Condensed Matter Physics

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The Group's main field of research is the development and characterisation of materials with new or improved properties. To this end, radiation is used as a tool to investigate the structure and to induce structural modifications in special samples. Special polymeric materials are currently investigated in collaboration with groups from the Universities of Aveiro and Coimbra, the University of Sophia, Bulgaria, Laboratoire Léon Brillouin (CEA-CNRS-Saclay), KFKI, Budapest, and the Budapest Neutron Centre. During 2005 the main effort was put into the preparation and characterisation of hybrid materials with new properties by radiation induced polymer cross-linking.

The Group is also active in the area of hardware and software instrument development, with emphasis in the design, construction, and testing of systems and components for neutron beam work.

Collaboration with other research groups and a policy of open access for external users to facilities operated by the Group including students are placed high in the ranking of priorities. Two students are presently working for a MSc degree.

The Group's work is supported by funds from FCT, IAEA, ITN and income from services.

Neutron beam facilities. A Two-Axis Neutron Diffractometer (DIDE), and a Small Angle Neutron Scattering Instrument (EPA) are currently under installation in two of the Portuguese RPI research reactor beam tubes. A rotating chopper Time-of-Flight Diffractometer, ETV, is operational. Routine operation of DIDE and EPA is expected to contribute significantly to increase the reactor utilisation and give an additional impetus to the continued operation of the reactor. The TOF Diffractometer is a dedicated instrument for student training. The present RPI reactor flux is adequate for measuring highly dispersive classes of samples and for preliminary measurements preceding data collection at higher flux neutron sources. The reactor also allows in-beam testing of devices such as detectors, collimators and neutron optical components. Upgrading the reactor facility by increasing the power from 1 MW to 5 MW and eventually installing a cold neutron source would open new perspectives for neutron scattering work and the reactor utilisation in general.

Development of components for neutron beam work
Development of the Converging Multichannel Collimator (CMC) based on an original concept was temporarily interrupted in 2005 due to the shortage of manpower especially technicians. The design of a prototype having been completed in 2004, it is intended to proceed in 2006 with the construction and testing of the device.

In-pile components for the Greek research reactor GRR-1, in Athens, were designed, fabricated and already partly installed in December 2005. Fabrication was subcontracted by ITN to ARSOPI, Vale de Cambra, Portugal. The equipment included five aluminium irradiation tubes, two shielding units and two in-pile collimators incorporating beam shutters. The supply of this equipment was contracted with IAEA, Vienna, and the Demokritos Greek Nuclear Research Centre. The in-pile collimators and shutters are to be part of new neutron scattering beam lines.

Design of a new detector assembly for the Two-Axis Neutron Diffractometer DIDE was completed. The assembly with 8 linear position-sensitive ^{3}He counters is projected to replace the old "banana" detector with improved count-rate and equivalent angular resolution. Component acquisition awaits a funding decision.

Research Team

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Preparation of Silica Based Hybrid Materials by γ Irradiation

F.M.A. Margaça, S.R. Gomes, A.N. Falcão, M. Carrapiço, L.M. Ferreira, F.G. Carvalho and I.M.M. Salvado[1]

Objectives

The purpose of the work is to investigate the influence of the various parameters involved in the preparation process of hybrid materials by irradiation using the ITN ^{60}Co source. This new preparation method avoids the use of solvents and catalysts, required by the sol-gel process, yet undesirable in biomaterial processing.

Results

Silica-based hybrid materials have been prepared by γ irradiation of the precursors polydimethilsiloxane, PDMS, tetraethylortosilicate, TEOS, and zirconium propoxide, PrZr. All samples are macroscopically homogeneous and transparent. Their drying was carried out in air, at room temperature.

The dried mass, normalized to the initial mass, M_{dried}, varies with drying time and with the composition. The pure PDMS sample shows no mass variation. The mass of any sample containing alkoxides decreases with drying time. Fig. 2 shows data plotted in a normalized way by taking the ratio:

$$r = \frac{M_{dried} - M_{PDMS}}{M_{ALK}} \quad \text{with} \quad r \in \Re$$

where M_{PDMS} and M_{ALK} are, respectively, the mass of PDMS and alkoxide used in the sample preparation.

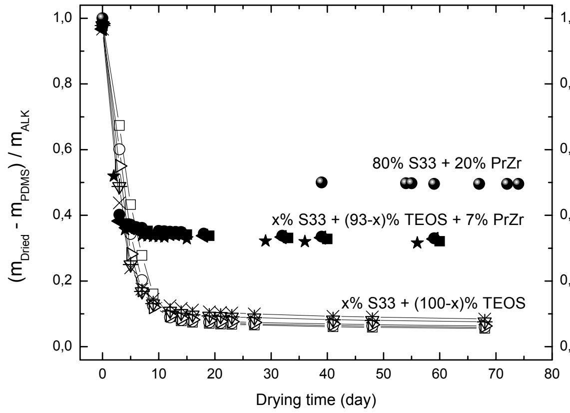

Fig. 2. Normalized dried mass for different content of PrZr and ratios ALK/PDMS = ¼, ½, 1, 2 and 4.

Since the metallic alkoxides are linked to the polymer chains and/or to each other, they lose part of their radicals during the reaction. This process leads to a certain mass loss. For instance, the PrZr molecule $Zr(OC_3H_7)_4$ will loose some carbons and/or hydrogens to link to some other species and in the limit it might become ZrO_2. In terms of molecular weight this means a decrease from 327,57 to 123,22 g/mol. A similar reasoning for TEOS, provides a maximum reduction from the molecular weight variation, when the TEOS molecule $Si(OC_2H_5)_4$ of M_w = 208,3 g/mol becomes SiO_2 for which M_w = 60,09 g/mol. Assuming that, during irradiation and drying, the polymer loses no mass and the alkoxides lose their radicals above, the dried mass, *M*, could be written as:

$$M = M_{PDMS} + \frac{M_w(SiO_2)}{M_w(TEOS)} \times M_{TEOS} + \frac{M_w(ZrO_2)}{M_w(\Pr Zr)} \times M_{\Pr Zr}$$

Fig. 3 shows the measured and the fitted values M.

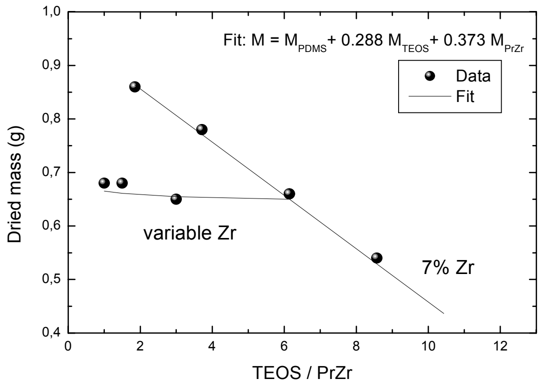

Fig. 3. Data and fit for the dried mass for hybrids prepared with PrZr, TEOS and PDMS.

There is good agreement between the fitting and the experimental data for different alkoxide contents. This evidences the presence of inorganic oxide regions, composed of silica and zirconia. The inorganic oxide mass is ca. 30% of that of the alkoxide. However, when one of the alkoxides is absent there is no formation of an inorganic oxide network, there being only a few individual derived molecules of the other alkoxide linked to the polymer. Both, the dose rate and the polymer molecular weight, were found to have no significant impact in the prepared material. Further investigation, as to the thermal behaviour and the microscopic structure, is in progress.

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New LDPE Copolymeric Films with Enhanced Hydrophilic Properties Prepared by Gamma Irradiation

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Graft copolymerisation induced by gamma radiation, from a ^{60}Co source, has being used for the grafting of 2-hydroxyethyl methacrylate (HEMA) branches onto low-density polyethylene (LDPE) films, in order to improve its hydrophilic properties and adequate the material for bio applications.
Sample preparation protocols were selected from previous kinetic studies in order to obtain films with high grafting yields. The obtained PE-g-HEMA copolymeric films were characterized by thermal analysis techniques (DSC and TGA), and by Fourier transform infrared spectroscopy (FTIR).
The results obtained point out that, upon irradiation, there is some loss of cristalinity of the copolymer backbone, but also that the samples keep a good thermal stability. Evaluation of sample water uptake has shown hydration levels up to 95% with a hydration/dehydration average ratio of 1:5, their use as a bio material thus looking promising.
Further work involves haemolysis and oxygen permeability studies of the prepared films as well as the application of the developed technique to other polyethylene based supports, i.e., flexible linear low-density polyethylene (LDPE) tubes.

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Neutron Spectrometers at the Portuguese Research Reactor

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Design work of a new detector assembly for the 2-Axis Neutron Diffractometer DIDE was completed. The new assembly incorporating a set of linear ^{3}He counters is projected to replace the old “banana” detector with improved count-rate and equivalent angular resolution. Implementation of the project awaits a funding decision.
Assembly of the out-of-pile components of the SANS instrument was completed and followed by an extensive series of tests that included beam alignment, improvement of the shielding setup and the measurement of signal-to-noise ratios under different shielding configurations and instrument operating conditions. The tests have shown the necessity of using a cooled beryllium filter preferably before the shutter or immediately after the mechanical velocity selector. First results on the response of the position sensitive detector count-rate to variations of the anode voltage indicate that a further reduction of noise from neutrons of energies above the cadmium cutoff can be obtained by a convenient choice of the high voltage applied.

Design of beam-line components for installation at the Greek research reactor GRR-1

A.N. Falcão, M. Carrapiço, I.F. Gonçalves

In-pile components were designed for the Greek nuclear research reactor GRR-1 installed at the National Research Center for Scientific Research Demokritos. The equipment included five aluminium irradiation tubes, two shielding units and two in-pile collimators incorporating beam shutters. The irradiation tubes and shielding units were ordered to replace existing equipment, whereas the in-pile collimators and shutters are to be part of new neutron scattering beam lines. Design of the new irradiation tubes and shielding units was preceded by careful measurement of the equipment requiring replacement, performed in a delicate operation under high radiation environment. The tubes were measured in different positions, obtained by rotating them around their axis in steps of 30º, using a long ruler and two strategically placed photographic cameras operated by remote control. The final reference dimensions used in the design were calculated using a fitting routine and the data collected with the cameras. The design of the shielding units and that of the in-pile collimators was aided by the Monte-Carlo MCNP code, using semi-empiric approaches of the neutron and gamma ray energy distributions. Construction of the equipment was carried out at the Portuguese company ARSOPI, Indústrias Metalúrgicas Arlindo S. Pinho, Lda, under ITN supervision. Final installation of three irradiation tubes and of the in-pile collimators and shutters was successfully done in December. Installation of the remaining equipment is scheduled to February 2006.