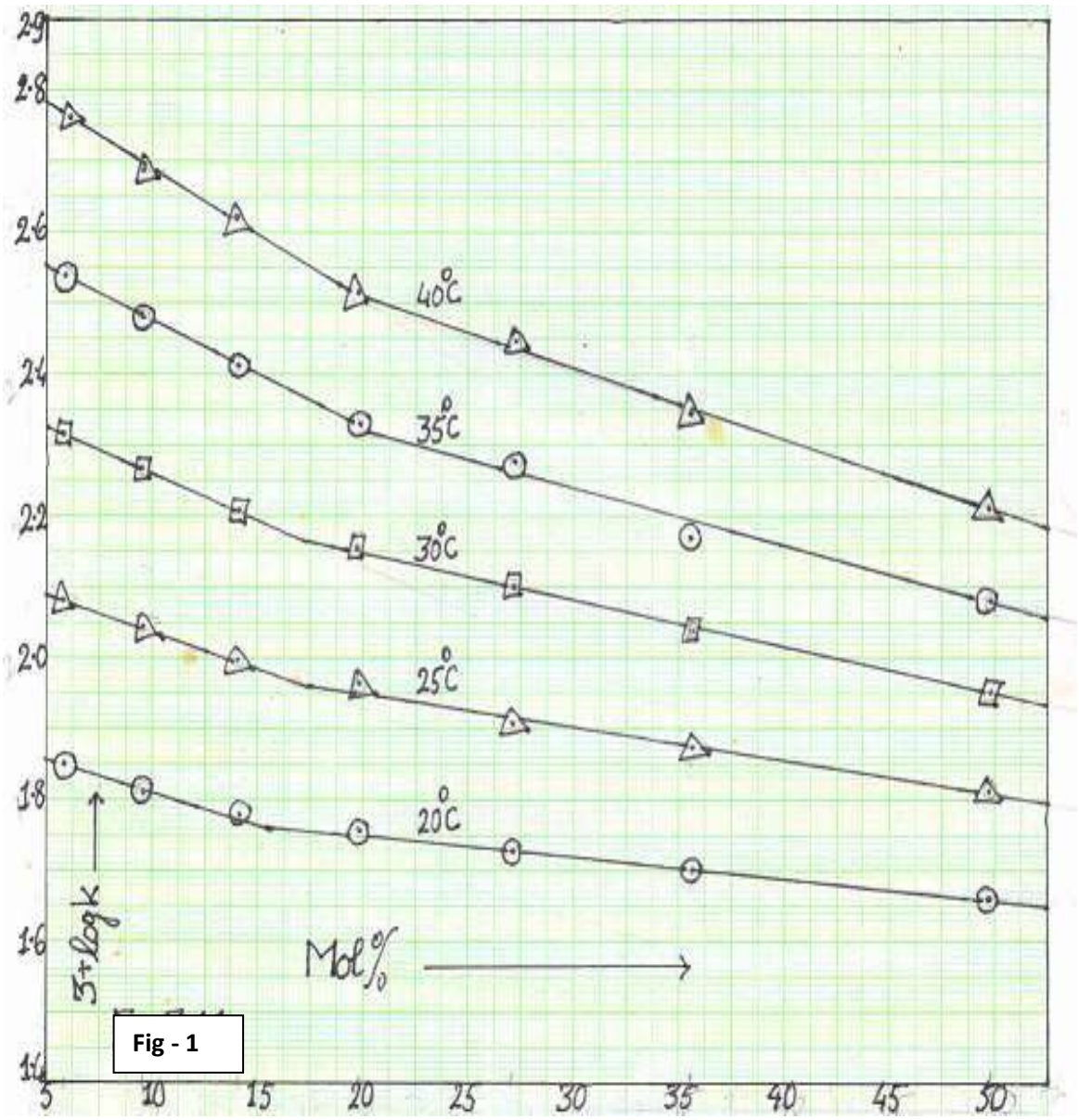

Fig - 1

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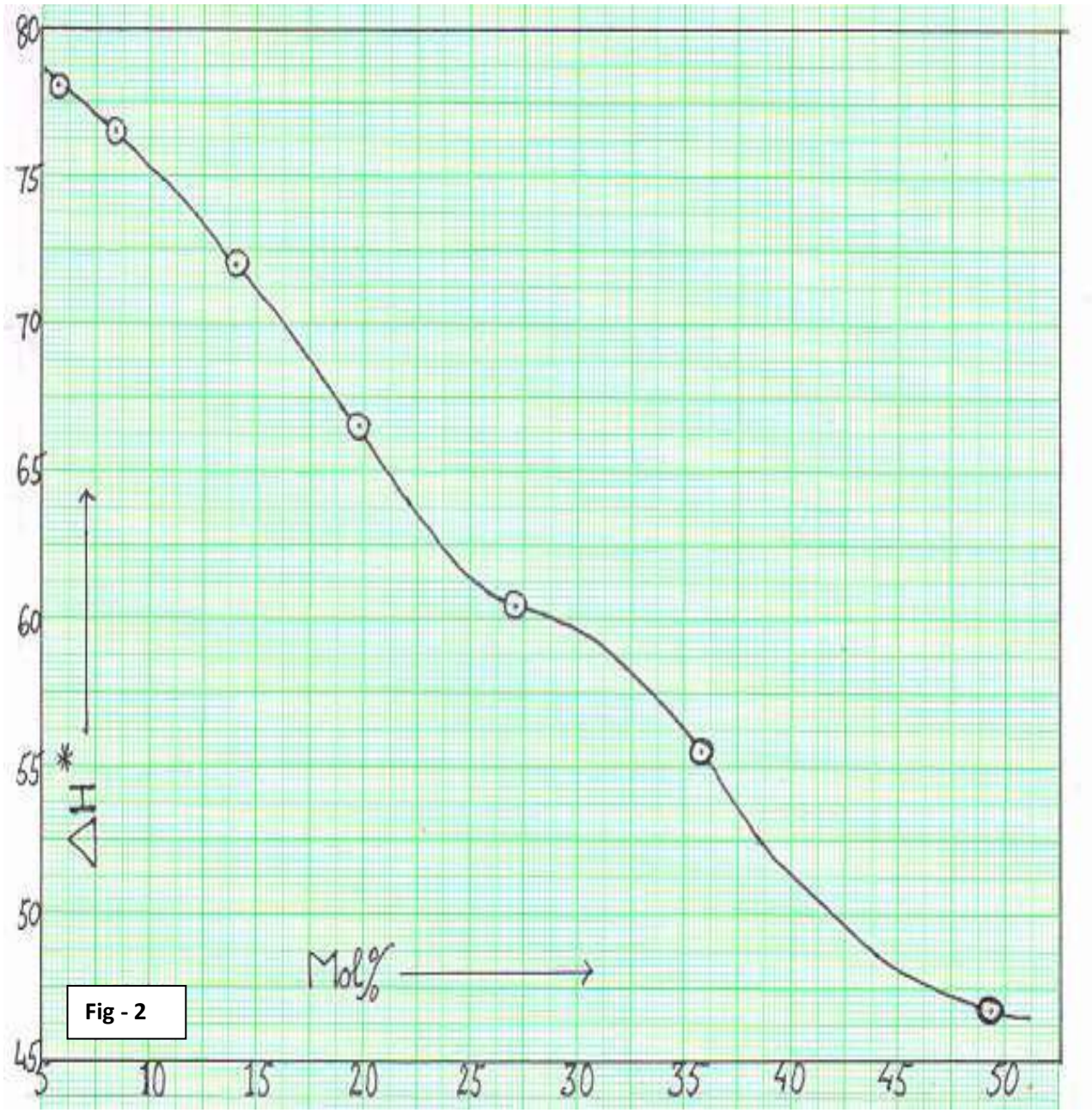

Fig - 2

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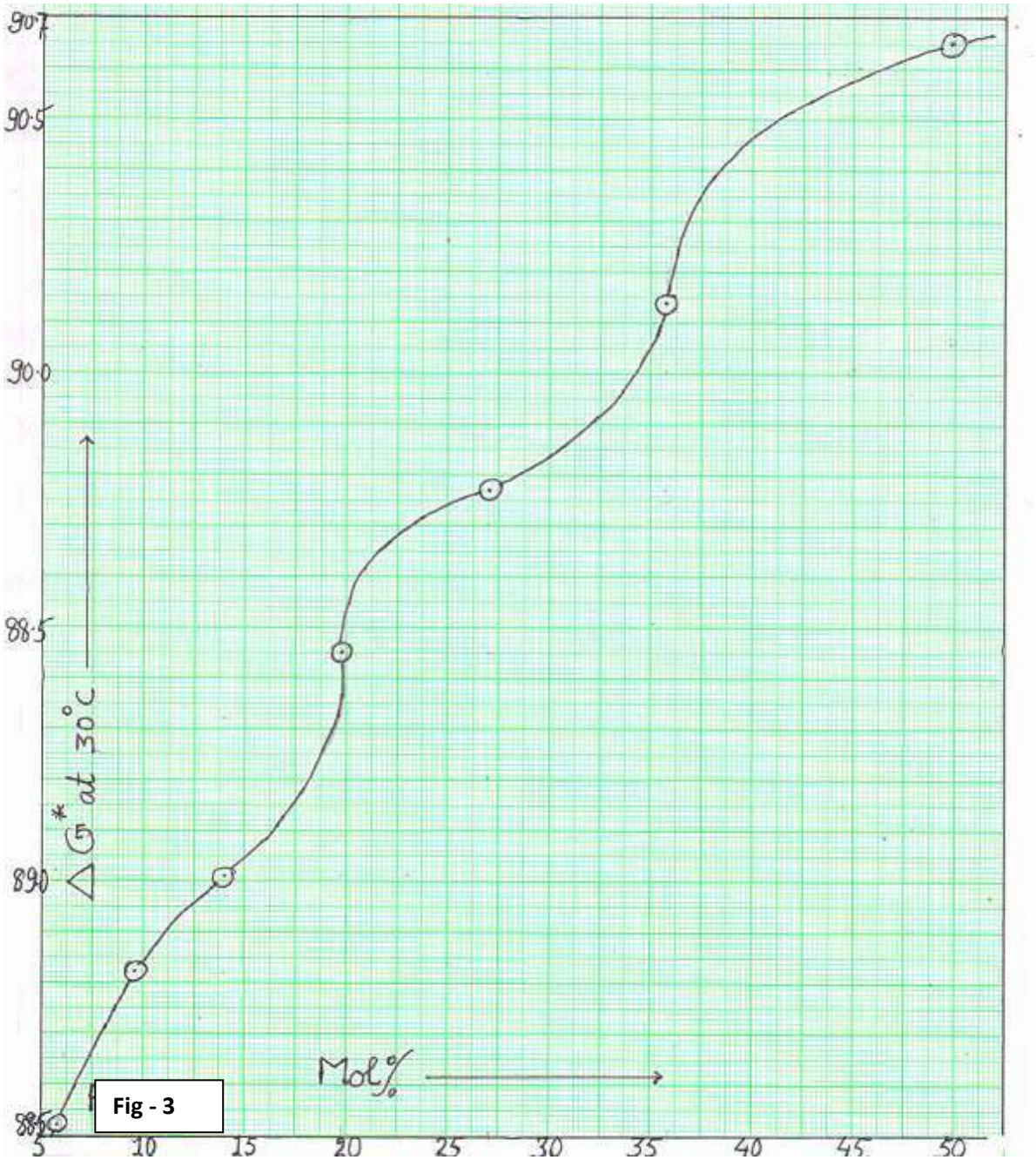

Fig - 3

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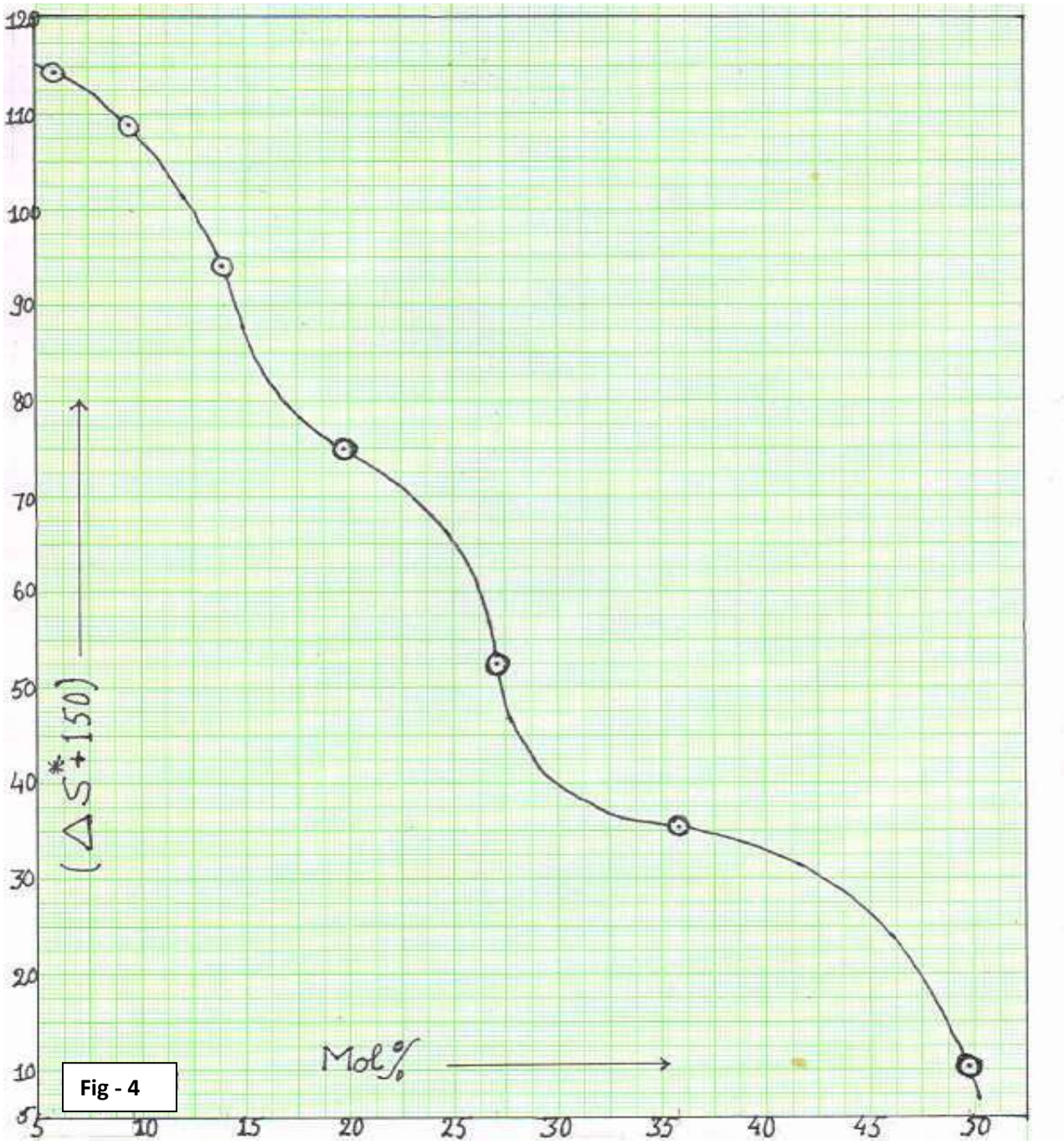

Fig - 4

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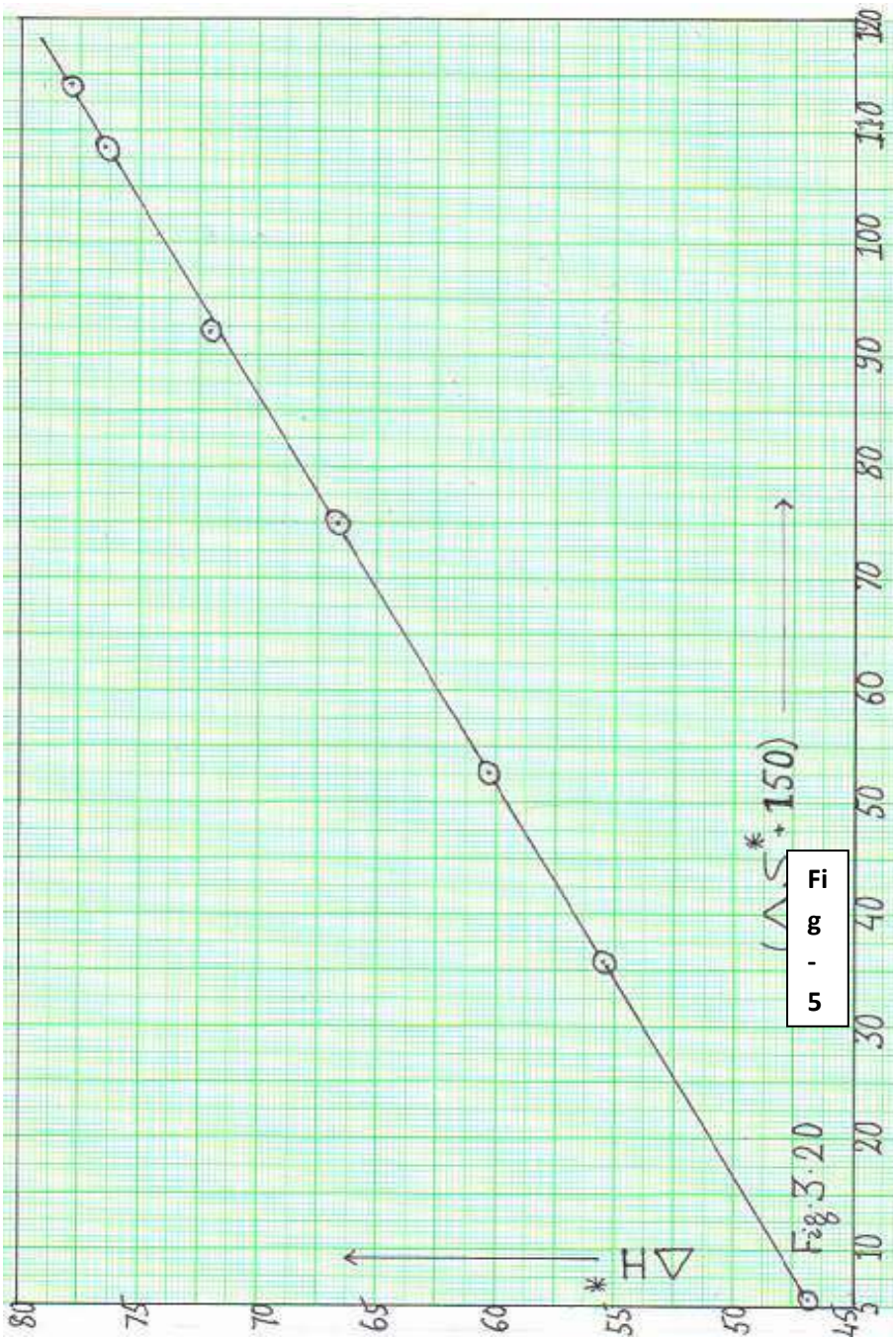

Fig - 5

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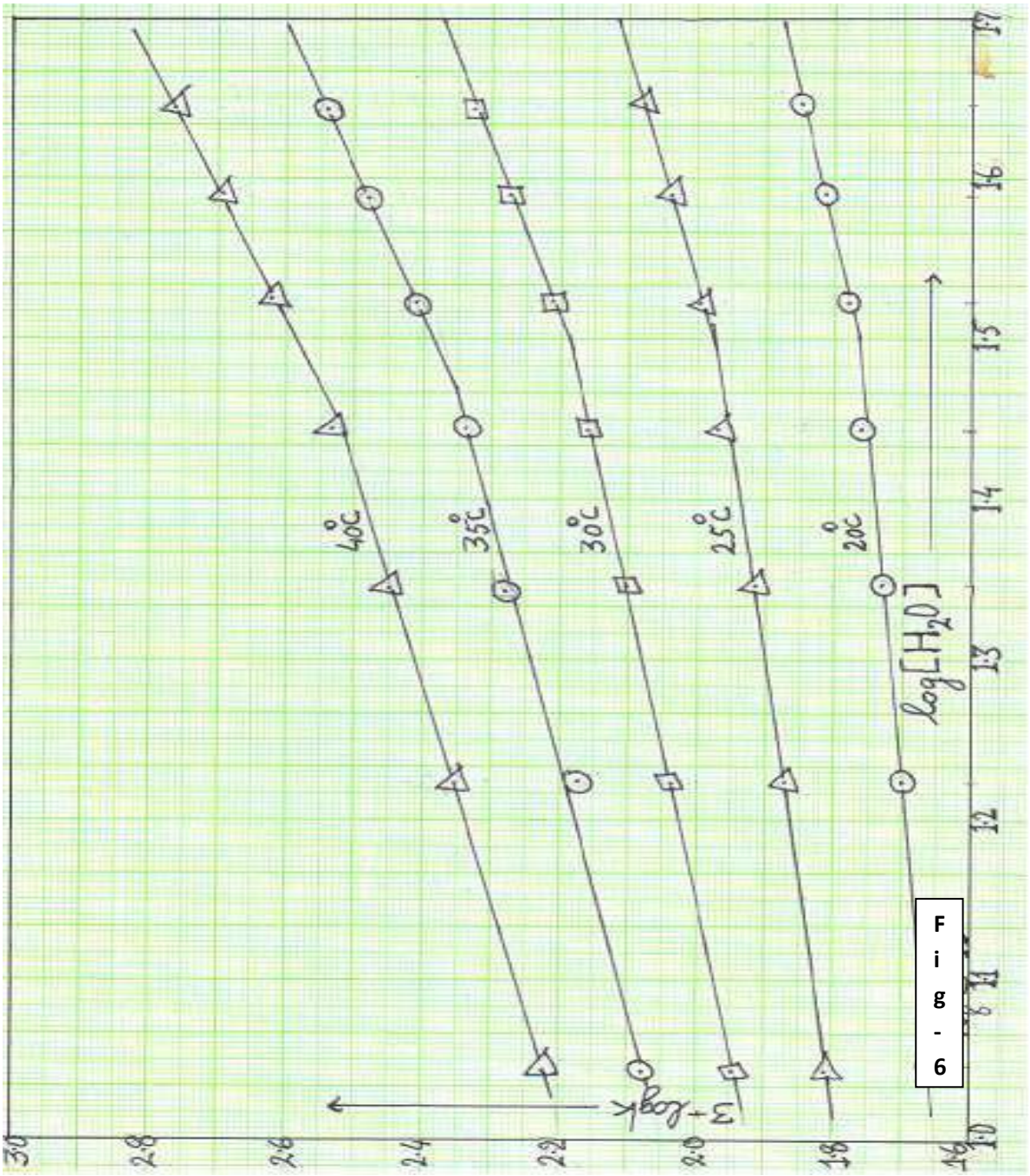

Fig - 6