PHOTOTHERMAL & PHOTOACOUSTIC

from Basic Research
to Industrial Applications

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Contents

INTRODUCTION

Many optical techniques in basic research rely on sound, heat and light. What makes some techniques stand out more than others in their ability to address the needs in basic research and at the same time respond to some of the driving forces in industrial applications? Among the optical techniques that satisfy the above criteria are transmittance and reflectance spectroscopy, photothermal and surface plasmon resonance methods. The latter two techniques have several advantages over the conventional optical techniques.

Photothermal methods rely on the excitation and detection of thermal waves in gaseous and condensed matter. Photothermal techniques are distinguished by two interesting properties. Firstly, the dissipation of energy of optically excited states into thermal energy provides a means to explore non-radiative de-excitation channels in matter. Secondly, monitoring the time and spatial dependence of the oscillating temperature field offers unique possibilities in optical and thermal parameters. In the last two decades several photothermal techniques have been developed, which are based on the same physical principle but differing by the kind of excitation and detection of the thermal waves. Deposition of heat into the sample through absorption of electromagnetic radiation with laser is the most frequently used excitation source. Detection of the thermal response is performed either by direct contact (gas contact) or without contact with the sample. These detection schemes of photothermal phenomena can be summarized as follows;

For solid-and liquid-contact

1. Pyroelectric detection of thermal waves.
2. Piezoelectric detection of thermaelastic effect from solids
3. Piezoelectric detection of the photoacoustic effect from liquids

For gas-contact

1. Microphone detection of Photoacoustic effects
2. Laser beam deflection due to Mirage effect
3. Microphone detection of photoacoustic effect from gases

For Non-contact

1. Photothermal Radiometry, IR radiation thermal waves
2. Thermally modulated reflection (thermoreflection)
3. Laser beam deflection (surface displacement and thermal lens)

In basic research, the main emphasis has been devoted to improving the sensitivity, the spectroscopic and spatial resolution of the photothermal techniques within the frame of laboratory scale equipment and experiments. The detection limits of low concentration of optical absorbers could be considerably increased by the application of photoacoustic methods for gases and solids. Due to the fact that the photon noise of the heating beam is not seen by the photothermal detector, the signal to noise ratio can be considerably enhanced by increasing the heating beam intensity. In our Applied Optics Laboratory, we focus our research on the development of photoacoustic and surface plasmon resonance as a powerful optical technique for characterizing the thermal and optical properties of materials and optical sensor applications.

WHY PHOTOACOUSTIC AND SURFACE PLASMON RESONANCE TECHNIQUES:

These two optical techniques, which are employed in our Applied Optics Laboratory, have several advantages over other conventional optical techniques :

- They are easy and inexpensive to set up, reliable, cost effective and efficient alternative approaches to other techniques for monitoring and qualitative determination of various optical and thermal properties of solids, liquids and gases.
- Since the optical beam can be focussed to a tiny spot, the photoacoustic and surface plasmon resonance techniques require only a small sized sample and involve minimum sample preparation. In addition to that, non-contact and non-destructive measurements are possible.
- The two techniques also work with low intensity optical beam in visible to infrared regions, thus the techniques are completely safe and do not involve any hazardous materials.
- Both techniques are sensitive, fast and accurate which makes performing measurements less time and work-consuming. Since their response time is within a few minutes, real-time measurement is possible.
- Photoacoustic spectroscopy technique permits continuous monitoring of constituents in solutions without the need to dilute samples, thus allowing analysis of mixtures in situ, under real-life conditions, and if needed, online.
- Photoacoustic and surface plasmon resonance spectroscopy enable measurements at microliter and nanoliter sample volumes with wide range of visible-infrared wavelengths. Both are 10 to 1000 times more sensitive than conventional UV-VIS spectroscopy.
- Considering the optical components involved and the development of modern light sources such as the solid state laser and OPO laser, the technique can be implemented through small and compact sized units suitable for industrial applications.

PHOTOACOUSTIC TECHNIQUE

The sound effect induced in a heated solid by an intermittent light beam was first observed by Graham Bell[1] in the eighteenth century. This effect was understood as a photothermal phenomenon and was the origin of the so-called photoacoustic effect. Nowadays, there is a range of Photothermal techniques and most of them were derived from photoacoustic spectroscopy (PAS). Basically, their differences are related to the detection schemes employed. There are detection systems using photodiodes pyroelectrics, piezoelectrics, thermopiles and microphone. Photoacoustic (PA) is a photothermal phenomenon that has been widely used to study the thermo-optical properties of materials. In brief, the photoacoustic effect consists of illuminating a given sample with a modulated light beam and measuring the subsequent temperature fluctuation induced in the sample resulting from light absorption (i.e. and due to the non-radiative de-excitation processes within the sample). Since the signal responds only to the absorbed light, the effect of scattered light plays no significant role in this photoacoustic technique. In addition, this method provides a solution to the difficulties presented by conventional optical spectroscopy which does not generally allow studies of very weak absorbing materials and also opaque samples. Some photoacoustic studies in infrared are based on the Fourier Transform method (FTIR-PA), which allows the measurements to be carried out in infrared regions up to 400 cm^{-1}. Photoacoustic is particularly useful in this region since it can detect the hydrogen bonds (C-H, O-H and N-H) that may present overtones and contributions from the combination of the stretching and vibrational modes of the bands.

PHOTOACOUSTIC THEORY, R-G MODEL

The photoacoustic effect can be produced by any kind of absorption that results in periodical heating. Some schemes of photoacoustic generation are, thermal expansion, thermal diffusion, thermalelastic bending and photobaric effects (gas evolution). In general the possible PA generating mechanisms in solids are shown in Figure 1. Among these, thermal diffusion is the most common mechanism responsible for the generation of the photoacoustic signal.

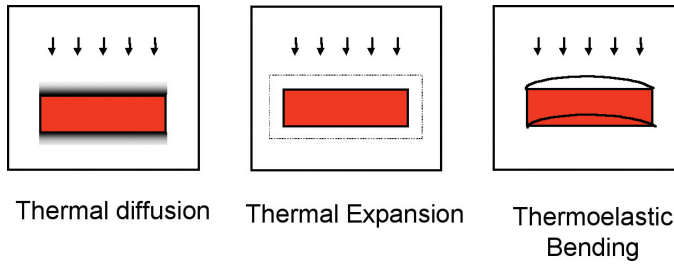

Figure 1 Photoacoustic generating mechanisms

All these effects result in pressure variation inside the gas chamber of the photoacoustic cell. The Rosencwaig-Gersho[2] thermal piston model (Figure 2a) considers the pressure inside the photoacoustic cell to be proportional to the heat generated by the absorbed light and dependent on both the geometry of the cell and the thermal-optical properties of the investigated sample.

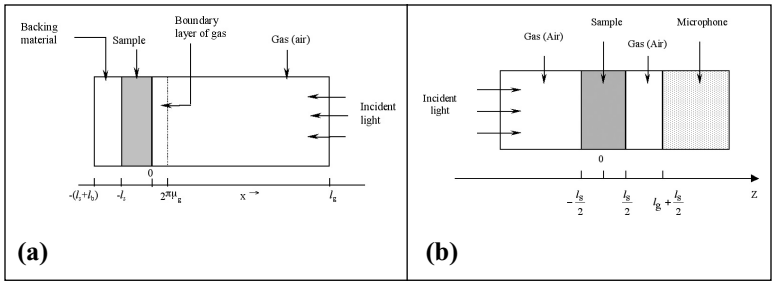

Figure 2 Rosencwaig-Gersho thermal piston model

For a thermally thick sample, the complex amplitude of the pressure variation inside the cavity is given by

$$Q = \frac{-i\beta\mu^2\gamma P_o I_o}{4\sqrt{2}T_o l' a' k} \tag{1}$$

where β is the optical absorption coefficient, μ is the thermal diffusion length of the sample and μ is the ratio of heat capacity of media.

OPEN CELL PHOTOACOUSTIC

The modified Rosencwaig-Gersho model[2] can be established using the photoacoustic geometry in Figure 2(b). It is known as the transmittance photoacoustic technique or open photoacoustic cell method[3]. The open photoacoustic cell is based on a front illumination configuration and it consists of a commercial electret microphone that receives sound waves directly from a flat slab of solid sample. If we consider the geometry of Figure 2(b) where the microphone is located in the backing position and the sample is opaque, the pressure variation could be expressed as

$$P = \frac{\gamma P_o I_o (\alpha_g \alpha_s)^{1/2}}{2\pi l_g T_o k_s f} \frac{e^{j(\omega t - \pi/2)}}{\sinh(\sigma_s l_s)} \quad (2)$$

For thermally thin samples, the pressure variation is dependent on $(f)^{-3/2}$ and for thermally thick samples, the signal decays exponentially with frequency, $S \propto (1/f)e^{-b\sqrt{f}}$. By fitting the experimental data to the equation for thermally thick samples, the thermal diffusivity is obtained as $\alpha_s = (\pi l_s^2 / b^2)$ [4].

Development of Photoacoustic Technique in Basic Research

Figure 3(a) shows the experimental setup for photoacoustic spectroscopy which could be used for open or closed cell configurations. The light source could be a white light that (Halogen or Xe arc lamp, 150Watts to 1000 Watts) passes through a monochromator (180 nm to 3800 nm) or a laser source. A mechanical chopper modulates the light and the beam goes to the photoacoustic cell (closed or open photoacoustic cell). Closed photoacoustic cells must have an optical window to allow radiation to reach the test sample placed inside the photoacoustic chamber.

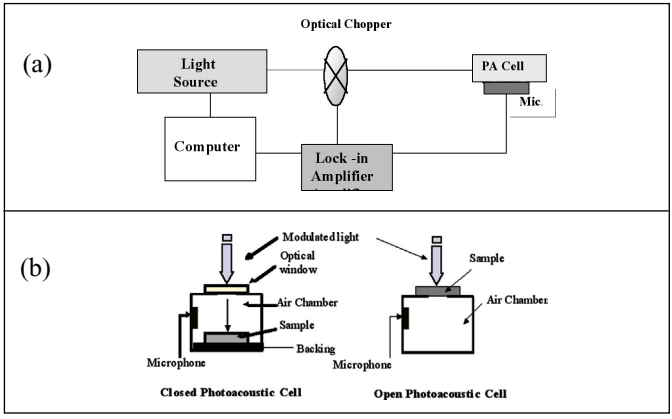

Figure 3 Basic photoacoustic experimental set up

A chopper driver (1 Hz to 3 kHz) gives the pulse reference that synchronizes the photoacoustic signal (normally 1 μV to 200 mV) collected by the microphone (20 mV/Pa to 50 mV/Pa), which is monitored by a lock-in amplifier. The photoacoustic experiment can normally be carried out in two modes i.e. wavelength scan or modulation frequency scan. For the open photoacoustic cell the norm is to use a fixed wavelength (UV, VIS or IR), such as a laser beam or a white light source, with a dedicated monochromator. The PA cell associated with open or closed cell configuration is shown in Figure 3(b). These two photoacoustic techniques (open photoacoustic cell[5-7] and closed cell[8-10]) have been used for characterizing the thermal and optical properties of liquids and solid samples.

PHOTOACOUSTIC - MODULATION FREQUENCY SCAN

If the light is absorbed in a length l_β, the only detectable heat is inside the thermal diffusion length $\mu=(\alpha / \pi f)^{1/2}$, which gives the "thermal skin depth" of the sample. With varying modulation frequencies f, one can perform the depth profiling of a layered sample. The parameter μ is used to classify the sample either as thermally thick ($\mu_s << l_s$) or thermally thin ($\mu_s >> l_s$). When $\mu_s = l_s$ we can define the critical frequency $f_c = (\alpha / \pi f)_s^2$), which is applied for both spectroscopic analysis (varying wavelength of the excitation source) and thermal properties measurement (fixed excitation wavelength and varying modulation frequency). Figure 4 explains the principle of depth profiling photoacoustic measurement using the modulation frequency scan technique.

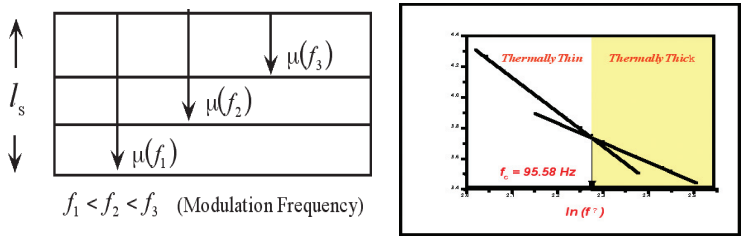

Figure 4 Depth profiling measurement technique

Study of Non-radiative Recombination Mechanisms in Semiconductors

In semiconductors, three fundamental heat sources are well identified i.e. due to intraband thermalization of the photoexcited carriers generated by the absorption of incident light, the non-radiative recombination of photoexcited carriers in the bulk and on the surface (bulk and surface recombination processes). It has been found that for optically opaque semiconductors and in the region where the non-radiative recombination processes are the dominant processes, the photoacoustic signal amplitude and phase are given as:

$$\partial P = \frac{2\varepsilon I_o P_o F}{T_o l_g k_s D\gamma\tau\sigma_g}\left[\frac{1}{\sigma_s^2 - \gamma^2} + \frac{\nu\tau}{\sigma_s}\right] \tag{3}$$

$$\phi = \pi/2 + \tan\left[\frac{(aD/\nu)(\omega\tau_{eff} + 1)}{(aD/\nu)(1 - \omega\tau_{eff}) - 1 - (\omega\tau_{eff})}\right] \tag{4}$$

where

$$\tau_{eff} = \tau\left[(D/\alpha_s) - 1\right]$$

Here ε is the ratio between the gap energy E_g of the semiconductor and the photon energy of the incident light of intensity I_o, $\gamma=(D\tau)^{-1/2}$, where D is the carrier diffusion coefficient and τ is the carrier bulk recombination time, and F is a parameter independent of the modulation frequency *f* which depends on τ and on the surface recombination velocity, ν.

This technique was used to characterize the thermal and carrier transport properties of Si (different thickness and different surface quality, Figure 5)[11] and metal-silicides (Au/n-Si. Au/p-Si, Ag/n-Si and Ag/p-Si (table 1))[12, 13]. The sensitivity of the technique provides a way of simultaneously measuring the thermal and carrier parameters of semiconductor compounds. The results related to this study have been reported in journals and conferences[11-14]. Obviously, the surface recombination velocity increases after the formation of silicide compounds, which is likely to be an n-type semiconductor. This phenomenon was clearly observed for Ag (150 nm)/p-Si sample annealed at 350°C for one hour. However, this important property was not clearly shown by Au/Si interfaces. Table 1 shows the thermal, electrical and carrier transport properties of Si and Ag/p-Si systems obtained by OPC measurements and conventional four point probe technique.

Table 1 Carrier transport parameters for metal-silicide obtained by frequency scan PA.

Sample / Physical Parameters	p-Si	Ag/p-Si as deposited (l_{Au} = 100 nm)	Ag/p-Si annealed at 350°C (l_{Au} = 100 nm)	Ag/p-Si as deposited (l_{Au} = 150 nm)	Ag/p-Si annealed at 350°C (l_{Au} = 150 nm)
Thermal Diffusivity, (cm^2/s)	1.063	0.850	0.800	0.849	0.801
Diffusion Coefficient, (cm^2/s)	14.99	15.00	14.99	14.99	15.08
Surface Recombination Velocity (cm/s)	390.3	262.6	431.1	297.5	432.8
Surface Recombination Lifetime (μs)	13.55	24.96	9.86	19.62	9.77
Resistivity (Ωcm)	50.608	0.033	curve	0.021	Schottky curve
Conductivity (S/cm)	0.019	30.303	curve	47.619	Schottky curve

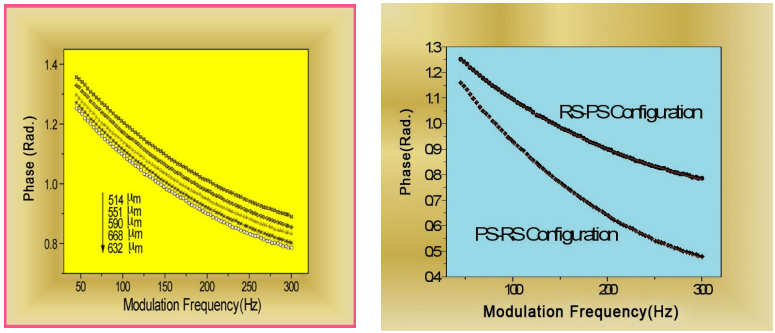

Figure 5 a). Photoacoustic phase signal at different sample thickness. b). effect surface quality to photoacoustic phase signal. PS-RS is polish-Rough surface.

Optical and Thermal Characterization of Materials

In this application, the materials could be semiconductors, ceramics, polymers, woods and metals. The main physical parameters measured in this aspect are thermal diffusivity, thermal effusivity, optical absorption and monitoring of chemical processes.

a) Thermal Diffusivity, α

In order to perform the photoacoustic measurements for thermal diffusivity, circular pieces of the sample (about 300 μm thickness, 10 mm radius) are needed and can be prepared by pressing the powder at a particular pressure (≈Mpa). The sample could be arranged in such a way as to use the a closed photoacoustic cell or an open photoacoustic cell detection technique. Both the amplitude and phase signals are recorded as a function of the light modulation frequency, *f,* using a lock-in amplifier interfaced with a personal computer. By using the Rosencwaig and Gersho theory[2] for a closed photoacoustic cell or a modified R-G theory for an open photoacoustic cell detection technique[17], the thermal diffusivity

values of the samples are obtained using fitting procedures for the amplitude and phase photoacoustic signals.

Table 2 Thermal characterization of 3 types of Malaysian hardwoods

Commercial names	Botanical names	Density (kg/m^3)	Thermal Diffusivity ($x\ 10^{-7}\ m^2/s$)	Heat Capacity ($x\ 10^6\ Jm^{-2}K^{-1}$)	Thermal Conductivity ($x10^{-1}\ Wm^{-1}K^{-1}$)	Thermal Effusivity ($Ws^{1/2}m^{-2}K^{-1}$)
Heavy Hardwoods						
Merbau	*Intsia palembanica*	800	1.40	1.13	1.58	422
Tembusu	*Fagrea spp.*	800	1.34	1.37	1.89	501
Chengal	*Neobalanocarpus heimii*	945	1.66	1.56	2.59	635
Keranji	*Dialium spp.*	960	1.41	1.39	1.95	521
Giam	*Hopea spp.*	975	1.37	1.75	2.39	647
Kekatong	*Cynometra spp.*	975	1.45	1.26	1.83	479
Balau	*Shorea spp.*	975	1.39	1.08	1.50	403
Bitis	*Madhuca utilis*	1105	1.36	1.41	1.91	519
Medium Hardwoods						
Simpoh	*Dillenia spp.*	735	1.35	1.70	2.26	621
Kempas	*Koompassia malaccensis*	880	1.56	1.61	2.51	635
Rengas	*Gluta spp., Melanochyla spp.*	835	1.42	1.76	2.49	663
Punah	*Tetramerista glabra*	720	1.33	1.39	1.84	506
Tualang	*Koompassia excelsa*	835	1.61	1.10	1.77	440
Kulim	*Scorodocarpus*	835	1.23	1.73	2.13	607
Keruing	*Dipterocarpus*	880	1.17	1.63	1.91	557
Kasai	*Pometia spp.*	800	1.46	1.12	1.64	428
Light Hardwoods						
Dark Red Meranti	*Shorea spp.*	705	1.51	1.09	1.65	423
Gerutu	*Parashorea spp.*	690	1.52	0.89	1.35	346
Medang	*Spp. of Lauraceae*	610	1.45	1.09	1.58	414
Merawan	*Hopea spp.*	690	1.36	1.07	1.46	394
White Meranti	*Shorea spp.*	675	1.45	1.05	1.52	400
Melunak	*Pentace spp.*	755	1.32	0.90	1.19	327
Kungkur	*Pithecellobium spp.*	675	1.46	1.09	1.59	415
Bintangor	*Calophyllum spp.*	690	1.36	1.10	1.49	405

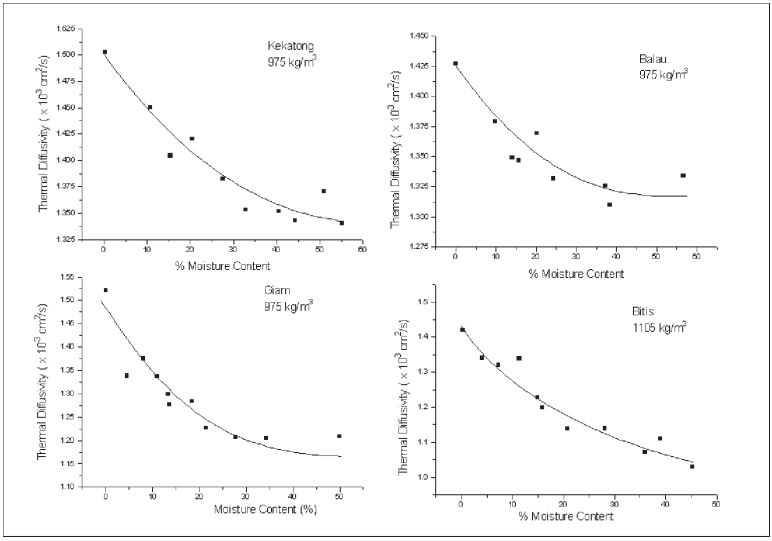

Figure 6 The effect of the variable moisture content on thermal diffusivity of Malaysian hardwood

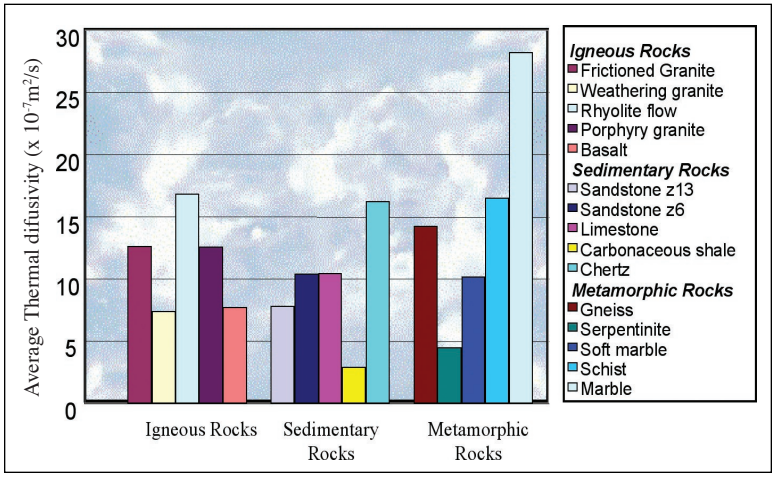

Figure 7 Thermal characterization of 3 types of rocks found in Peninsular Malaysia

Table 2 shows part of our thermal diffusivity measurements of Malaysian hard woods[15-18]. Since the moisture content of the sample plays an important role, the effects of moisture on thermal diffusivity are displayed in Figure 6. The results revealed that the technique is a useful tool, which is capable of effectively measuring the thermal diffusivity values in the range of moisture content (0-20) %. Other natural samples including igneous, sedimentary and metamorphic rock samples were extensively measured using this method (Figure 7)[19].

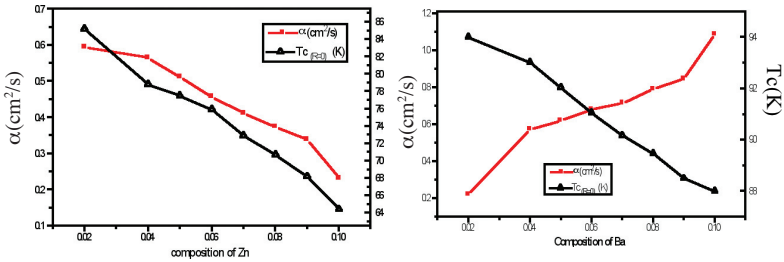

Figure 8 Variation of thermal diffusivity of superconducting samples with composition.

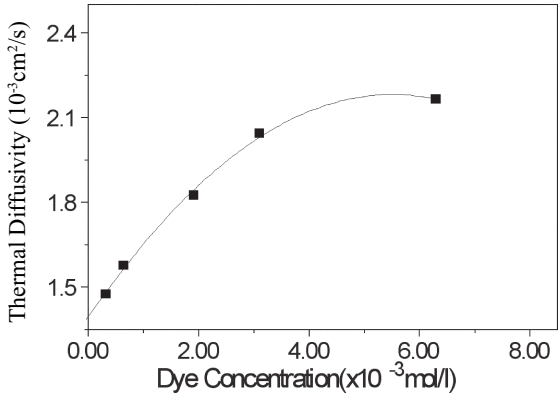

Figure 9 Thermal diffusivity of PMMA doped with R6G.

Recently a study was been carried out to determine the thermal diffusivity of ceramic superconductor (Figure 8)[7, 20-22] and R6G doped PMMA (Figure 9)[23]. The results revealed that a good correlation exists between thermal diffusivity and the T_c value of the investigated superconductor samples.

Wood products, such as abrasive papers (Figure 10)[24], commercial papers (Figure 11)[25] and MDF woods were also studied in our laboratory[19]. Generally the PA method can be used as an objective non-destructive technique with a simple experimental setup. Only a small sample size is needed and the approach can be used on various types of sample. By knowing the sample density and using the PA technique together with the continuous photothermal technique, we can characterize the thermal properties of samples completely, such as measurements of thermal diffusivity, thermal conductivity and specific heat[19].

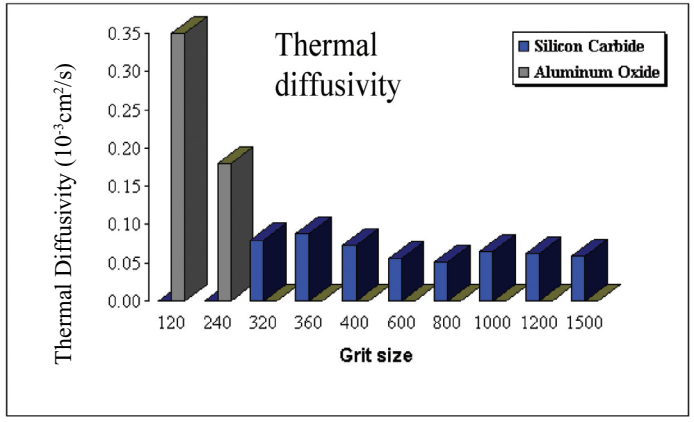

Figure 10 Thermal diffusivity of silicon carbide and aluminum oxide abrasive papers.

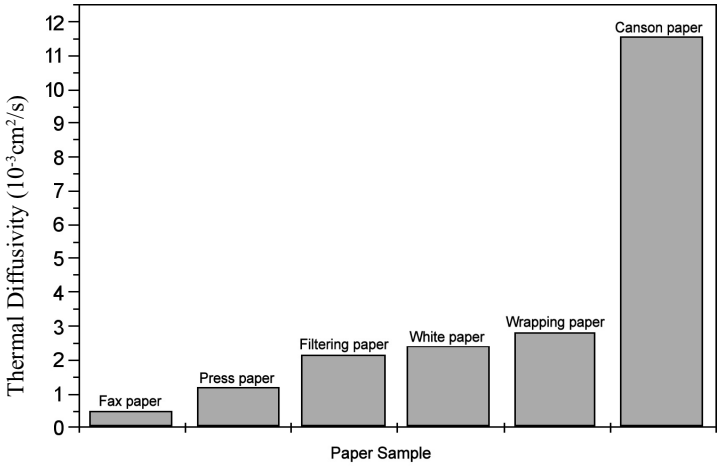

Figure 11 Thermal diffusivity of commercial papers

b) Thermal Effusivity, ε

Thermal effusivity values can be related to thermal diffusivity as, $\alpha = k^2 / \varepsilon^2$. Hence, by knowing ε and α, the thermal conductivity of the sample can be determined without measuring the ρC_p, values. For thermal effusivity measurements, the sample position is similar to those used in the thermal diffusivity measurement. To increase sensitivity and make it suitable for liquid samples we developed a new photoacoustic configuration (Figure 12)[26]. In this setup a modulated laser beam (632.8 nm, 30 mWatts) was focused onto an Al disc (thickness ≅100 μm) and the sample was attached to the opposite end of the illumination side. For liquid samples the Al disc can be the bottom plate of the liquid cell. However, for solid samples, a thin thermal paste is applied to create good thermal contact between the sample and the Al disc. With this new configuration and using a procedure developed based on the R-G model, thermal effusivity could be easily obtained by taking the

ratio of photoacoustic signal of thin Al disc to the photoacoustic signal when the sample has thermal contact with the Al absorber, $(\delta P_f)/(\delta P_F^A)$.

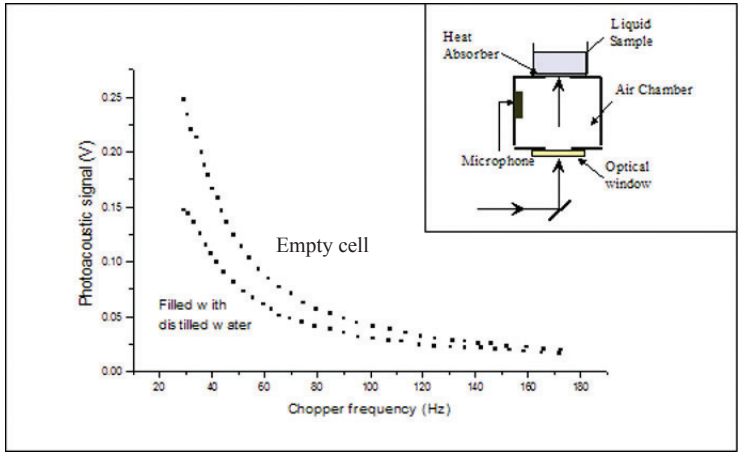

Figure 12 Measurement of thermal effusivity by photoacoustic technique.

This photoacoustic configuration has also been used by our Applied Optics Group to study the thermal effusivity of edible oils (13 samples), glycerol and creamy samples (body lotion, and hair cream)[27]. PA signal of Al disc and water in contact with Al shown in Figure 12 indicate that the method is sensitive enough to characterise the thermal properties of liquid samples.

Table 3 Thermal Effusivity of Various Samples Obtained By Photoacoustic Technique

Liquid sample	**Thermal effusivity e_s ($Ws^{1/2}cm^{-2}°C^{-1}$)**	**Literature Values[28] e_s ($Ws^{1/2}cm^{-2}°C^{-1}$)**
Distilled water	0.1592	0.1588
Glycerol	0.1072	0.0934
Palm Olein	0.060	NA
Super Olein	0.058	NA
Engine Oil	0.055	0.0435
Lubricant	0.051	NA
Body lotion	0.136	NA
Hair cream (Code 10)	0.108	NA
Hair cream (Byrcream)	0.092	NA
Edible Oil		
Canola oil	0.0559	N/A
Coconut oil	0.0596	N/A
Corn oil	0.0537	0.064
Grape seed oil	0.0423	N/A
Groundnut oil	0.0588	N/A
Hazelnut oil	0.0528	N/A
Olive oil	0.0549	N/A
Safflower oil	0.0524	N/A
Saseme oil	0.0539	N/A
Sunflower oil	0.0766	N/A
Soya bean oil	0.0541	N/A
Walnut oil	0.0571	N/A
Cooking oil (Knife brand)	0.0643	N/A

Part of our results on the thermal effusivity of oils and lubricants are listed in Table 3. With regard to the capabilities of the methods and procedures that we have developed; the on going research by our Applied Optics Group is focused on the determination of water in transmission oils, crude oils and lubricants.

WAVELENGTH SCAN PHOTOACOUSTIC

Optical Absorption and Band gap Energy, E_g

For the purpose of measuring band gap energy of semiconducting materials the photoacoustic optical absorption spectra was obtained in the range of 300-1100 nm. A home made photoacoustic spectrometer was developed which consisted of a Xe lamp (450 Watts), a variable frequency mechanical chopper (normally set at 10 Hz and 33 Hz), a monochromator and a photoacoustic cell fitted with a highly sensitive microphone. The sample was placed in a cylindrical chamber and the photoacoustic signal obtained was detected by a lock-in amplifier (SR530) interfaced to a personal computer, which simultaneously displayed the wavelength-dependent signal amplitude and phase which described the absorption spectra of the samples.

Each band gap value was obtained by considering that the band gap energy is located at the position where the maximum derivative at the edge of this photoacoustic spectrum occurs. The other alternative to obtain the energy gap is by directly plotting absorption coefficients (photoacoustic signal) as a function of optical energy.

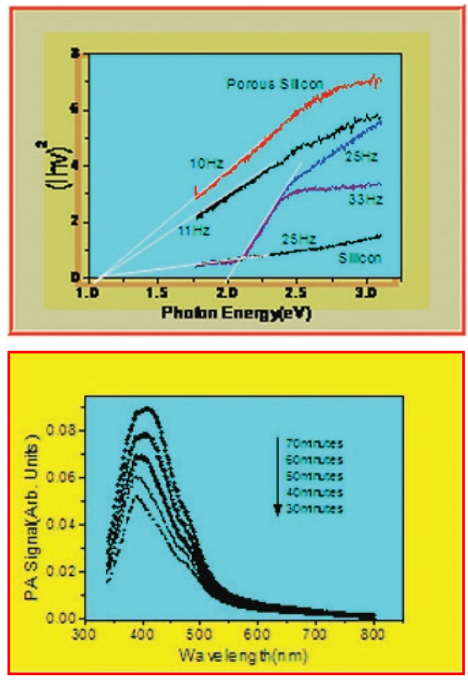

Figure 13 Optical absorption and energy band gap of porous silicon

Recently we measured the optical band gap of porous silicon (Figure 13)[29, 30]. The main advantage of this method is that measurements can be done simultaneously for the porous layer and the silicon substrate. Typical values of the energy band gap for porous silicon and silicon substrate obtained were 2.08 eV and 1.06 eV respectively. Now, we are actively preparing, characterizing and studying the recombination processes in semiconductor samples with different porosity by measuring the non-radiative decay phenomena.

REAL TIME MEASUREMENT

Monitoring Photobleaching

Several studies on photo-induced bleaching of organic dye molecules trapped in transparent solid matrices have been carried out over the past few decades. Such studies have great importance due to the various applications of these materials, such as for active laser element, optical data storage and processing, optical waveguides etc. Since 1968, methylene blue (MB) has been used as a holographic recording medium for formation of high spatial frequency amplitude and phase holograms with a conventional He-Ne laser as a light source. Several techniques have been used to study the photostability of methylene blue doped in polymer matrix. Our group has carried out, for the first time, photostability studies on methylene blue (MB) doped in gelatine, poly(methylmethacrylate) PMMA and Poly(vinyl alcohol)(PVA) matrices using the photoacoustic (PA) technique and fibre optics spectrophotometer[31-37]. Photoacoustic signals were measured as a function of time, chopping frequency and the beam power. The results indicate that the photobleaching rate is proportional to the incident laser power (260-300 mWatts) and decreases with increasing concentration of dopant molecules. Methylene blue embedded in PVA shows the highest photobleaching rate followed by gelatine and PMMA (Figure 14). The increase of photobleaching rate in PVA is also noticeable when the beam power is increased. The thermal diffusivity values obtained for bleached PMMA, PVA and gelatine were 1.46×10^{-3} cm^2/s, 0.83×10^{-3} cm^2/s and 1.10×10^{-3} cm^2/s respectively.

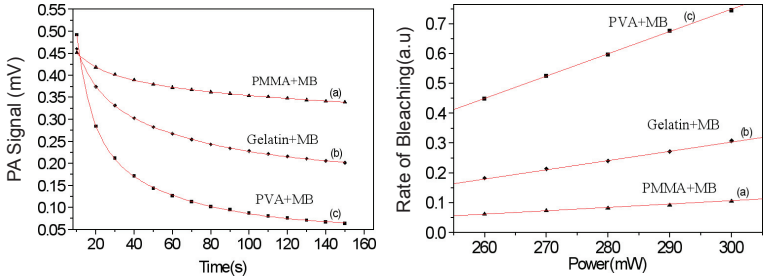

Figure 14 Photoacoustic signal and photobleaching rate of PMMA, gelatin and PVA doped with MB

It is believed that the photobleaching mechanism of MB occurred through photochemical reactions which can be described as follows[38] :

$$MB \xrightarrow{h\upsilon} {}^{1}MB^{*} \rightarrow {}^{3}MB^{*} \xrightarrow{+e^{-}} \text{Intermediate product} \rightarrow \text{leuco MB} \quad (5)$$

$$\searrow^{MB} \quad \searrow^{MB}$$

$$2MB \quad 2MB$$

Other photobleaching studies include MB doped in sago starch, chitosan and sago starch blends[38,39]. Studies of dye concentration effect and nature of solvents on the photobleaching process are being carried out in our laboratory using both the photoacoustic spectrometer and fiber optics spectrophotometer.

PHOTOACOUSTIC GAS SENSOR

Photoacoustic and photothermal gas sensors are better than surface sensitive devices with respect to reliability, because they use a volume effect. Using an optical filter for a particular wavelength will open up a wide field of applications. Process control, atmospheric control, environmental monitoring and HVAC (heat, ventilation and air conditioning) are areas where these sensors could be used. This

flexibility is another attraction for both manufacturers and users. A number of world class companies have chosen photoacoustic sensors for these reasons.

To be able to comply with current and future regulations for air-pollution control and safety, process industry, public institutions and private households strongly demand high sensitivity and low cost reliable gas sensors. In satisfying this demand, the photoacoustic sensor has met the current status of sensor development which can be categorized as a low cost, highly reliable, highly selective and sufficiently sensitive in many applications.

a) The Principle of a Photoacoustic Gas Sensor

In a conventional photoacoustic gas sensor, the gas to be analyzed is pumped into an absorption chamber that is sealed with mechanical valves during measurement. The cell is irradiated with modulated IR light filtered at the wavelengths at which the gases of interest are strongly absorbed.

The photoacoustic principle for the gas sensor is based on the phenomenon where when a gas is irradiated with infrared (IR) light, it absorbs incident radiation within its own characteristic absorption spectrum. The amount of absorbed radiation, which follows the Beer-Lambert absorption law, is a function of gas concentration, path length and the specific absorption coefficient of the gas. This absorbed radiation, which for a very short period of time is stored as intra-molecular vibrational-rotational energy, is quickly released by relaxation to translational energy. Translational energy is equivalent to heat and when the absorption chamber is sealed, this will cause the pressure to rise. Each gas has a unique IR spectrum and strong absorption takes place only at a certain wavelength. When the incident light is modulated at a given frequency, a periodic pressure change is generated in the absorption chamber. This photoacoustic

signal can be detected or measured with a sensitive microphone (Figure 15a*)*.

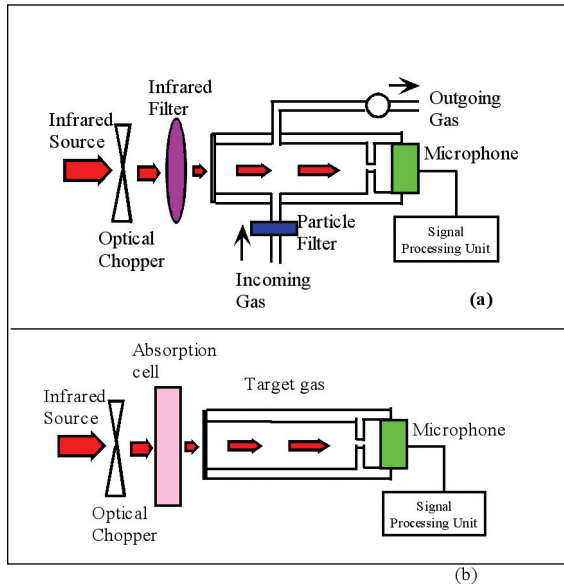

Figure 15 Principle of photoacoustic gas sensor

b) The principle of Selective Photoacoustic Gas Sensor

A schematic diagram of the operation principles of the selective photoacoustic gas sensor is shown in Figure 15b together with the required electronics. In this sensor, the absorption cell is pre-filled with the actual target gas and sealed. When modulated IR light is passed into the absorption cell, a photoacoustic signal is generated. If the target gas is introduced in the absorption path outside the cell, a reduction of the photoacoustic signal is observed. This reduction is nearly proportional to the concentration of target gas in the absorption path.

It is important to note that a photoacoustic signal reduction is observed only if the gas outside the cell is absorbing the IR radiation at the same wavelength as that inside the pre-filled cell. In this way a good selective gas sensor can be obtained without the use of any additional optical filtering. The gas inside the absorption cell then acts as a narrow band selective filter. Another important point is that in a conventional photoacoustic sensor the test gas has to be continuously pumped into and out of the absorption cell. In this new concept photoacoustic sensor, no mechanical pump or valves are required. In testing the performance of the photoacoustic sensor we have developed a new PA cell gas sensor. Figure 16 shows the preliminary results of our measurements.

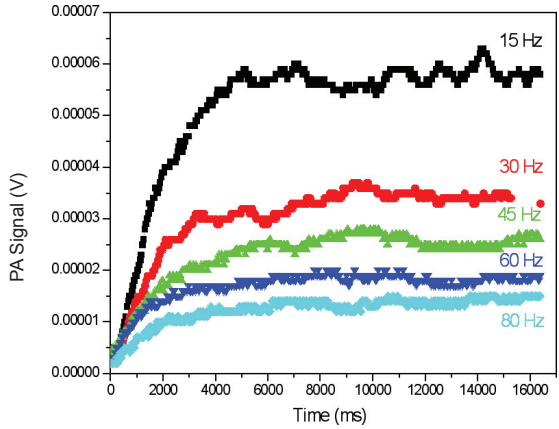

Figure 16 Photoacoustic signal of air detected at different chopping frequencies. The Photoacoustic cell is a cylindrical glass of 12 mm diameter and 10 cm in length

Photoacoustic Industrial Applications

The improvement in sensitivity and accuracy of photoacoustic techniques has led to increasing interest for application in industry. The transfer from laboratory scale to industrial plant scale however, requires additional optimization comprising cost considerations, security and reliability aspects. Most importantly, the device has to work under special conditions as dictated by industry. This in general also includes a considerable reduction in the available measuring time whereas in laboratory experiments extended frequency scans and multiple repletion of the measurement are applied to increase the signal to noise ratio. At industrial scale, the available measuring time is limited to a few pre-selected modulation frequencies or to a time limited pulse experiment. In addition, sophisticated electronic signal treatments are excluded due to cost reasons.

So far, only photoacoustic gas detector/sensor measurement techniques have been successfully transferred to actual industrial applications. Two main areas which have been satisfied are concerned with quality control and control of pollution and impurity in products or the environment. An impressive example of an industrial adapted device is the gas trace detectors of different companies (e.g. Innova-Air Techinstruments[40], MTEC Photoacoustic Inc. [41], Quantum Northwest[42], MSA Chargard[43], USA Instruments Inc.[44] and Bruel and Kjaer[45]). These detectors, which are based on the photoacoustic effect from gases, are distinguished by their very simple, robust and space saving designs as compared to spectroscopic arrangements on laboratory scale. The latter generally works with tunable lasers adjusted to an absorption line of the trace molecule or uses spectrometers to scan the whole absorption spectrum. In contrast, the industrial detector of photoacoustic sensor is adapted to different gases by simply changing a narrow band IR-filter in front of the lamp. The use of only a limited number of

electronic, mechanical and optical components makes the system more reliable and less sensitive to environmental changes. On the other hand, most consumers request simple handling of the detector output which is achieved by automisation and stabilisation with an online computer. The development of adequate software has become one of the key requirements in modern technology for industrial application. This is also true for the photothermal devices developed and applied for quality control. In this aspect, Innova Air Techinstruments is the leading company which produces various types of gas sensors based on photoacoustic effect. Two companies, i.e. Jenopik in Europe[46] and Thermawave[47] in US, sell apparatus for quality control of silicon wafers which are based on optically and thermally modulated optical reflection from semiconductors. Infrared radiometry, the remote detection technique using thermal waves, for corrosion detection in airplanes has been developed by the Thermalwave group in US, led by Dr. R. L. Thomas[47].

SURFACE PLASMON RESONANCE

The phenomenon of anomalous diffraction on diffraction gratings due to the excitation of surface plasma waves was first described at the beginning of the twentieth century by Wood [48]. In the late sixties, optical excitation of surface plasmons by method of attenuated total reflection was demonstrated by Kretschmann[49] (Figure 17) and Otto[50]. Since then, surface plasmons have been intensively studied and their major properties assessed.

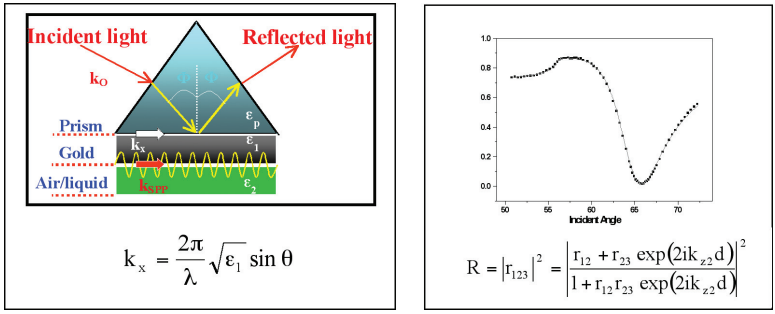

Figure 17 Kretschmann configuration and surface plasmon resonance curve

Surface plasmon resonance is a charge-density oscillation that may exist at the interface of two types of media with dielectric constants of opposite signs, for instance, a metal and a dielectric. The charge density wave is associated with electromagnetic waves, the field vectors of which reach their maximum amplitude at the interface and decay evanescently into both media. This surface plasma wave (SPW) is a p-polarized wave. The propagation constant of the surface plasma wave propagating at the interface between a semi-infinite dielectric and metal is given by the following expression:

$$k_{SPP} = \frac{\omega}{c}\sqrt{\left(\frac{\varepsilon_m \varepsilon_s}{\varepsilon_m + \varepsilon_s}\right)} \qquad (6)$$

At optical wavelengths, this condition is fulfilled by several metals,[51] of which gold and silver are the most commonly used.

Surface plasmon resonance (SPR) reflectivity measurements are surface-sensitive, spectroscopic methods that can be used to characterize the thickness and/or index of refraction of ultra thin organic and biopolymer films on noble metal (Au, Ag, Cu) surfaces. Surface plasmon resonance spectroscopy has become widely

used in the fields of chemistry and biochemistry to characterize biological surfaces and to monitor binding events. The success of these SPR measurements is primarily due to three factors: (i) with SPR spectroscopy the kinetics of biomolecular interactions can be measured in real time, (ii) the adsorption of unlabeled analyte molecules to the surface can be monitored, and (iii) SPR has a high degree of surface sensitivity that allows weakly bound interactions to be monitored in the presence of excess solution species. SPR spectroscopy has been used to monitor such events as antibody-antigen binding, DNA hybridization and protein-DNA interactions.

a) Refractive Index Measurement

Since the technique is sensitive to the optical properties of the third medium, it was used effectively as a tool in the determination of the refractive index of liquids, solids (thin film form), powders and emulsions[51-59]. Figure 18 shows several different types of samples that have been tested in our laboratory using the SPR technique. Their values (real and imaginary part of optical dielectric constants) are displayed in Table 4.

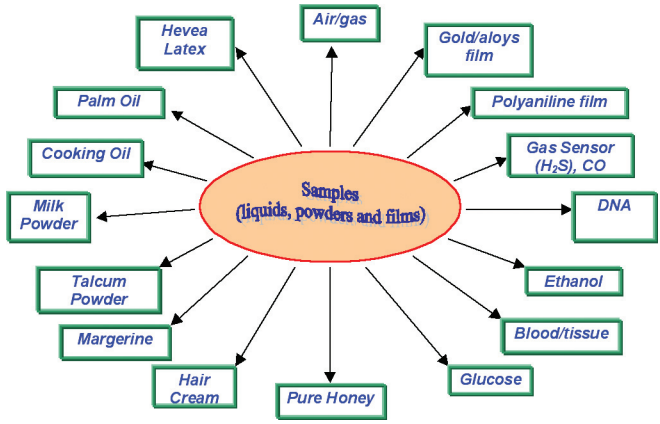

Figure 18 Solid, powder, liquid, emulsion and liquid samples tested by Surface Plasmon Resonance technique

Table 4 The values of the real and imaginary part of optical dielectric constants measured by Surface Plasmon Resonance Technique

Sample	**ε_r (λ=632.8 nm)**	**ε_i (λ=632.8 nm)**
Distilled water	1.768	0.030
Etahanol (15 %)	1.753	0.015
Ink (0.13 %)	1.788	0.010
Latex (37.7 %)	2.075	0.035
Cooking Oil	2.170	0.010
Honey	2.250	0.010
Magerine	2.170	0.023
Hair Cream	1.975	0.020
Talc	1.010	0.020
Milk Powder	1.010	0.005
Gold (99.999 %)	-10.50 (60.7nm)	1.97
Gold (22 Carat)	-10.14 (63.8nm)	2.70
Gold (18 Carat)	-10.01 (70.3nm)	2.45
RBD Palm Oil	2.143	0.018
RBD Palm Olein	2.146	0.015
RBD Palm Stearin	2.143	0.017

b) Surface Plasmon Resonance Optical Sensor

Generally, an SPR optical sensor comprises of an optical system, a transducing medium which interrelates the optical and (bio)-chemical domains and an electronic system supporting the optoelectronic components of the sensor and allowing data processing. The transducing medium transforms changes in the quantity of interest into changes in the refractive index which may be determined by optically exciting the SPR. The optical part of the SPR sensor contains a source of optical radiation and an optical structure in which the SPW is excited and measured. Major properties of an SPR sensor are determined by the properties of the sensor's subsystems. Leiedberg (1993)[52] was the first researcher who used the SPR as a chemical sensor. The sensor sensitivity, stability and resolution depend on properties of both the optical system and the transducing medium. The selectivity and response time of the sensor are primarily determined by the properties of the transducing medium.

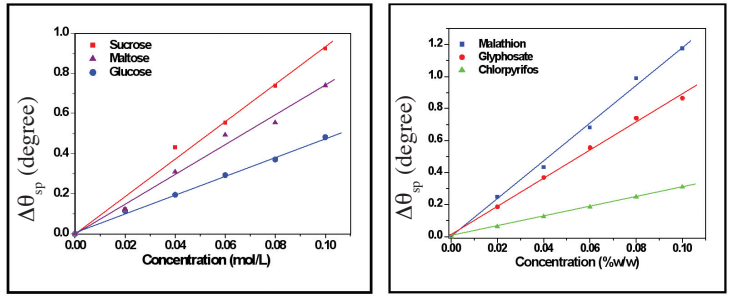

Figure 19 Surface plasmon resonance as a function of concentration for discaride and pesticide samples

Surface plasmon resonance (SPR) was used for biosensing for the first time in 1983[60] and for imaging applications in 1987[61]. In our

laboratory this type of optical sensor has been used for detection of various samples, such as detection of sacharides, pesticides, DNA and antigen, and ethanol in solution, detection of H_2S and CO gases in air, and measuring moisture content in honey, hevea latex and milk[62-67]. Figure 19 shows the typical sensitivity and detection limit of our SPR optical sensor for disaccharides and pesticides samples in water. Obviously the chemical structure and kinetic behavior of the molecule involved in the tested sample affect the sensitivity and detection limit. Other samples tested by this method include, G90 (Trichloroisacyanuric acid), G70 (Calcium hypochlorite), swimming pool water, fresh milk and fatty hydroxamic acid[68].

b) Kinetic Study of Adsorption Molecule

The thin gold layer in the SPR Kretschmann configuration creates the physical conditions required for surface plasmon resonance. Essentially, SPR detects changes in mass of dissolved material in the aqueous layer close to the sensor chip surface by measuring changes in the refractive index. When molecules in the test solution bind to a target molecule/gold surface, the mass increases but when they dissociate, the mass falls.

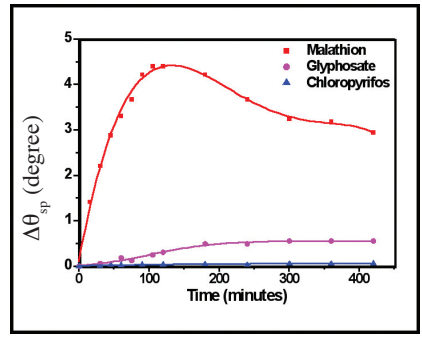

Figure 20 Kinetic response of DNA and pesticide molecules to gold thin layer

This simple principle forms the basis of the SPR sensorgram which provides continuous real-time monitoring of the association and dissociation of the interacting molecules. The sensorgram provides quantitative information in real-time on specificity of binding, active concentration of molecule in a sample, kinetics and affinity. Studies can be carried out in opaque and transparent samples. Figure 20 shows the examples of kinetic behaviour of DNA and pesticide molecules[69]. In general, the association process can be explained as:

$$\Delta\theta \equiv \Delta\theta_{\infty}[1 - \exp(-t/\tau)^{\beta}] \qquad (7)$$

where τ and β are time constant and stretching coefficient respectively.

SURFACE PLASMON RESONANCE IN INDUSTRY

In this area, SPR, as a surface-oriented method, has shown great potential for affinity biosensors, allowing real-time analysis of biospecific interactions without the use of labeled molecules. The SPR sensor technology has been commercialized by several companies and has become a leading technology in the field of direct real-time observation of biomolecular interactions.

Systematic development of the SPR biosensor technology by Swedish BIAcore AB (originally Pharmacia Biosensor AB) led to the launch of the first commercial SPR biosensor in 1990. Since then the BIAcore sensor technology has been further refined in terms of speed, throughput and accuracy. Currently, BIAcore offers several models of SPR biosensors (BIACORE® 3000, BIACORE® 2000, BIACORE® X, BIACORE® 1000, BiacoreQuant™)[70]. Increasing interest in commercialization of optical biosensor systems has resulted in the development of another SPR biosensor system (TI-SPR-1 Experimenters Kit, Spreeta™ Evaluation Kit) by Texas

Instruments (USA).[71] Both BIAcore and Texas Instrument sensor systems utilize prism couplers. An SPR biosensor instrument relying on wavelength interrogation of SPW in grating-based structures is offered by Quantech (USA)[72]. Another SPR biosensor system (called IBIS) developed by Intersens Instruments was also presented by Xantec (Germany)[73].

CURRENT AND FUTURE RESEARCH IN PHOTOACOUSTIC AND SURFACE PLASMON RESONANCE

It is difficult to predict what the future of basic research in photoacoustic and surface plasmon resonance will hold. However, with current developments in lasers such as the diode laser and OPO laser, basic research in photoacoustic/SPR will focus on the improvement of sensitivity and good signal to noise ratio. With better instruments and fuller understanding of the mechanisms involved, photoacoustic/SPR will be an analytical tool which enables steady improvements to be made in sensitivity, precision, and accuracy. Demand for the photoacoustic/SPR technique as an analytical instrument package has been steadily increasing since the OPO laser was used successfully in photoacoustic/SPR experiments.

FUTURE OUTLOOK

There is a need for detection and analysis of chemical and biochemical substances in many important areas including medicine, environmental monitoring, biotechnology, drug and food monitoring. Photoacoustic and surface plasmon resonance sensor technology hold potential for applications in these areas. Currently, SPR biosensor devices compete with other types of biosensors such as real-time optical immunosensors[74]. However,

the major competitors of biosensors are immunoassays which are commonly and widely used for determination of numerous important substances and offer low-cost tests of high specificity and sensitivity. Today, the commercially available thermal wave and biosensors cover a very limited area of the (bio)chemical monitoring market aimed primarily at research and analytical laboratories. In order to branch out from specialized laboratories and centralized testing sites and gain a fair share of the (bio)chemical monitoring market, thermal wave and SPR sensors have to compete with existing technologies on the basis of factors such as low cost, ease of use, robustness, sensitivity and stability. It is envisaged that this will drive research and development of thermal wave and SPR-sensing devices in future applications.

Undoubtedly, in the future, the thermal wave and SPR technologies will benefit from the use of optical waveguide technology which offers potential for development of miniaturized, compact and rugged sensing elements with the prospect of fabrication of multiple sensors on a single chip.

CONCLUSION

Apparatus for industrial applications are from the experimental point of view a "slim version" of a laboratory setup. On the other hand, the evaluation of the reliability of the measured data, which in a laboratory environment is the responsibility of the scientist, has to be performed automatically in an industrial device by an online computer equipped with adequate and refined software. We may argue that laboratory experiments with its high standard technical equipment and detailed measurement procedures are not necessary in view of later industrial application of a "slim version". Why can we not immediately develop the simplest setup instead of performing extensive experiments at laboratory scale? The answer is

not simple. If we have to search for the shortest and cheapest path to get from one place to another, we have to explore all possible paths between the two places through extensive investigation in order to select the best and most adequate one. The same holds true for the transfer of photoacoustic and surface plasmon resonance techniques to efficient industrial applications. Both efforts are needed, the thorough and extensive investigation at laboratory scale followed by the adaptation of the most suitable method for industrial needs.

Even though some photoacoustic and surface plasmon resonance methods have been commercialized for industrial and research use, basic research in these two areas is still needed to improve the sensitivity, accuracy and even the selectivity (for gas detection) of the sensors. In addition to that development of compact, robust, small sized portable units need to be considered for industrial use.

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BIOGRAPHY

Dr. W. Mahmood Mat Yunus was born on April 10, 1955 in Bachok, Kelantan. His early education was in Kota Baharu which is where he completed his secondary school education, at Sultan Ismail Petra, Kota Baharu, Kelantan. He subsequently enrolled in Universiti Kebangsaan Malaysia in 1975 and received the degree of Bachelor of Science with Honours in Physics in 1979. In the same year he joined Universiti Pertanian Malaysia[1] as a tutor in the Science Unit of UPM Sarawak. He obtained his Master and Doctoral degrees in Ionization Physics from the University College of Swansea in 1982 and 1985 respectively.

Dr. W. Mahmood Mat Yunus was appointed as a lecturer in 1985 and was promoted to Associate Professor in 1995. He has todate served with UPM for over 23 years. In recognition of his excellent services, the University honoured him with the Excellent Service Award in 1997 and again in 2004, in the highest category.. From 1997 to 1999 he was appointed the Head of the Department of Physics and later reappointed to serve the Department until 2002. He was promoted to the post of professorship on the 1st of May, 2002.

Dr. W. Mahmood truly enjoys teaching and supervising students. He teaches various subjects in physics, such as Introductory Physics, Electromagnetism, Materials Science, Instrumentation, Electromagnetic Wave and Optics, Modern Optics, Electricity and Magnetism and Semiconductor Physics and Devices. Realizing the importance of the optical technique as a tool in characterizing materials and as a non-destructive optical sensor, he successfully developed the Photothermal-Photoacoustic and Surface Plasmon Resonance laboratories in the Department. Dr. W. Mahmood is

1 now Universiti Putra Malaysia

also actively involved in students' supervision and todate he has supervised 50 postgraduates and 60 undergraduate final year students. Since 2000, he has served as an external examiner at UTM, UKM, UM and UiTM.

Over the past twelve years he embarked on research in the fields of Photothermal-Photoacoustic and Surface Plasmon Resonance. Most of the research funding was from the IRPA programs (Ministry of Science, Technology and Environment) and University Putra Malaysia. Todate, he has been awarded several projects under the MOHE and SAGA programs totalling over RM 700,000.00.

In 1994, Dr. W. Mahmood developed a surface plasmon resonance laboratory funded by the Malaysia Toray Science Foundation (MTSF). In 1995 he was awarded a Commonwealth Fellowship to explore Surface Plasmon applications at the University of Exeter, UK. Four years later (1999) he was awarded the "Japan Society for the Promotion of Science (JSPS) Fellowship" to visit University of Kyushu to strengthen his research activities in photoacoustic gas detection. Subsequently in 2002, under the "Japan Society for the Promotion of Science (JSPS)" he worked together with professor Taro Toyoda at the Department of Applied Physics and Chemistry, University of Electro-Communication, Tokyo, Japan to gain knowledge in characterizing semiconductor materials using the photoacoustic technique.

Since 1985 he has been directly responsible for more than 175 papers, related to his specialized field in applied optics, being published in local and international journals and more than 70 papers published in proceedings. He has actively involved himself in presenting working papers and chairing technical sessions at conferences and seminars. Todate, he has contributed more than 200 papers to Seminars/conferences and workshops.

At present Dr. W. Mahmood is a member of the Institute of Physics Malaysia, Optical Society of America and a Fellow of Malaysia Solid State and Technology Society. He has served as a referee for the Journal of Applied Optics (US), Optical Express Journal (US), Int. J. Heat Mass Transfer (UK), Jurnal Sains dan Teknologi Keadaan Pepejal (MAL), Jurnal Sains Malaysiana (MAL) and Jurnal Teknologi (siri C) (MAL). He was also a member of the editorial board of the PERTANIKA journal for more than 9 years (1997 – 2006).

Dr. W. Mahmood truly enjoys serving society as a scientist and will continue to do so in the future.

Photoacoustic Technique

The photoacoustic effect is based on the generation of acoustic waves as a consequence of modulated light absorption. The absorption of the incident energy beam and the subsequent non-radiative de-excitation relaxation processes give rise to a heat source in the sample. In the photoacoustic technique, the sample is enclosed in an airtight cell where the temperature oscillations from the illuminated surface of the sample will be communicated to the gas in the cell. The effect can be sensed by a sensitive microphone that forms part of the inner cell wall. Physically the heating of the sample depends on the optical absorption and the heat diffuses from the sample to the surface. These two factors lend versatility to the technique for studying optical absorption as well as thermal diffusivity of samples. The ultimate goal is to use this technique in determining glucose content in blood by in-situ measurements.

Surface Plasmons Resonance Technique

A surface plasmon (SP) is a collective excitation of the electrons at the interface between a conductor and an insulator. Surface plasmons on a plane surface are non-radiative electromagnetic modes, that is, SPs cannot be generated directly by light nor can they decay spontaneously into photons. The origin of the non-radiative nature of SPs is that the interaction between light and SPs cannot simultaneously satisfy energy and momentum conservation. This restriction can be overcome by increasing the effective wave vector (and hence momentum) of the light using a prism coupling technique.

His interest at present is more in the applications of Surface Plasmon Resonance particularly in multiple thin film systems. The presence of even very thin film results in measurable shifting of the resonance angle and broadening the reflectivity dip. These effects can be utilized to develop devices such as optical modulators and chemical sensors. To further enhance the lab's capabilities in analyzing thin films, he is equipping his ATR reflectivity system with the ability to make ellipsometric measurements for variable angle and variable wavelength measurements.

ACKNOWLEDGEMENT

I am most grateful to Allah SWT for His blessings and guidance. I sincerely thank Universiti Putra Malaysia, the Science Faculty and the Sponsors of the research grants for the opportunities provided to explore and excel in my area of research interest. I am indebted to my late father and mother for their prayers and "doa", support and encouragement in my early upbringing. Special thanks to my wife and our five children for their sacrifices, endless support, love and understanding. Last but not least, my heartfelt gratitude to all my colleagues, friends and students who rendered me support and assistance. No matter the magnitude of your contribution each and every one of you has contributed to my success in one way or another. Thank you.

LIST OF INAUGURAL LECTURES

1. Prof. Dr. Sulaiman M. Yassin
   The Challenge to Communication Research in Extension
   22 July 1989

2. Prof. Ir. Abang Abdullah Abang Ali
   Indigenous Materials and Technology for Low Cost Housing
   30 August 1990

3. Prof. Dr. Abdul Rahman Abdul Razak
   Plant Parasitic Nematodes, Lesser Known Pests of Agricultural Crops
   30 January 1993

4. Prof. Dr. Mohamed Suleiman
   Numerical Solution of Ordinary Differential Equations: A Historical Perspective
   11 December 1993

5. Prof. Dr. Mohd. Ariff Hussein
   Changing Roles of Agricultural Economics
   5 March 1994

6. Prof. Dr. Mohd. Ismail Ahmad
   Marketing Management: Prospects and Challenges for Agriculture
   6 April 1994

7. Prof. Dr. Mohamed Mahyuddin Mohd. Dahan
   The Changing Demand for Livestock Products
   20 April 1994

8. Prof. Dr. Ruth Kiew
   Plant Taxonomy, Biodiversity and Conservation
   11 May 1994

9. Prof. Ir. Dr. Mohd. Zohadie Bardaie
   Engineering Technological Developments Propelling Agriculture into the 21st Century
   28 May 1994

10. Prof. Dr. Shamsuddin Jusop
    Rock, Mineral and Soil
    18 June 1994

11. Prof. Dr. Abdul Salam Abdullah
Natural Toxicants Affecting Animal Health and Production
29 June 1994

12. Prof. Dr. Mohd. Yusof Hussein
Pest Control: A Challenge in Applied Ecology
9 July 1994

13. Prof. Dr. Kapt. Mohd. Ibrahim Haji Mohamed
Managing Challenges in Fisheries Development through Science and Technology
23 July 1994

14. Prof. Dr. Hj. Amat Juhari Moain
Sejarah Keagungan Bahasa Melayu
6 Ogos 1994

15. Prof. Dr. Law Ah Theem
Oil Pollution in the Malaysian Seas
24 September 1994

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Fine Chemicals from Biological Resources: The Wealth from Nature
21 January 1995

17. Prof. Dr. Sheikh Omar Abdul Rahman
Health, Disease and Death in Creatures Great and Small
25 February 1995

18. Prof. Dr. Mohamed Shariff Mohamed Din
Fish Health: An Odyssey through the Asia - Pacific Region
25 March 1995

19. Prof. Dr. Tengku Azmi Tengku Ibrahim
Chromosome Distribution and Production Performance of Water Buffaloes
6 May 1995

20. Prof. Dr. Abdul Hamid Mahmood
Bahasa Melayu sebagai Bahasa Ilmu- Cabaran dan Harapan
10 Jun 1995

21. Prof. Dr. Rahim Md. Sail
Extension Education for Industrialising Malaysia: Trends, Priorities and Emerging Issues
22 July 1995

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The Diminishing Tropical Rain Forest: Causes, Symptoms and Cure
19 August 1995

23. Prof. Dr. Ang Kok Jee
The Evolution of an Environmentally Friendly Hatchery Technology for Udang Galah, the King of Freshwater Prawns and a Glimpse into the Future of Aquaculture in the 21st Century
14 October 1995

24. Prof. Dr. Sharifuddin Haji Abdul Hamid
Management of Highly Weathered Acid Soils for Sustainable Crop Production
28 October 1995

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Fish Processing and Preservation: Recent Advances and Future Directions
9 December 1995

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Pesticide Usage: Concern and Options
10 February 1996

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Microbial Fermentation and Utilization of Agricultural Bioresources and Wastes in Malaysia
2 March 1996

28. Prof. Dr. Wan Sulaiman Wan Harun
Soil Physics: From Glass Beads to Precision Agriculture
16 March 1996

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Sustained Growth and Sustainable Development: Is there a Trade-Off 1 or Malaysia
13 April 1996

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Sharecropping in Perfectly Competitive Markets: A Contradiction in Terms
27 April 1996

31. Prof. Dr. Mohd. Yusuf Sulaiman
Back to the Future with the Sun
18 May 1996

32. Prof. Dr. Abu Bakar Salleh
Enzyme Technology: The Basis for Biotechnological Development
8 June 1996

33. Prof. Dr. Kamel Ariffin Mohd. Atan
The Fascinating Numbers
29 June 1996

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Fungi: Friends or Foes
27 July 1996

35. Prof. Dr. Tan Soon Guan
Genetic Diversity of Some Southeast Asian Animals: Of Buffaloes and Goats and Fishes Too
10 August 1996

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Will Rural Sociology Remain Relevant in the 21st Century?
21 September 1996

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Leptospirosis-A Model for Epidemiology, Diagnosis and Control of Infectious Diseases
16 November 1996

38. Prof. Dr. Marziah Mahmood
Plant Biotechnology - Strategies for Commercialization
21 December 1996

39. Prof. Dr. Ishak Hj. Omar
Market Relationships in the Malaysian Fish Trade: Theory and Application
22 March 1997

40. Prof. Dr. Suhaila Mohamad
Food and Its Healing Power
12 April 1997

41. Prof. Dr. Malay Raj Mukerjee
A Distributed Collaborative Environment for Distance Learning Applications
17 June 1998

42. Prof. Dr. Wong Kai Choo
Advancing the Fruit Industry in Malaysia: A Need to Shift Research Emphasis
15 May 1999

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Avian Respiratory and Immunosuppressive Diseases- A Fatal Attraction
10 July 1999

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Biological Control of Plant Pathogens: Harnessing the Richness of Microbial Diversity
14 August 1999

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The Endomycorrhiza: A Futile Investment?
23 Oktober 1999

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Molecular Plant Virology: The Way Forward
2 February 2000

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Do We Have Enough Clean Air to Breathe?
7 April 2000

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Green Environment, Clean Power
24 June 2000

49. Prof. Dr. Mohd. Ghazali Mohayidin
Managing Change in the Agriculture Sector: The Need for Innovative Educational Initiatives
12 January 2002

50. Prof. Dr. Fatimah Mohd. Arshad
Analisis Pemasaran Pertanian di Malaysia: Keperluan Agenda Pembaharuan
26 Januari 2002

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Fisheries Co-Management: An Institutional Innovation Towards Sustainable Fisheries Industry
28 February 2002

52. Prof. Dr. Gulam Rusul Rahmat Ali
Food Safety: Perspectives and Challenges
23 March 2002

53. Prof. Dr. Zaharah A. Rahman
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13 April 2002

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Productivity Driven Growth: Problems & Possibilities
27 April 2002

55. Prof. Dr. Wan Omar Abdullah
Immunodiagnosis and Vaccination for Brugian Filariasis: Direct Rewards from Research Investments
6 June 2002

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Agro-ento Bioinformation: Towards the Edge of Reality
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Sustainability of Tropical Animal-Agricultural Production Systems: Integration of Dynamic Complex Systems
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The Economics of Exchange Rates in the East Asian Countries
26 October 2002

59. Prof. Dr. Shaik Md. Noor Alam S.M. Hussain
Contractual Justice in Asean: A Comparative View of Coercion
31 October 2002

60. Prof. Dr. Wan Md. Zin Wan Yunus
Chemical Modification of Polymers: Current and Future Routes for Synthesizing New Polymeric Compounds
9 November 2002

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Is the KLSE Efficient? Efficient Market Hypothesis vs Behavioural Finance
23 November 2002

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Road Safety Interventions in Malaysia: How Effective Are They?
21 February 2003

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The New Shares Market: Regulatory Intervention, Forecast Errors and Challenges
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Blueprint for Transformation or Business as Usual? A Structurational Perspective of the Knowledge-Based Economy in Malaysia
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Chemical Diversity of Malaysian Flora: Potential Source of Rich Therapeutic Chemicals
26 July 2003

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An Ecological Approach: A Viable Option for Aquaculture Industry in Malaysia
9 August 2003

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The Essential Fatty Acids-Revisited
23 August 2003

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Psychotheraphy for Rural Malays - Does it Work?
13 September 2003

69. Prof. Dr. Mohd. Zamri Saad
Respiratory Tract Infection: Establishment and Control
27 September 2003

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Cocoa-Wonders for Chocolate Lovers
14 February 2004

71. Prof. Dr. Abdul Halim Shaari
High Temperature Superconductivity: Puzzle & Promises
13 March 2004

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Oils and Fats Analysis - Recent Advances and Future Prospects
27 March 2004

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Microwave Aquametry: A Growing Technology
24 April 2004

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11 May 2004

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The Spider Mite Saga: Quest for Biorational Management Strategies
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The Education of At-Risk Children: The Challenges Ahead
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14 August 2004

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11 March 2005

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Helping Malaysian Youth Move Forward: Unleashing the Prime Enablers
29 April 2005

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Smart Partnership: Plant-Rhizobacteria Associations
17 June 2005

84. Prof. Dr. Mohd Khanif Yusop
From the Soil to the Table
1 July 2005

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Materials Science and Technology: Past, Present and the Future
8 July 2005

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Enhancing Career Development Counselling and the Beauty of Career Games
12 August 2005

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Engineering Agricultural Water Management Towards Precision Framing
26 August 2005

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Bioremediation-A Hope Yet for the Environment?
9 September 2005

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The Wonder of Our Neuromotor System and the Technological Challenges They Pose
23 December 2005

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Rumen Microbes and Some of Their Biotechnological Applications
27 January 2006

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Haemorrhagic Septicaemia in Cattle and Buffaloes: Are We Ready for Freedom?
24 February 2006

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3 March 2006

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Internet Unwired
24 March 2006

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Development and Innovation in the Fracture Management of Animals
31 March 2006

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Strategic Feeding for a Sustainable Ruminant Farming
19 May 2006

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Green Organic Chemistry: Enzyme at Work
14 July 2006

97. Prof. Dr. Malik Hj. Abu Hassan
Towards Large Scale Unconstrained Optimization
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98. Prof. Dr. Khalid Abdul Rahim
Trade and Sustainable Development: Lessons from Malaysia's Experience
22 Jun 2007

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Econometric Modelling for Agricultural Policy Analysis and Forecasting: Between Theory and Reality
13 July 2007

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Managing Change - The Fads and The Realities: A Look at Process Reengineering, Knowledge Management and Blue Ocean Strategy
9 November 2007

101. Prof. Ir. Dr. Mohamed Daud
Expert Systems for Environmental Impacts and Ecotourism Assessments
23 November 2007

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Pathogens and Residues; How Safe is Our Meat?
30 November 2007

103. Prof. Dr. Jayum A. Jawan
Hubungan Sesama Manusia
7 Disember 2007

104. Prof. Dr. Zakariah Abdul Rashid
Planning for Equal Income Distribution in Malaysia: A General Equilibrium Approach
28 December 2007

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Newcastle Disease virus: A Journey from Poultry to Cancer
11 January 2008

106. Prof. Dr. Dzulkefly Kuang Abdullah
Palm Oil: Still the Best Choice
1 February 2008

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Probing the Microscopic Worlds by Lonizing Radiation
22 February 2008

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Waste-to-Wealth Through Biotechnology: For Profit, People and Planet
28 March 2008

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11 April 2008

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The Future of Pesticides Technology in Agriculture: Maximum Target Kill with Minimum Collateral Damage
25 April 2008

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Probiotics: Your Friendly Gut Bacteria
9 May 2008

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Sustainable Supply of Wood and Fibre: Does Malaysia have Enough?
23 May 2008

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Connecting the Bee Dots
20 June 2008

114. Prof. Dr. Maimunah Ismail
Gender & Career: Realities and Challenges
25 July 2008

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Biochemistry of Xenobiotics: Towards a Healthy Lifestyle and Safe Environment
1 August 2008

116. Prof. Dr. Mohd Yunus Abdullah
Penjagaan Kesihatan Primer di Malaysia: Cabaran Prospek dan Implikasi dalam Latihan dan Penyelidikan Perubatan serta Sains Kesihatan di Universiti Putra Malaysia
8 Ogos 2008

117. Prof. Dr. Musa Abu Hassan
Memanfaatkan Teknologi Maklumat & Komunikasi ICT untuk Semua
15 Ogos 2008

118. Prof. Dr. Md. Salleh Hj. Hassan
Role of Media in Development: Strategies, Issues & Challenges
22 August 2008

119. Prof. Dr. Jariah Masud
Gender in Everyday Life
10 October 2008

120 Prof. Dr. Mohd Shahwahid Haji Othman
Mainstreaming Environment: Incorporating Economic Valuation and Market-Based Instruments in Decision Making
24 October 2008

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Big Questions Small Worlds: Following Diverse Vistas
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Responding to Changing Lifestyles: Engineering the Convenience Foods
28 November 2008

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Aesthetics in the Environment an Exploration of Environmental: Perception Through Landscape Preference
9 January 2009

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Leadership Theories, Research & Practices: Farming Future Leadership Thinking
16 January 2009

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Waste Management, What is the Choice: Land Disposal or Biofuel?
23 January 2009

126. Prof. Dr. Jamilah Bakar
Freshwater Fish: The Overlooked Alternative
30 January 2009

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The Chemistry of Nanomaterial and Nanobiomaterial
6 February 2009

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Crop Breeding: Exploiting Genes for Food and Feed
6 March 2009

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Money Demand
27 March 2009

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3 April 2009

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Volunteerism: Expanding the Frontiers of Youth Development
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8 Mei 2009

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Mechanics of Tillage Implements
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