Abstract of Exhibit
Taiwan International Science Fair

CATEGORY: **BioChemistry**

TITLE: **Stimuli-responsive Fullerene Grafted Polymers for Enhanced Drug Delivery Applications**

NAME: **Chai Jishan**

COUNTRY: **Singapore**

Purpose of Research

The physiochemical properties of fullerenes have aroused wide interest, such as its ability to accept and lose electrons and relatively high reactivity that permit various modes of structural modifications. However, obstacles to further research include its complete lack of solubility in water and low processability.

This project investigated the morphology and microstructure of a fullerene-grafted polymer as a potential candidate for better and novel systems for drug delivery. In this research, hydrophilic functionalities were introduced to the C_{60} fullerene by chemical modifications, through the attachment of poly(acrylic acid) (PAA) chain. The objective was to investigate the dynamics and the self-assembly properties of this polymer in aqueous solutions, and the knowledge gained would enhance the development of such system for potential applications in drug delivery and nanotechnology.

Procedures

Atom Transfer Radical Polymerisation Technique (ATRP) is a versatile and controlled technique, extremely suitable for the synthesis of C_{60}-PAA. The following experiments were conducted on the synthesised C_{60}-PAA:

1. Conductometric and potentiometric titrations were conducted to investigate the pH dependence of the polymer in aqueous solution, and to deduce the number-averaged molecular weight, M_n of the PAA grafted onto fullerene.
2. Dynamic Light Scattering (DLS) was conducted to study the dynamics of the polymers or aggregates in solution and to determine the hydrodynamic radius (R_h) of the polymer. The variables consist of varying concentration, angle, addition of 0.5M NaCl electrolyte and degree of neutralisation (α). The R_h can be obtained through the Stokes-Einstein equation $R_h = \frac{kT}{6\pi\eta_o D_o}$.
3. Static Light Scattering (SLS) was conducted to determine the weight-averaged molecular weight, M_w, and the radius of gyration, R_g.
4. Transmission Electron Microscopy (TEM) was conducted to detect the morphology of large C_{60}-PAA aggregates and to observe the aggregates' fractal pattern obtained with the addition of electrolyte.

Data and Discussion

A well-defined C_{60}-PAA polymer was obtained using ATRP. Mono-grafting existed between C_{60} and PAA and the degree of polymerization was found to be 90 (i.e. 90 acrylic acid monomers are attached to each fullerene molecule).

Using conductometric and potentiometric titrations, the degree of polymerization is found be 86 which corresponds with ATRP results. It is found that a pH-responsive C_{60} containing water-soluble polymer was achieved. Counter-ion condensation effect was also observed, whereby due to high charge density on the PAA chains, positive ions are attracted and shield further increase in negative charges. This allows transportation of charged or hydrophilic elements, through their attachment onto the outer PAA shells of the aggregates.

From DLS experiments, the average R_h was found to be 235 nm for all 5 concentrations tested. The consistent shift in the peaks shows that translational diffusion has taken place with varying angles**.** The other observations are:

- R_h is independent of concentrations
- With addition of electrolyte, R_h = 186 nm (due to shielding effect by the Na^+ ions)

From SLS, the true particle size, R_g is 280 nm and the number of polymers per aggregate, N_{agg} is 44500. The ratio of R_g/R_h is 1.2 which shows that the aggregates exist in a spherical formation with a dense core, similar to the theoretical projections based on its amphiphilic properties.

TEM images revealed that the polymers exist in regular spherical micelles, which can be modified to allow the incorporation of hydrophobic drugs or gene segments in biomedical sciences. With the addition of electrolyte, a continuous network of aggregates is formed in a fractal pattern.

Conclusion

Through ATRP, a well-defined water-soluble pH-responsive fullerene system was achieved. Its properties were studied through titrations, light scattering and TEM. With the addition of NaCl electrolyte, a fractal pattern is formed under TEM. This has wide potential in nanotechnology. For example, copper can be incorporated into such polymers in order to form defined structures, and the polymers can then be burnt off to create nano-wires. The specific conformational change of the system as it responds to varying pH, and the counter-ion condensation effect observed, prove useful in the area of biotechnology, especially in the areas of drug delivery and gene therapy, through its ability to transport both hydrophilic and hydrophilic drugs