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Laser-Raman Technique for Determination of Products Concentration Exhausted from I.C. Engines.

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LASER-RAMAN TECHNIQUE FOR DETERMINATION OF PRODUCTS
CONCENTRATION EXHAUSTED FROM I.C.ENGINES

استخدام أشعة اللـيزر فى قياس تركيز النواتج الـعادمة من آلات الاحتراق

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الخلاصة ــ فى هذا البحث تم استخدام اشعة اللـيزر فى قياس نواتج الاحتراق الـعادمـــة
من آلات الاحتراق الداخلى . تم تطبيق النظام على قياس الاكسجين وأول أكسيد الكربون.
قياسات معملية باستخدام ليزر He - Ne بطول موجى ٦٣٢ نم كمصدر وتم مقارنـــــة
النتائج بالقياس باستخدام جهاز تحليل النواتج . وتم فى هذا البحث تصميم وانشــــاء
جهاز لتجميع الطيف الناتج من تفاعل اللـيزر مع الجزئيات وفصل الموجات المختلفـــــة
من بعضها ذلك لان كل فار لـه طول موجى محدد . وبالتالى أمكن التغلب على مشكلـــــة
رئيسية فى عملية قياس كل فار على حده دون التداخل مع الغازات الأخرى لمكونات العادم
وظهضا من هذا البحث العملى أن هذا النظام صالح للقياس حيث وجد توافق بين النتائـج
المحسوبة والقيم المقاسه بجهاز المعايرة ووجدتا أيضا أن هذا النظام أسرع مــــــــن
القياس حيث أن زمن القياس ٢٥٠ x ١٠ـ⁹ ثانية أما جهاز المعايرة فانه يستغرق مـــن
٢ : ٣ دقيقة وبذلك يمكن أن يكون القياس بالنظام المقترح لحظى . أما المشاكـــــل
التى ظهرت أثناء اجراء البحث هو عدم توافر جهاز ليزر بقدرة مناسبة وكذلـــــــك
مكبـــــــر ذو حساسية مناسبة .

ABSTRACT

In this paper, the Laser Raman technique for the
measurements of the species concentration of the gases exhaus ed
from I.C Engines is presented. An application of the method for
oxygen concentration by Raman spectroscopy has been made.
Experimental data obtained using He-Ne laser beam with 632.8 nm
wave length as a source are presented and compared with
conventional method for analysis. The main parameters and outline
of the apparatus are described. An oxygen analyzer has been used
to evaluate the results. The results show a good agreement as
compared with the results measured by the oxygen analyzer. The
Laser-Raman technique proved very rapid for measuring the species
concentration of the exhaust gases. For example, the measuring
time when using the oxygen analyzer is approximately 2-3 minutes ,
while by Raman technique is slightly more than the response time
of the photodiode detector which is approximately 250 nano
seconds. The problems associated with this experimental work are
the low cross section area of Raman scattern, the low laser power
and the low sensitivity of the amplifier.

1-INTRODUCTION

The current method of the detection and measurements of combustion products exhausted from internal combustion engines is based on sampling of these products followed by subsequent analysis in laboratories. The main disadvantage of this method is in the long period expended in data collection during which thermal and chemical variations may distort the actual pollutants content.

Laser-Raman method of the detections and identification of the products of combustion has the advantages of remote controlling and the capability of conducting a qualitative and quantitative analysis of these products. It is also more rapid than the classical chemical methods. Another advantage for this method is its independence of excited wavelength (laser) of Raman spectra. Thus, we can use any one wavelength laser to measure the concentrations of any molecule in the exhaust products. However, the most significant limitations of this technique arise from the low cross section from Raman scattering.

The Raman effect arises from internal energy changes within an individual molecules as a result of the interaction of its electromagnetic field with the incident light and is thus characteristic of the particular molecular species involved [1]. The literature in this field [2,3] indicate that Raman spectroscopy is a very suitable method for the determination of the species concentration of gases exhausted from internal combustion engines and other combustion gases properties such as temperature.

Bechtel [4] used both Raman and Laser induced florescence for measurement of the temperature and species concentration profiles. The results show a good agreement between the theoretical predictions and the measured concentrations of CH_4, O_2, H_2O, CO_2 and OH in several CH_4-air flames. In the field of burners, the Raman spectroscopy is a useful method for temperature and species concentration profiles[5]. This reference used the spontaneous Raman spectroscopy for temperature and molecular concentration profiles in a premixed laminar flame, generated by a multihole burner. The results obtained by using chopped Ar ion-Laser beam as a source.

The aim of this work is to investigate the measurements of combustion products exhausted from internal combustion engine by using Raman spectroscopy. The emphasis will be directed for the measurements of O_2 and CO. The latter has a direct impact on the environment and combustion efficiency.

2-EXPERIMENTAL ARRANGEMENT

The Raman experimental set-up used in this work for measurements of O_2 and CO concentrations exhausted from internal combustion engine is shown schematically in Fig.(1). It consists of PASCO model Os-9171 He-Ne laser. The power of the laser is 0.5 mW, the wavelength of the beam radiation is 632.8 nm and the laser beam diameter is 1 mm. It is a rather safe unit for using in the laboratory, however, according to its data specifications, one should never look directly into the laser beam, nor should one look into the reflection of a laser beam from a mirror. The laser

beam cuts and scatters the gases exhausted from a single cylinder, air cooled, four stroke Diesel engine. The laser beam enters the collimator and wavelength separator apparatus. This apparatus collimates also the scattered radiation and separates it according to its wavelength. The detector, which is a photodiode detector, mounted in a TO18 style case with end window. It can detect a certain required scattered wavelength by setting it at certain position in the wavelength separator's outlet. The photodiode current due to scattered radiation is the input of the amplifier, and the output voltage is measured by the oscilloscope.

The photodiode current is proportional to the scattered beam intensity which is proportional to a certain species concentration. The output voltage appears on the oscilloscope is directly proportional to the photodiode current. Then the oscilloscope output is an indication of a certain molecular concentration of species exhausted from the engine.

3-THE COLLIMATOR AND WAVELENGTH SEPARATOR

In our experiments we have designed and constructed the collimator and wavelength separator. This apparatus is used to overcome the problem of the detection for a certain scattered wavelength. The physical idea of it is shown in Fig.(2) which illustrates two beams in the same line with different wavelength and when any of the two beams passes through a transparent prism, the deviation angle of each beam depends upon its refractive index which depends upon its wavelength. Consider ψ is the angle of the prism's head, ϕ_1 is deviation angle of a beam of wavelength λ_1, ϕ_2 is the deviation angle of beam of wavelength λ_2, $\Delta\phi$ is the difference between the two deviation angles ϕ_2 and ϕ_1, r_1, r_2 are the refractive index of each beam and z is the distance between the deviated beams at distance Y from the prism. The required from the design is to calculate the distance z for O_2 and CO molecules.

According to the fundamental of optics we have the relation,

$$\Delta\phi = \psi \ (r_2 - r_1) \tag{1}$$

But λ_1=632.8 nm is the He-Ne laser wavelength (reference beam), the Raman spectra for the oxygen and carbon monoxide are 1556 and 2145 Cm^{-1} respectively[6&7]. The scattered wavelength for oxygen becomes 701.9 nm and for CO becomes 732.2 nm.

The refractive index for a beam with wavelength 632.8 nm in the prism's material r_1=1.519, refractive index for wavelength of 701.9 nm (O_2) is 1.51 and the refractive index for wavelength of 732.2 nm (CO) is 1.506 [8]. Then in the cases of O_2 and CO from equation (1), where ψ =60 degrees,

$$\Delta\phi \ (O_2) = \ 0.54 \ degree \tag{2a}$$

$$\Delta\phi \ (CO) = \ 0.78 \ " \ " \tag{2b}$$

Thus, the distance z for the two cases can be calculated knowing that the distance Y in our apparatus is equal to 350 mm and $z=Y \tan(\Delta\phi)$. Then $z(O_2) = 3.3$ mm and $z(CO) = 4.8$ mm. The position of the photodiode detector is 3.3 mm from the laser beam in order to measure the oxygen concentration and 4.8 mm for carbon monoxide detecting. Fig.(3) shows a schematic diagram for the constituent of the collimator and wavelength separator. In this work the apparatus is adjusted in order to make the distance between the collimating lens and the measured point is equal to the focal length of the collimating lens.

4-EXPERIMENTAL RELATION

Consider Io to be the intensity of the excited beam (laser beam) in mW/cm^2 and ΔI is the scattered power within an element of solid angle. Then we have the relation [9]:

$$\Delta I = Io \; N \; \Omega \; (d\delta/d\Omega) \quad mW \qquad (3)$$

Where, N is the number of molecules in the measured volume (V), $d\delta/d\Omega$ is the Raman scattering cross section within $d\Omega$ an element of solid angle and Ω is the solid angle of the collimation.

Consider $\Delta I(D)$ to be the power incident on the detector which must be less than I in equation(3) because of collimation efficiency of about 0.9 [6] in this experiments then,

$$\Delta I(D) = 0.9 \; Io \; N \; \Omega \; (d\delta/d\Omega) \; mW \qquad (4)$$

and the detection area A(D) can be introduced;

$$\Delta I(D) = (0.9/A(D)) \; Io \; N \; \Omega \; (d\delta/d\Omega) \quad mW/cm^2 \qquad (5)$$

and introducing the photodiode specifications, then id, which is the leakage current with I(D) incident light can be written as;

$$id = (33.3/A(D)) \; Io \; N \; \Omega \; (d\delta/d\Omega) \quad mA \qquad (6)$$

and introducing the relation between ΔVo and id from the amplifier characteristics;

$$\Delta Vo = (1.665*10^9/A(D)) \; Io \; N \; \Omega \; (d\delta/d\Omega) \quad mV \qquad (7)$$

where ΔVo is the increasing of the output voltage.

The measured volume (v) in the experiment is approximately 0.25 cc. Consider in that volume, the total number of molecules Nt, where Nt contains the measured molecules and the others. According to the equation of state;

$$PV = (Nt \; R \; T/ Nov) \qquad (8)$$

where P is the gas pressure (1 atm.), v a measured volume, Nov the Avogadro's number ($6.022045*10^{23}$ molecule/gm.mole), R is the universal gas constant (.082 atm.lit/gm mole. K) and T is the temperature of the measured gas.

Then; $Nt = 1.05*10^{19}$ molecule   (9)

Consider x the percentage molecular of the measured gas, then $x=100*N/Nt$ %; $A(D)=0.00785$ cm.cm we get;

$$x = 4.49*10^{-29} \ Io \ \Omega \ \Delta Vo \ (1/(d\delta/d\Omega))$$ (10)

In this work, the solid angle   is the ratio between   the diameter of the collimating lens and its focus lens which is equal to 0.5672. For He-Ne laser Io equal to 63.7 mW/cm.cm, then equation (10) becomes;

$$x = 1.24*10^{-30} \ (\Delta Vo/(d\delta \ /d\Omega \ )) \ \%$$ (11)

Equation(11) is the general experimental relation in this work. It is useful for detecting any molecules according to its Raman cross section area $(d\delta/d\Omega)$. For oxygen measurements the Raman cross section area is equal to $1.23^{-1} \ Cm^2/Sr$.[6&7]., then the general equation(11) becomes;

$$x = 10.1 \ \Delta Vo$$ (12)

Equation (12) is the our experimental relation for measuring the percentage oxygen concentration, x where, the quantity ΔVo is the increase in the output voltage above the noise of detection circuit in mV.

5-RESULTS AND DISCUSSIONS

The results shown in Table 1. illustrate the quantities measured by the oscilloscope (ΔVo) in mV and the oxygen concentration measured   by the Oxygen analyzer at different engine loads. We notice from the results in the table that the measured values of ΔVo is a good indication for oxygen concentration, where ΔVo increases with the increase of the oxygen concentration. This is because when the oxygen concentration increases. the scattered radiation intensity increases. Then the diode current then increases which causes the increase of the output voltage (ΔVo).

Fig.(4) shows a comparison between the quantities of x which is calculated from Raman technique and that measured by the oxygen analyzer. It can be seen from the results shown in the figure that the values of low oxygen concentration ($x < 2.5\%$) do not agree with the measured values by the oxygen analyzer. This is due to the low sensitivity of the oscilloscope and the lower power of the laser used.

Fig.(5) shows the calibration curve (solid curve) which is the relation between the increase of output voltage (ΔVo) and the values of oxygen concentration measured by the oxygen analyzer The curve indicates the calibrated values of oxygen concentration corresponding to the increase of the output voltage (ΔVo). In this figure, the dashed line is the calculated one corresponding to the experimental relation(12). The results shown in Figs.(4 and 5) show that the Raman technique is very valuable for oxygen concentration measurements. The sensitivity of the present apparatus fails for values less than 2.5% and that is not a defect in our technique . It is just the sensitivity of the oscilloscope is not suitable for voltages less than 0.1 mV.

CONCLUSIONS

In this work, we have used the Raman technique to measure the species concentration of the gases exhausted from internal combustion engine. The following conclusions could be made;

1.The Raman technique is very more rapid than eitherr the classical chemical methods or the oxygen analyzer techniques. The measuring time of the oxygen analyzer is about 2 minutes and in the classical chemical methods is more than this time. In Raman technique the measuring time is slightly more than the response time of the photodiodee detector. This is approximately 250 nano seconds. This more rapid tool helps us to measure the instant change in species concentration such as engine start and the sudden variation in its load.

2.This technique is applicable to measure the concentration of any molecule exhausted from the internal combustion engine by knowing the Raman cross section area of the scattered molecule.

3.The collimator and wavelength separator designed and constructed in this work are available for collimating the scattered radiation from a certain point and separate it according to its wavelength to a certain point at a distance (z) from the laser beam (works as monochromator).

4.The problems of this experimental work are the low cross section of Raman scattering, the low power of the laser and the low sensitivity of the oscilloscope. These problems made the technique not useful to measure the low concentration of the gases.

5.As a future work, it would be useful to get a high sensitivity measurements of the species concentration. These need power pulsed laser, monochromator and suitable amplifier .

NOMENCLATURE

A(D)	Detection area
I	Intensity of light
i	Electric current
Io	Laser beam intensity
id	Diode current
Nov	Avogadro's number
Nt	Total number of molecules
P	Gas pressure
R	Universal gas constant
r	Refractive index
T	Temperature
t	Time
v	Measuring volume
Vo	The output voltage of the amplifierx
x	Percentage gas concentration in a mixture
Y	Distance from the prism and the wavelength separator
z	Distance from the detector position and the laser beam
ϕ	Deviation angle
ψ	The angle of the prism head
λ	Wavelength

δ   Cross sectional area of scattering
Ω   Solid   angle

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Table 1 ΔV_o, Calculated values from Eq.(12) and the values of
x from oxygen analyzer at different engine loads

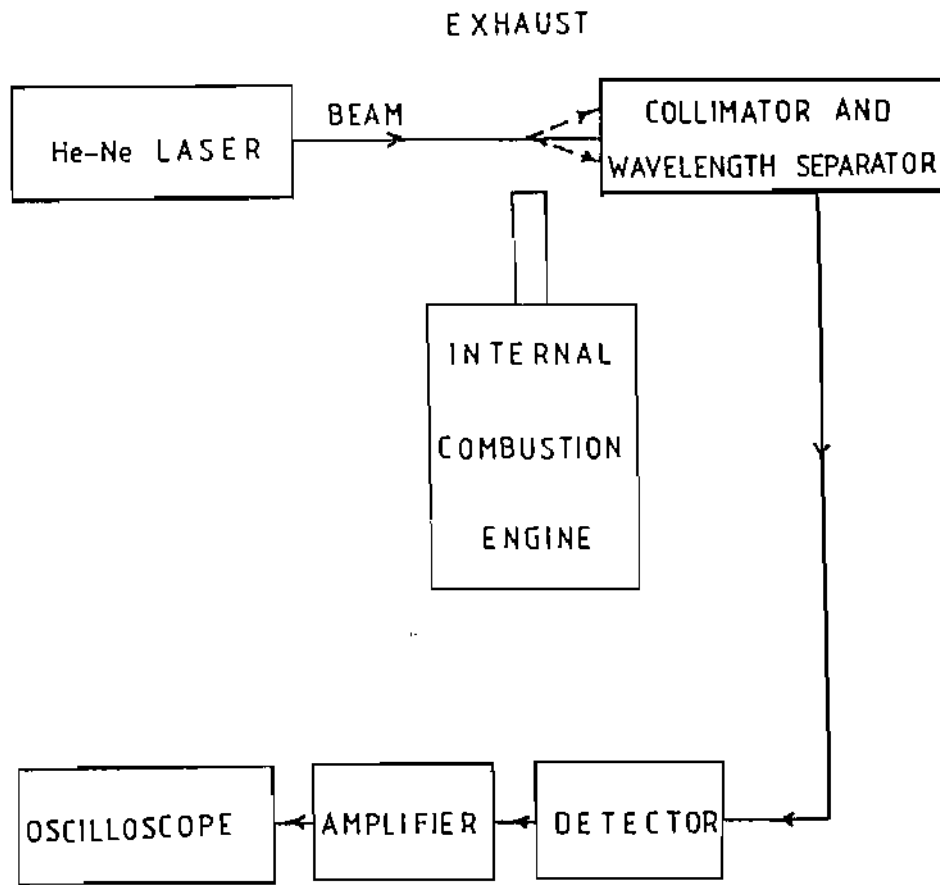| Engine load (%)	ΔV_o (mv)	Calculted x	Measured x
0.0	0.6	6.06	6.35
25.0	0.45	4.545	4.4
50.0	0.35	3.535	3.3
75.0	0.3	3.03	2.55
100.0	0.3	3.03	2.1

EXHAUST

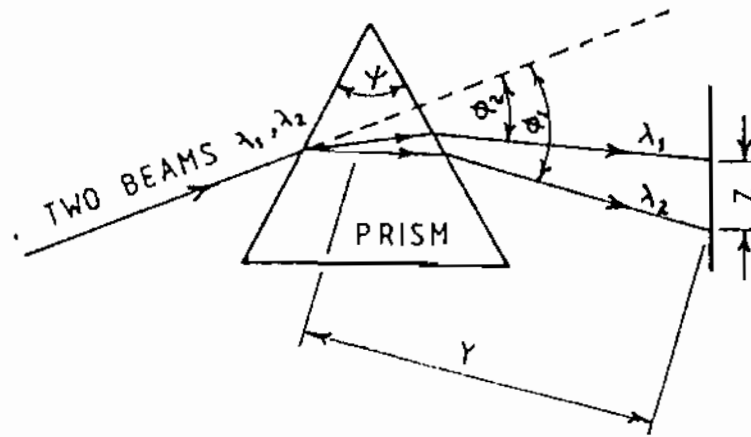Fig.(1) A schematic Diagram of the Set up of the Experiment.

Fig.(2) A Schematic Drawing Shows the Physical Idea of
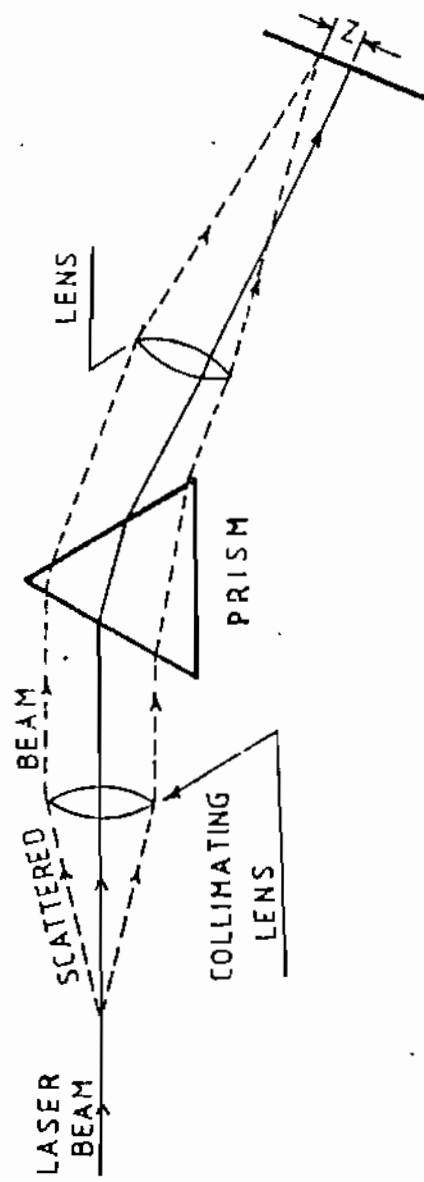the Wavelength Separator.

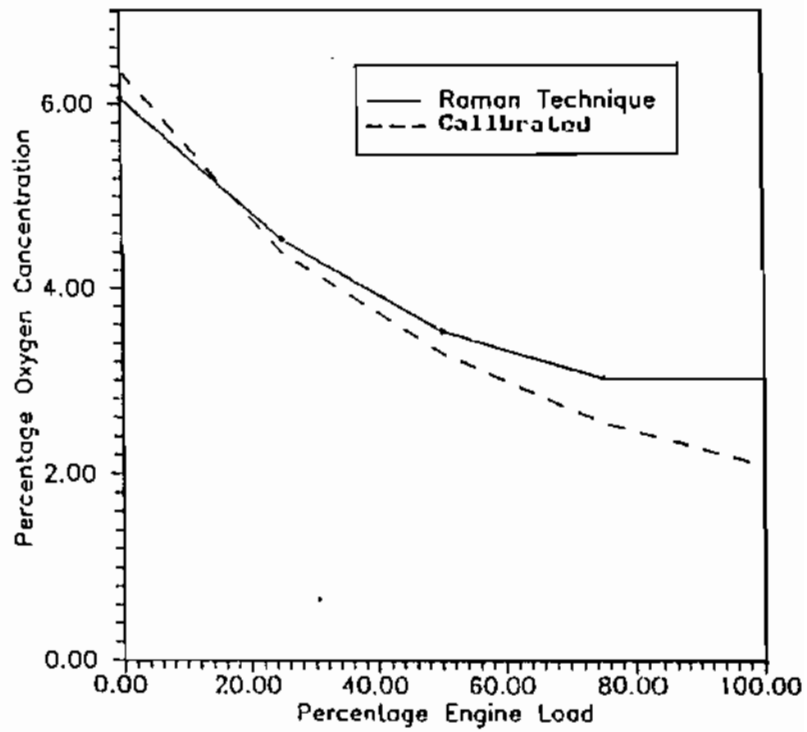Fig.(3)   The Constituent of the Collimator and Wavelength Separator Apparatus.

Fig. (4) A Comparison Between Calculated Results
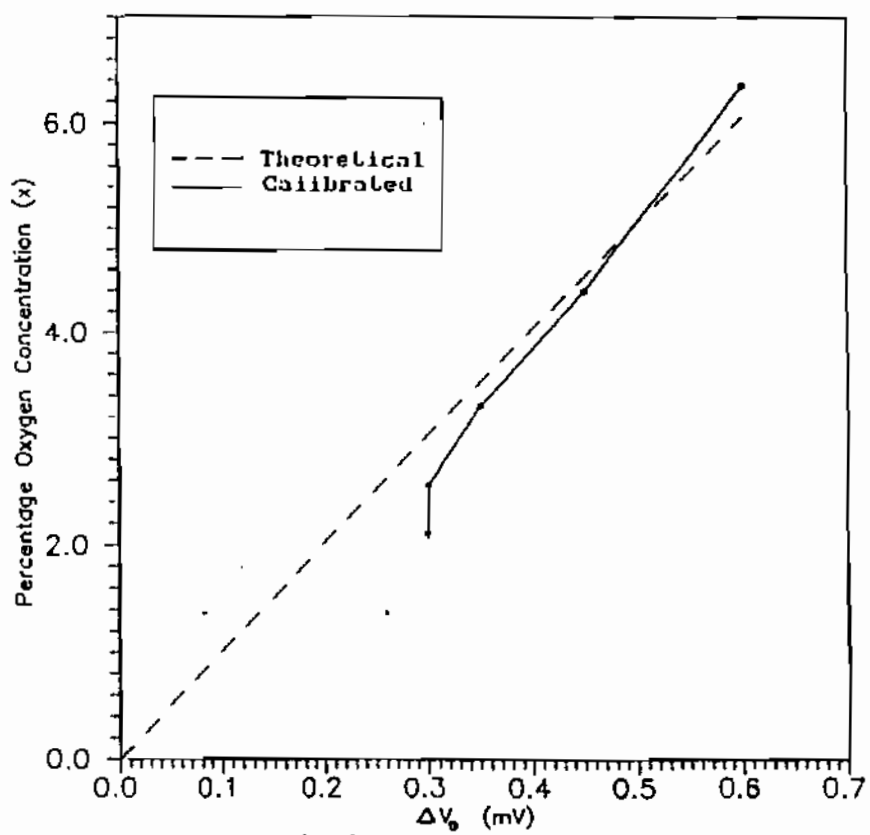and Calibrated Values

Fig.(5) The Calibration Curve