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Part -3 - Azeotrope Formation and Distillation Curve of Gasoline –Alcohol Mixtures

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ABSTRACT

Mixing of alcohol with hydrocarbon base gasoline causes an increase in the vapour pressure of the mixture .This is because alcohol (methanol or ethanol) combines with certain low molecular weight Hydrocarbons to form azeotropes. Azeotropes have lower boiling points than the hydrocarbons from which they are made, resulting in an increase in vapour pressure . This paper presents the estimation of the areas under the distillation curves of gasoline – methanol mixtures and that of the hydrocarbon base gasoline. Calculation of areas was carried out using the Numerical Trapezoid rule and Calculus Definite Integration method. Linear equations were developed relating the logarithmic values of the areas resulted from azeotrope formations and the alcohol concentrations, Also, correlation between azeotropic area and some volatility terms were discussed

KEYWORDS.:
Distillation Curve - Gasoline –methanol mixture-Azeotrope Formation -Area under distillation curve.

INTRODUCTION:

Methanol has been used as gasoline additive in many countries. It cheaper than gasoline and can be mass produced easily [1, 2]. It had a higher octane number and can be used as octane enhancer. It contains 50% oxygen which reduces CO emission [3]. However mixing methanol with gasoline, the physicochemical properties of the mixtures are changed from properties of both methanol and gasoline to some extend. The influences of methanol addition on the evaporation properties of methanol / gasoline mixtures, have been widely studied [4-6]. They all found that the elevation of vapour pressure and dropping of distillation temperatures, are common features of the investigated mixtures.

Other investigators studied the Reid vapou r pressures and distillation curves of alcohol – gasoline mixtures containing 5-85 vol. % of methanol, ethanol, 1- propanol, 2- propanol, i- butanol and t-butanol [7, 8] . They reached a conclusion that these alcohols have the ability to form mixtures with hydrocarbons of gasoline exhibiting near – azeotropic behaviours, which affected the vapour pressure of the mixture in a non-ideal manner and changed the shape of the distillation curves [9, 10]. In distillation curves of gasoline – methanol mixtures, the distillation temperature increases rapidly until the first 5% has been recovered. Subsequantly, there is substantial plateau region where the distillation temperature increases only slowly and approaches the boiling point of pure methanol as the mixture is distilled, following the plateau region, the temperature increases rapidly again and approaches, but does not quite reach, the gasoline distillation curves [11, 12] .The rapid increase in temperature is believed to occur when the methanol has been completely distilled from the mixture [13]. Near – a zeotropic behaviour is observed in the plateau region, but by completely distilling the alcohol, the near – azeotropic mixture is removed and the distillation temperature increases toward that of the remaining gasoline hydrocarbons [11].

Distillation curves for blends of methanol, ethanol and butanols with gasoline have been reported [3, 7, 8, 11-15] and all of them found the azeotropy phenomenon. However none of them looked further to identify the azeotropes or to calculate the area due to azeotrope formation. In the present study, the distillation curves of five methanol –gasoline mixtures (M5, M10, M15, M17.5 and M20) along with methanol – free gasoline (M0), were constructed using the standard ASTM D-86 Distillation apparatus . The areas under distillation curves of methanol – gasoline mixtures and that of hydrocarbon base gasoline, were calculated using the Numerical Trapesoid Rule and Calculus Definite Integration Method.

MATRIALS and METHODS :

Hydrocarbon base gasoline (HBG) was formulated volumetrically from 60% reformate and 40% light naphtha. These two refinery streams were kindly supplied from Cairo Petroleum Refining Co. .Mostorod Refinery Varian detailed hydrocarbon – type GC analysis was also provided [16] . Methyl alcohol, 99.99 %, was kindly supplied by Methanex – Egypt Co., Damietta, Egypt.

Fuel blends were prepared from HBG and methanol. Five fuel blends comprise 5,10,15,17.5 and 20 vol.% methanol. RON of these blends along with the base fuel, were determined (ASTM-D299) in Cairo Petroleum Refining Co. Physicochemical properties of the blends and base fuel, were reported in previous work [16].

Area Under Distillation Curve (AUDC)

[17,18] described that the area under a curve is the area between the curve and the x-axis . In calculus , the area under the curve is measured using definite integrals . Microsoft Excel does not have functions to calculate definite integrals, but it possible to approximate this area using one of the numerical integration methods like the Trapezoid Rule .Using this rule , the approximate area under the graph of the function F(X) between two (X) values . The curve is divided into smaller curves , each tending to line segment forming a series of trapezoids ,each with area = (average height width) .

If the area under the distillation curve was divided into equal 8 stripes each represents a single trapezoid (as shown in figure), then the required area under the curve given by **I** where ;

$$\mathbf{I} = \int_a^b f(x)dx \underset{*}{=} \sum \qquad {}_i$$

Where$\boldsymbol{Ai}$ is each trapezoid stripe area, and

$$_i \quad \frac{y_i \quad {}_{i+1}}{} \quad (\quad {}_{i+1} \qquad {}_i)\mathbf{or} \quad {}_i \quad \frac{{}_i \quad {}_{i+1}}{} \quad \Delta x,$$

Where $\Delta x = - \quad -$ for each stripe as they all have an equal width for each;

a is the 10% (or 0.1) , the suggested area under the curve starting limit , and
b is the 90% (or 0.9) , the suggested area under the curve ending limit .
So, the approximate integral **I** may be written as:

$$\mathbf{I} = \Delta x \quad \frac{{}_1 \quad {}_2}{} \qquad \frac{{}_2 \quad {}_3}{} \qquad \frac{{}_n \quad {}_{n+1}}{}$$

The trend line equation will be substituted as a function $f(x)$ in the area under the curve (definite integral) equation instead of the previous (approximate integral) numerical trapezoid theory equation and it will be calculated simply by calculus using the following equation;

$$\mathbf{I} = \int_a^b f(x)dx \ldots\ldots$$

Where, $f(x)$ = the trend line equation from each curve trend line
a is the 10% (or 0.1) , the require area under the curve starting limit , and **b** is the 90% (or 0.9) , the require area under the curve ending limit .

Area Due to Azeotrope Formation :

The area under distillation curve of hydrocarbon base gasoline $(AUDC)_g$, and that under distillation curve of each gasoline – methanol blend $(AUDC)_{g\text{-}m}$. The area due to azeotrope formation is located between the two curves. By subtracting the smaller $(AUDC)_{g\text{-}m}$ from the bigger $(AUDC)_g$ the area due to azeotrope formation could be obtained .

$$\text{Area due to azeotrope Formation} = AUDC_g - AUDC_{g\text{-}m}$$

RESULTS AND DISCUSSION:

Distillation temperatures of hydrocarbon – base gasoline (MO) and five different gasoline –methanol blends (M5, M10, M15, M17.5 and M20), were measured using the standard ASTM-D86-04b [19]. These distillation data were used to construct twelve distillation curves of the investigated base fuel and fuel blends. In previous work, volatility, consumption and emission of methanol – blended gasolines, were studied [16, 20] . In the present study, the impacts of methanol addition on the shape of distillation curve of gasoline –methanol blends and the effects of methanol addition on the area under distillation curves and the area due to azeotrope formation.

Methanol addition and Shape of Distillation Curve :

Figure 1 shows two distillation curves , the upper for methanol – free gasoline (M0) , and the lower for methanol – blended gasoline (M20) ,i.e., containing 20% by volume methanol . It is clear that the addition of methanol significantly impacts the shape of distillation curve, particularly for the temperatures of the first 50-60 percent of the fuel to evaporate .This phenomenon has been clearly demonstrated in the literature [21, 22] . In the present study **,** the methanol molecule forms azeotropes with hydrocarbon compounds and this manifests itself as a flat portion of the distillation curve effectively at the boiling point of the azeotropes (around 45-70 ^{O}C) . Increasing methanol addition causes a substantial decrease in distillation temperatures over the middle portion of the distillation curve [11, 13, 16, 20, 23] .

For better illustration of the impact of blended methanol concentration on the shape of distillation curve , Four couples of distillation curves were constructed These distillation couples comprise M0-M5, M0-M10 pictured in figure2 and M10-M15 , M0-M17.5 in Figure 3 . It can be seen from these figures that as more methanol is added, the flattened area expands to cover larger portion of the distillation curve [3, 7, 24] . Also further methanol addition leads to significant drop in T50 value which represents the 50% evaporated temperature of the fuel. For instance, T50 of the investigated base gasoline is 89 ^{O}C. After blending 20 % vol.% methanol , a significant decrease in T50 value to 62 ^{O}C (Figure 1) . Such low T50 value is out of the international gasoline blend volatility regulations which should have minimum T50 value of 77 ^{O}C (170 ^{O}F), [25-27] . One can reach a conclusion that addition of methanol causes severe alterations to the shape of distillation curve of hydrocarbon – base gasoline. These alterations have significant impacts on distillation characteristics, volatility criteria and all other performance parameters of methanol – blended gasoline.

Area Under Distillation Curves :

In calculus, the area under the curve is measured using definite integrals. Microsoft Excel does not have functions to calculate definite integrals, but it is possible to approximate this area using one of the Numerical integration Methods [17,18] which comprise methods to find the approximate area under the curve of the function $f(x)$ between two (x) values . Using the Trapezoid Rule , the curve is divided into smaller curves , each tending to a line segment forming a series of Trapezoids , each with an area = (average height$*$width) .

In the illustrative Figure 1 , the area under the curve is divided into equal 8 stripes as suggested for this study from 10% to 90% evaporated (i.e., from x = o.1 to x = o.9 and n = 8) . Each stripe represents a single trapezoid .

In Figure 1 , two distillation curves are shown : the hydrocarbon base gasoline (M0) and that of the same gasoline after blending 20 vol .% methanol (M20) . From the distillation data (x and y axis values) , the areas under distillation curves of M0 and M20 , were calculated using two methods : the Numerical Trapezoid Rule (NTR) and Calculus Definite Integration (CDI) methods . Area due to azeotrope formation **= $AUDC_g$-$AUDC_{g\text{-}m}$**

Distillation data of the investigated hydrocarbon base gasoline (M0) and the other gasoline – methanol blends (M5, M10, M15, M17.5, and M20), are listed in Table 1. Also the polynomial equations, representing each distillation curve, are listed in Table 2. Four couples of distillation curves: M0-M5, M0-M10, M0-15 and M0-M17.5 are pictured in Figures 2-5. Areas under distillation curves, were calculated using the Numerical Trapezoid Rule (NTR) and the Calculus Definite Integration (CDI) method. The calculated values are tabulated in Table 3.

Careful inspection of data in Table 3 shows that the area under distillation curve decreases by increasing the blended methanol content. For example , the calculated area under distillation curve of M5 blend , is 57.4 square units , while that under distillation curve of M20 blend is 46.80 square units using the Numerical Trapezoid Rule (NTR) and the distillation curve extends between 0.1 and o.8 distillate volume recovery (i.e., from 10% to 80% volume recovery) When Calculus Definite Integration (CDI) method was applied , almost the same areas under distillation curves of M5 and M20 blends were achieved . For any of the investigated gasoline – methanol blends, the calculated area under distillation curves extending from 10% to 90% recovery (0.1to 0.9 distillate volume fraction), are bigger than those under the corresponding curves extending from 10% to 80% recovery.

Figure 6 illustrates the relationship between the area under distillation curves of the investigated fuels and blended methanol Two linear equations were developed for the curves extending from 10% to 80 % and from 10% to 90 % recovery, the areas in equations [1] and [2] are expressed in square units.

$$AUDC_{10\text{-}80\%} = -0.6874\,x + 60.608 \;\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots [1]$$
$$R^2 = 0.9986$$
$$AUDC_{10\text{-}90\%} = -0.7206\,x + 74.949 \;\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots [2]$$
$$R^2 = 0.9996$$

Figure7depicts the same relation but using log area (non- dimensional units) instead of square units (dimensional ones) .R^2 values of the four developed equations give indications concerning the reliability of the fits .

$$\text{Log } AUDC_{10\text{-}80\%} = -0.0056x + 1.7798 \;\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots [3]$$
$$R^2 = 0.995$$
$$\text{Log } AUDC_{10\text{-}90\%} = -0.046\,x + 1.8712 \;\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots [4]$$
$$R^2 = 0.9939$$

Area as under distillation curves (AUDC) in Figures 6 and 7 are calculated using the numerical trapezoid rule

(NTR) whereas, in Figures 8 and 9, areas are calculated using calculus definite integration (CDI) and other linear equations were developed

$$AUDC_{10\text{-}80\%} = -0.6793\,x + 60.003 \;\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots [5]$$
$$R^2 = 0.9947$$
$$AUDC_{10\text{-}90\%} = -0.701\,x + 74.142 \;\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots [6]$$
$$R^2 = 0.9942$$
$$\text{Log } AUDC_{10\text{-}80\%} = -0.0056\,x + 1.7843 \;\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots [7]$$
$$R^2 = 0.9969$$
$$\text{Log } AUDC_{10\text{-}90\%} = -0.o46\,x + 1.8759 \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots [\ 8\ ]$$
$$R^2 = 0.9989$$

Area due to azeotrope formation is located between the distillation curve of hydrocarbon base gasoline and the distillation curve of gasoline – methanol blend and can estimated by subtracting (area under distillation curve of hydrocarbon base gasoline ($AUDC_g$) – area under distillation curve of gasoline – methanol blend ($AUDC_{g+m}$) Figure 10 and 11 illustrate the relation between the AUDC and volume percent of the blended methanol . Using CDI and NTR .It can be seen from these figures that the area due to azeotrope formation is directly proportional with the volume percentage of blended methanol. Insignificant difference in the calculated areas using the CDI and NTR methods as indicated from the two values plotted in one line.

Area due to azeotrope formation using:

CDI = 0.6391x+ 1.0618 …………………………………………………….[9]
$R^2 = 0.9954$
NTR = 0.5978 x+ 1.4203………………………………………………… [10]
$R^2 = 0.9882$

Log Area due to azeotrope formation using

CDI = 0.033x+ 0.5072 …………………………………………………… [11]
$R^2 = 0.9915$
NTR = 0.0307 x+ 0.5333 ………………………………………………… [12
$R^2 = 0.9981$

CONCLUSIONS:

Results achieved in this study can lead to the following conclusions:

1- More than 10 volume percent methanol addition will lead to unfavourable volatility and significant drop in T50 value.
2- Blends containing 15, 17.5 and 20 volume percent methanol, are out of the volatility regulations specified by ASTM.
3- Concerning the calculated area under the distillation curve, both numerical trapezoid rule and calculus definite integration, are reliable methods.
4- Area due to azeotrope formation is in direct relation with the volume percentage of blended methanol.

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Table 1- Distillation Temperatures of Hydrocarbon Gasoline (M0) and Gasoline –Methanol Blends

% Recovered	Distillation Temperature , ^{O}C					
	M0	**M5**	**M10**	**M 15**	**M17.5**	**M20**
IBP, ^{O}C	36	34	35	35	35	34
10 %	55	44	45	45	44	43
20 %	62	49	48	49	48	48
30%	69	55	52	51	50	50
40%	78	74	59	56	55	54
50%	89	85	81	63	63	62
60%	101	98	97	92	78	71
70%	114	112	111	109	106	105
80%	129	128	129	129	125	124
90%	152	151	152	149	150	149
FBP, ^{O}C	185	185	184	184	183	183

Table 2- Distillation Curve Equations

Fuel	Added Methanol VOL. %	Polynomial Equations of the Distillation Curves	R^2
M0	0.0	$Y= 91.55X^2 + 25.44X+ 52.61$	0.998
M5	5.0	$Y= 79.97 X^2 + 53.88 X+ 36.19$	0.995
M1O	10.0	$Y= 127.60 X^2 + 10.23X + 40.47$	0.992
M15	15.0	$Y= 173.20 X^2 + 40.10 X+ 47.40$	0.987
M17.5	17.5	$Y= 209.80 X^2-77.68 X+ 52.50$	0.993
M20	20.0	$Y= 226.60X^2-99.95X+ 56.21$	0.991

Table 3- Calculated Areas Under Distillation Curves Using NTR and CDI Methods

Fuel	Area Under Dist. Curve (From 10-80 vol.% recovery)		Area Under Dist. Curve (From 10-90. vol.% recovery)	
	NTR	CDI	NTR	CDI
M0	60.50	60.435	75.00	74.480
M5	57.40	55.917	71.35	69.898
M10	53.50	53.286	67.55	67.432
M15	50.55	50.505	64.30	63.909
M17.5	48.50	48.017	62.35	61.839
M20	46.80	46.460	60.50	59.976

NTR = Numerical Trapezoid Rule

CDI = Calculus Definite Integration

*all calculated areas are in square units.

Table 4- Calculated Areas Resulting from Azeotrope Formation

Fuel	Azeotropic Area * (from 10% to 80% recovery)		Azeotropic Area * (from 10% to 80% recovery)	
	NTR	CDI	NTR	CDI
M5	4.80	4.518	5.35	4.582
M10	7.00	7.149	7.45	7.048
M15	9.95	10.385	10.70	10.570
M17.5	12.00	12.418	12.35	12.641
M20	13.70	13.975	14.50	14.504

NTR = Numerical Trapezoid Rule

CDI = Calculus Definite Integration

*all calculated areas are in square units.

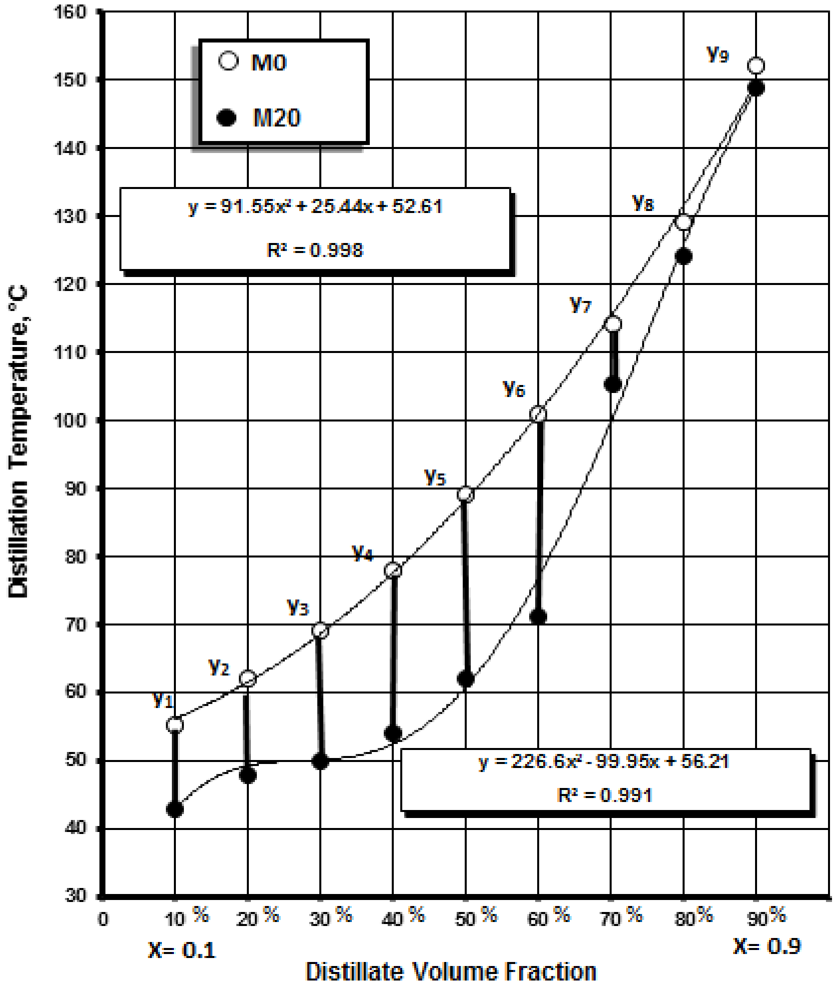

Figure 1 - Illustrates areas under M0 and M20 curves and the area due to azeotrope formation between the two curves.

Base Gasoline (M0)		
X axis	Y axis,°C	Area
10 % (0.1)	55	5.85
20%	62	6.55
30%	69	7.35
40%	78	8.35
50% (0.5)	89	9.50
60%	101	10.75
70%	114	12.15
80%	129	14.05
90% (0.9)	152	
AUDC g		**74.55**

(M20) blend		
X axis	Y axis,°C	Area
10 % (0.1)	43	4.55
20%	48	4.90
30%	50	5.20
40%	54	5.80
50% (0.5)	62	6.65
60%	71	8.80
70%	105	11.45
80%	124	13.65
90% (0.9)	149	
AUDC g+ m		**61.00**

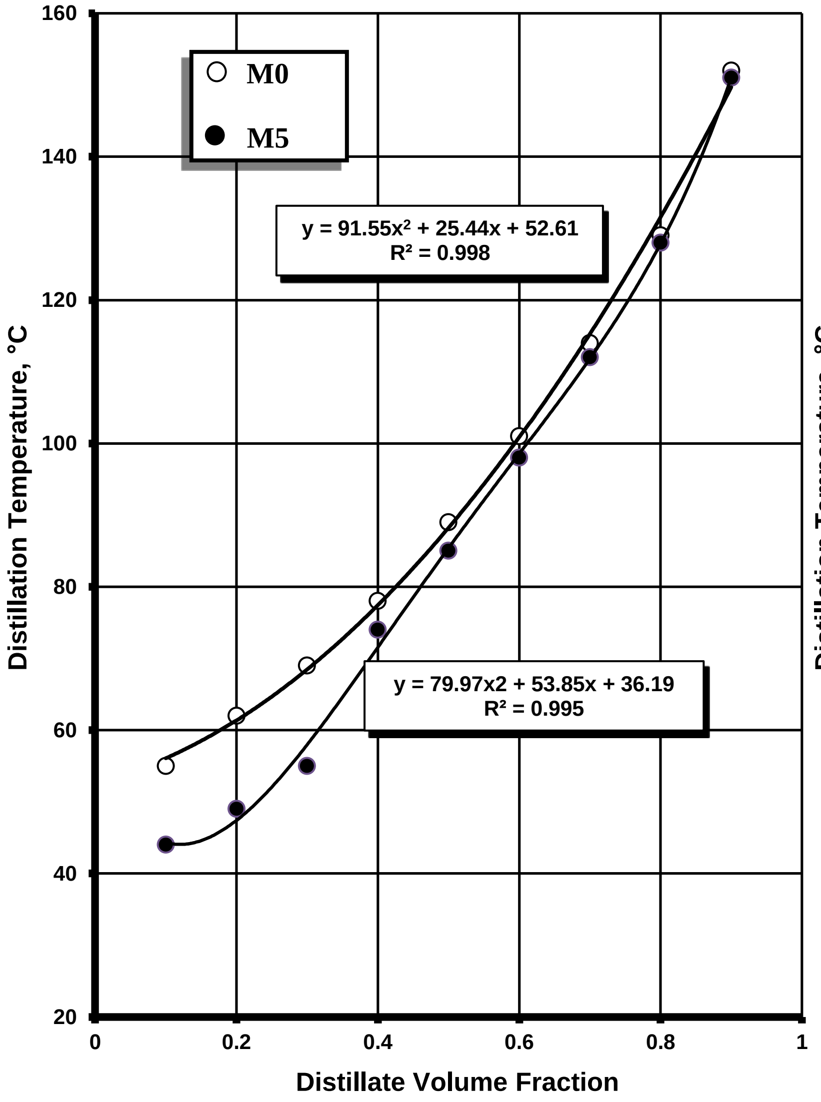

Figure 2- Distillation Profiles for M0 & M5.

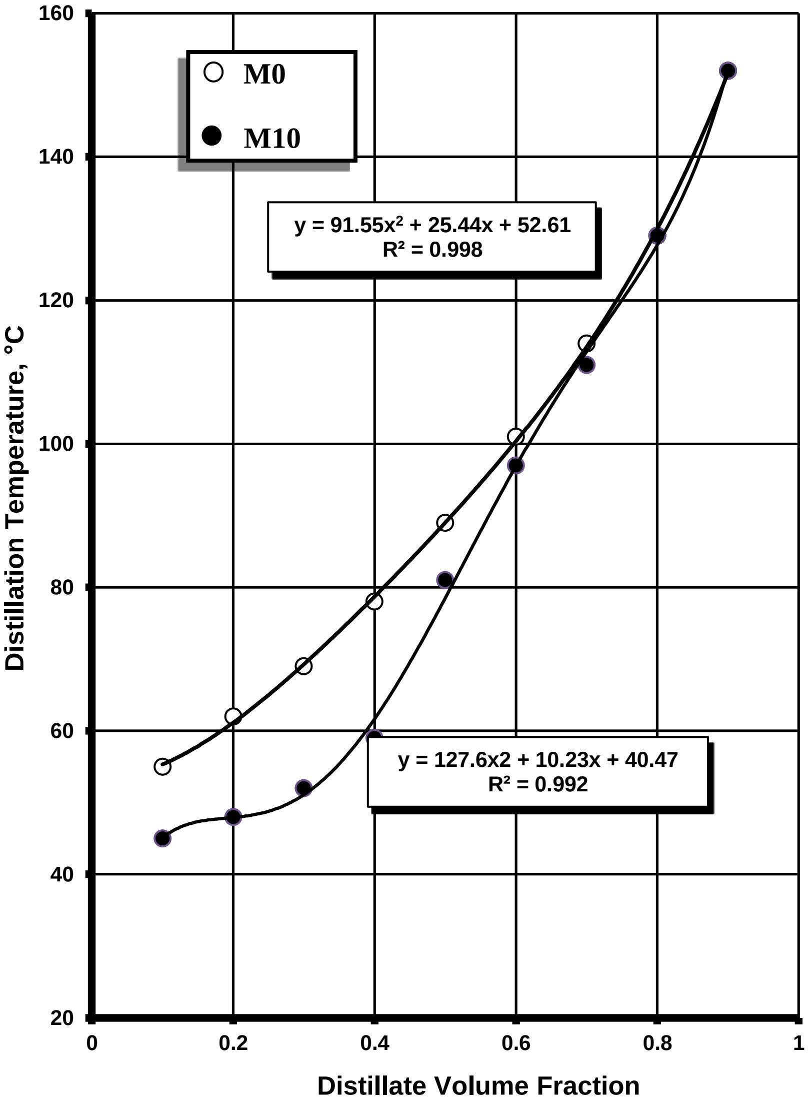

Figure 3 - Distillation Profiles for M0 & M10.

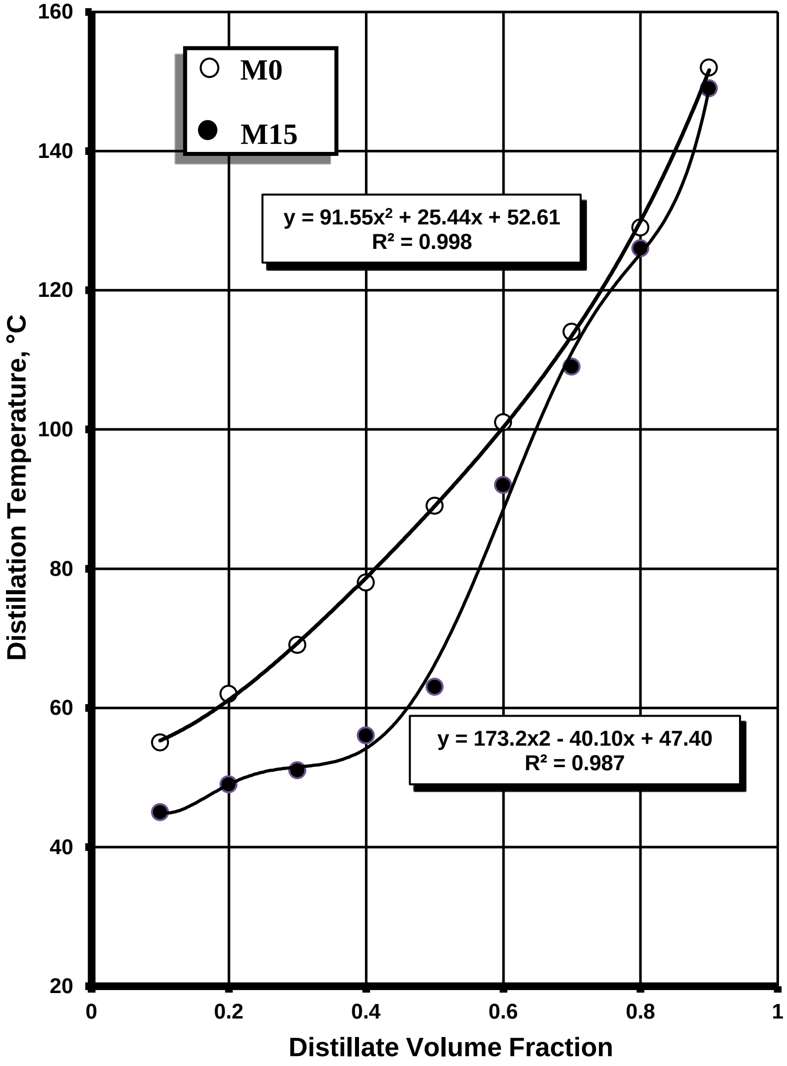

Figure 4- Distillation Profiles for M0 & M15 .

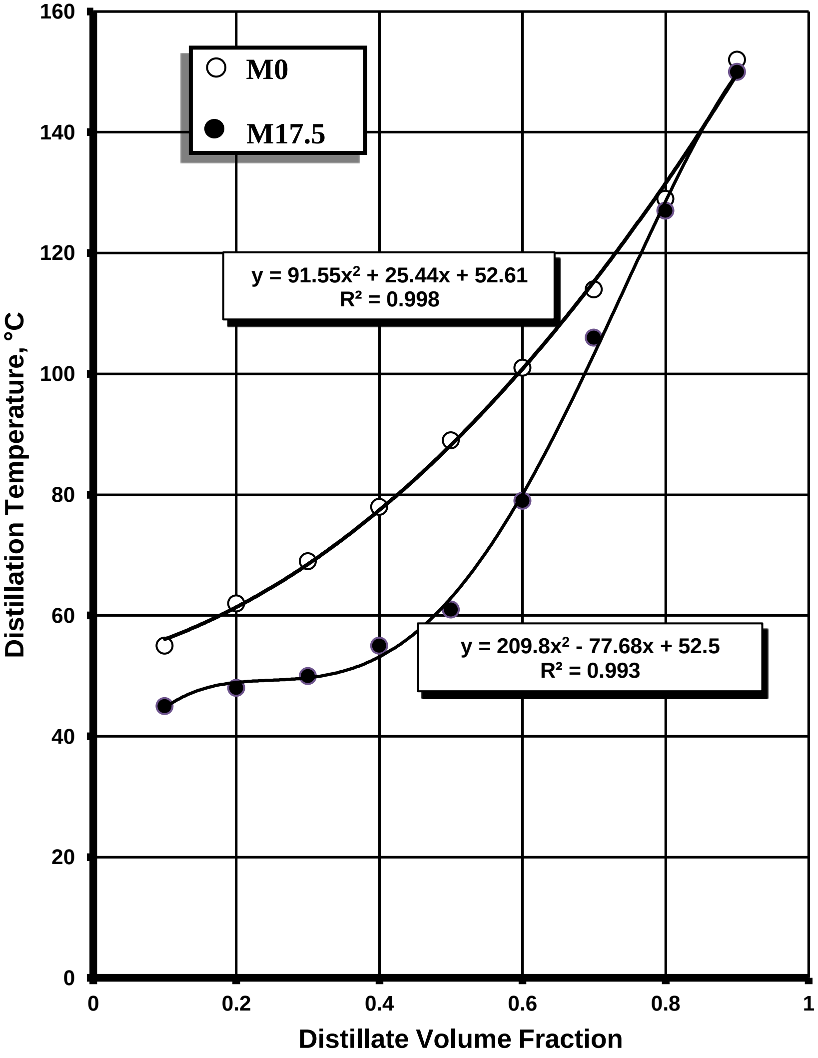

Figure 5- Distillation Profiles for M0 & M17.5.

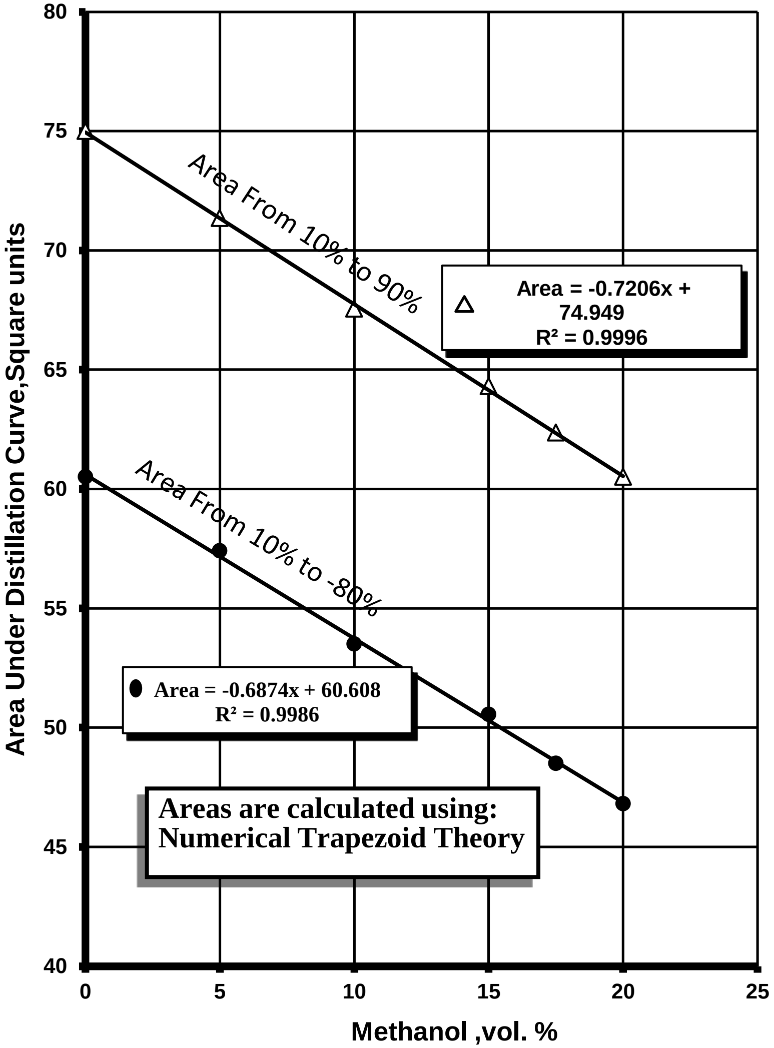

Figure 6: Area Under Distillation Curve as a Function of Blended Methonal using Numerical Trapezoid Theory .

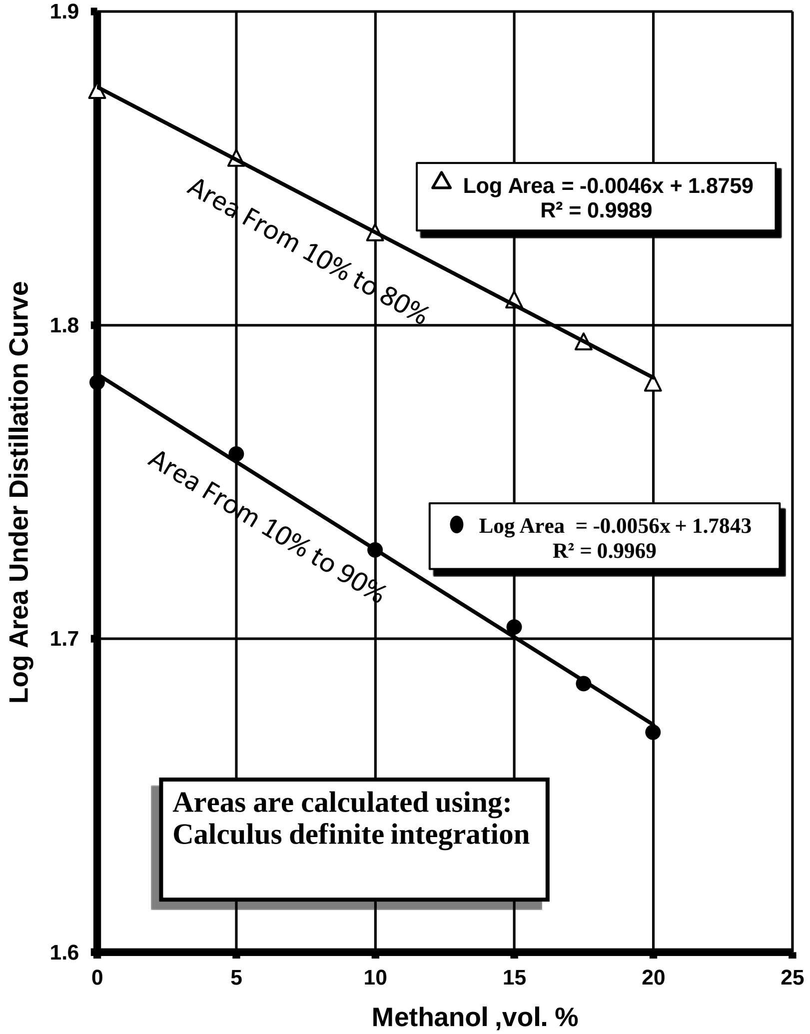

Figure 7: Log Area Under Distillation Curve as a Function of Blended Methonal using Numerical Trapezoid Theory.

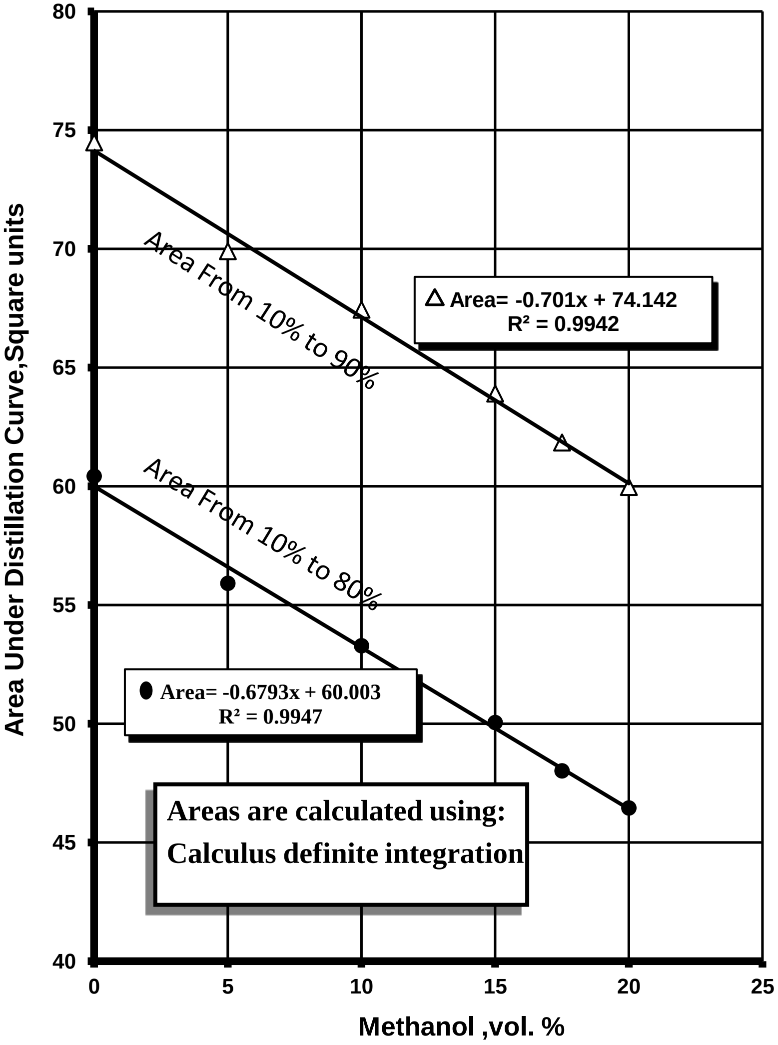

Figure 8- Area Under Distillation Curve as Function of Blended Methonal using Calculus Definite Integration

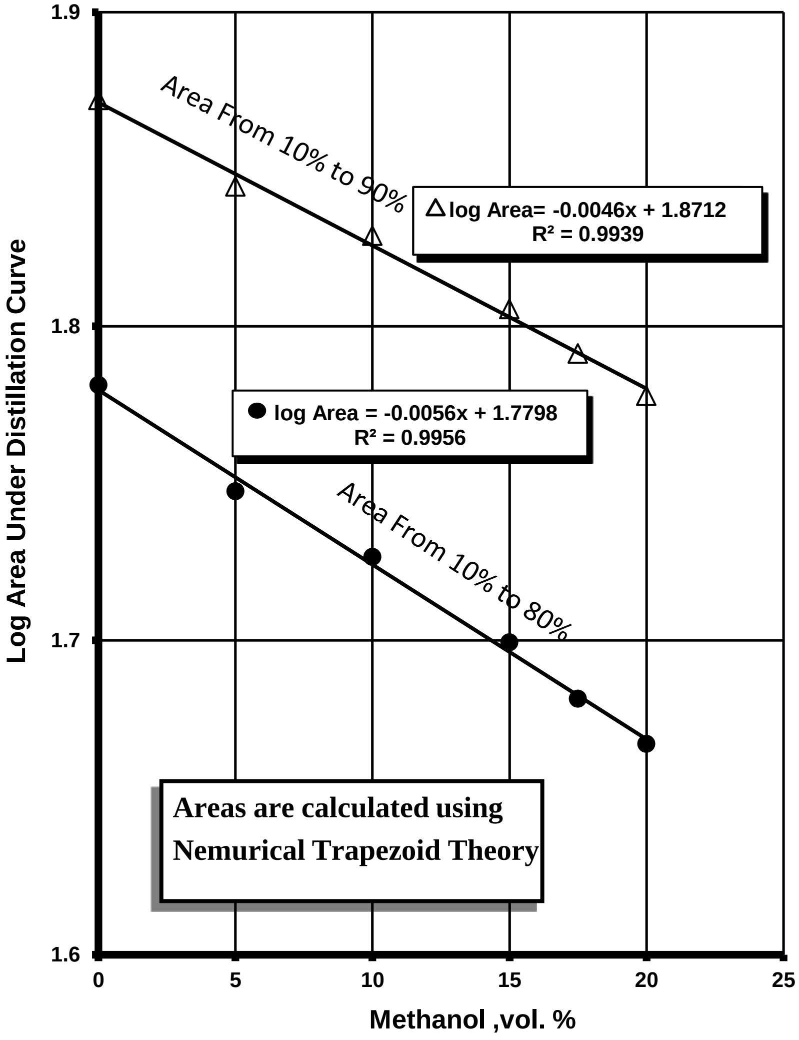

Figure 9- Log Area Under Distillation Curve as a Function of Blended Methonal using Calculus Definite Integration

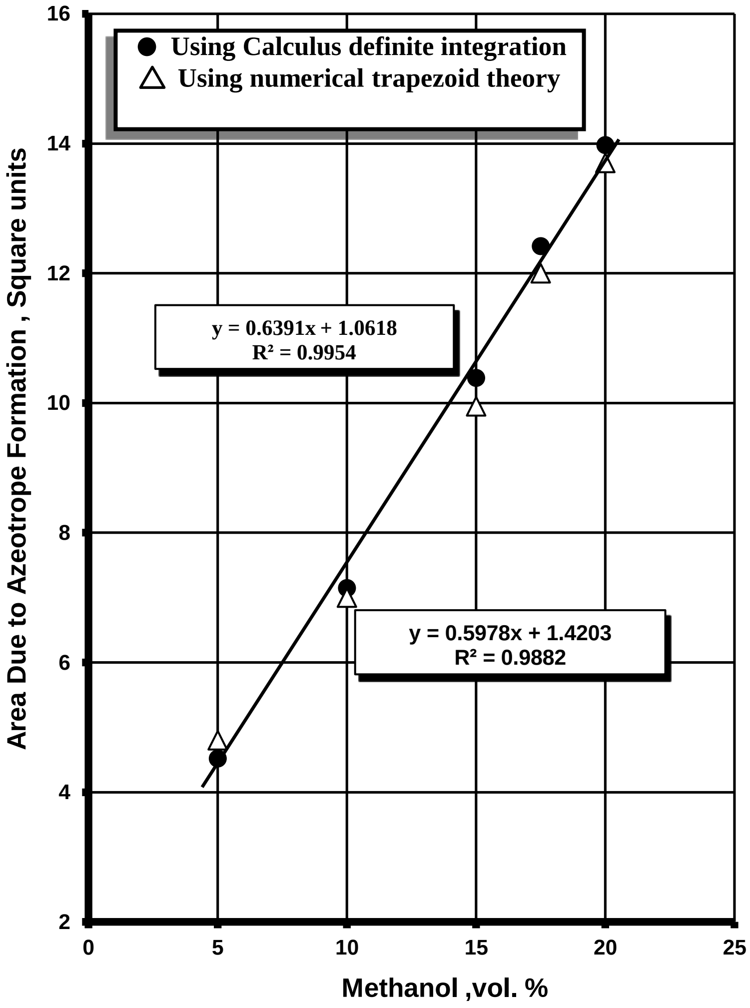

Figure 10- Area Due to Azeotrope Formation as a Function of Blended Methanol.

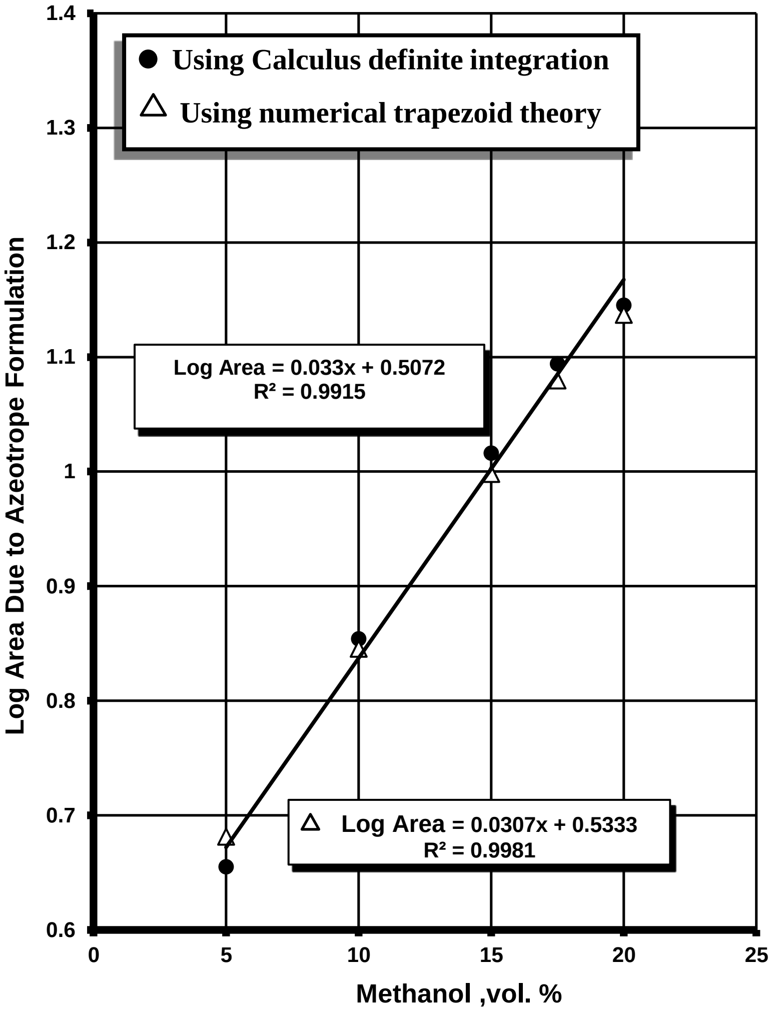

Figure 11- Log Area Due to Azeotrope Formation as a Function of Blended Methanol.