Symposium 20

Polymers for Biomedical and Pharmaceutical Applications

S20-211

Polymer Stents with Shape Memory as Drug Delivery Systems

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The treatment of vascular obstruction diseases (stenosis) with revasculating methods such as catheter dilatation and re-canalisation does not always result in the desired outcome. After the expansion of the obstructed vessel, there is the chance of an immediate recoil and a re-constriction of the vessel walls. Furthermore, dilatation leads to hardly controllable hyperplastic intimae reactions, especially in the coronary arteries. This may result in a restriction of the flow volume and thereby may lead to the necessity of further therapies or to bypass surgery. Therefore an endoprotheses is being implanted in the vessel after a revasculating treatment in order to avoid these complications, and to ensure a durable enlargement of the flow volume. These minimally invasive implanted vascular endoprothesis ("Stents") are made from hose-like wire nettings or slotted metal tubes. All coronary stents applied so far are made from metallic materials. The utilization of the shape memory effect of polymers allows the minimally invasive application of a fully polymeric stent for the first time [1], which enhances the use of the stent as drug delivery system and thereby improves the post-surgery course through specific application of medication. Active agents can be added to the polymer in rather high doses. A fraction of up to 35% by weight can be realized. This high dosage and along with it a high release of medication in the blood flow might allow for an application of the polymer stent also as a drug delivery system for example in cancer therapy. A loaded polymer stent may be placed in the very vessel that provides blood to the tumour. Thereby a locally restricted, continuous therapy can be realized, and the organism is less burdened with unwanted side effects. [1] H.M. Wache, D.J. Tartakowska, A. Hentrich, and M.H. Wagner, J. Mat. Sci.: Materials in Medicine 14 (2003) 109

S20-989

ENGINEERING OF FOAMED STRUCTURES FOR BIOMEDICAL APPLICATION

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One of the approach in tissue engineering is to design biocompatible and biodegradable scaffold for cells, allowing them to proliferate and maintain their differentiated function in vitro and/or in vivo. The scaffold must possess three-dimensional and highly open porous network and suitable mechanical and biological properties. Scaffolds with controlled porosity, pose size, pore interconnectivity and suitable anisotropic architectures are strongly required for the regeneration of highly complex three-dimensional tissues (such as bone and cartilage) and for the transport of nutrients and metabolic waste to and from the cells. Moreover, the delivery of specific bioactive molecules incorporated into the polymeric matrix is essential to guide cell-material interaction. Scaffold properties depend primarily on the nature of the biomaterial and the pore structure is defined by the fabrication process. In this study, several techniques to prepare porous scaffolds with controlled micro-architectures and specific drug delivery capability are investigated highlighting structure-process-property relationship of the obtained scaffold. In particular, the gas foaming process have been combined with templating techniques with the aim of preparing porous scaffolds characterized by multi-scaled porous network and gradients of porosity and pore size. By the phase separation process, we aim to emboss highly oriented porosity into the scaffold, able to guide the regeneration of specific tissues, such as nerve and blood vessel. Finally, by using a bottom-up approach in scaffold design, we show the possibility to control the spatio-temporal release of specific bioactive molecules from the scaffold. All the technique investigated in this study show great potential in the design of biodegradable scaffold for tissue regeneration and may define some possible future directions for tissue engineering.

S20-1278

In vitro polymer based models of tumors under electric fields : comparison with in silico simulations and in vivo measurements

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In this study, tumor tissues are simulated in vitro with natural polymer gels (collagens) to elucidate electrochemical processes taking place during electrochemical therapy. Use of electric currents in chemotherapy greatly enhances drug transport and delivery synergistically with diffusion mechanism, transport by migration, convection, or electroporation. Cancer electrochemical treatment is based upon the passage of an electric current, whether direct (electrochemical treatment, EChT) or micro -pulsed (electrochemotherapy, ECT), through two or more electrodes inserted locally in the tumor tissue. Extreme pH changes at tissue level (EChT) with significant water motion or the creation of membrane porous channels at the cell level (ECT), facilitating transmembrane penetration of anticancer drugs into the cell, are the main electrochemical effects. Here we explore a combination of ECT and EChT with drug-loaded nanoparticles with the final goal of enhancing drug transport and delivery to tumor tissues and cells. The study is accomplished through in vitro modeling with natural polymer gels (collagen). In vitro measurements are contrasted with in silico simulations (using a mathematical model based on the Nernst-Planck, Poisson and Navier-Stokes equations for ion transport, electric field distribution and fluid flow, respectively) and with in vivo measurements using BALB/c mice bearing a subcutaneous tumor under a dorsal plastic window chamber. Preliminary results suggest that the use of nanoparticle charged drug delivery systems and tuned electric fields significantly increases drug transport and delivery.

S20-506

Shape memory in uncrosslinked biodegradable polymers

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Shape Memory is extremely useful in minimally-invasive deployment of medical devices, particularly in cardiovascular stents. Current stents employ Nitinol, which exhibits shape memory on the basis of a phase transformation. Crosslinked polymers may also be made to exhibit shape memory effects, but are sometimes precluded for use in stents for other reasons. Thermoplastics poly (lactide-co-glycolide) (PLGA) and poly (L-lactic acid) (PLLA) have been used in biodegradable stent prototypes. By appropriate thermo-mechanical processing condition, shape memory effects were observed in both polymers and their shape memory properties were investigated in this paper. The three important parameters in shape memory properties are strain fixity, strain recovery and permanent strain. The effects of deformation temperature, deformation strain level and creeping time on those parameters were evaluated in depth. Current results showed that high deformation temperatures give high fixity, high recovery, but also high permanent deformation for both PLGA and PLLA. Generally, a lower stress applied for a longer duration (the “creeping” method) is preferred to a higher stress applied “instantly” for achieving shape memory. These observed effects could be explained on the basis of molecular orientation and slippage effects. Measurements of birefringence and x-ray diffraction (XRD) were used to derive the orientation function in the whole polymer and in the crystalline region. By making certain assumptions, degree of orientation in the amorphous region was derived as well. The results showed that higher molecular chain orientation is achieved at high deformation temperature and the creeping mode and the development of molecular orientation in the amorphous regions follows the pseudo-affine deformation mechanism. The results also suggested that recovery is associated with considerable disorientation of the amorphous regions, which is the primary mechanism of shape memory.

S20-510

A nanostructured hybrid device based on polymers infiltrated porous silicon layers for biotechnological applications

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Hydrophobic aliphatic polyesters, such as poly(ε-caprolattone) (PCL), polylactide (PLA) and their copolymers, have been intensively investigated in applications such as medical devices used in the regeneration of damaged tissues and also in the pharmaceutical field for the drugs controlled release systems. Porous silicon (PSi) is really an amazing platform material due to its physical and structural features: the PSi sponge-like morphology shows a high surface area (up to 500 m2/cm3) which strongly interacts with the surrounding environment. As a consequence, some physical parameters, such as the dielectric constant, the conductivity and the photoluminescence of PSi samples strongly change. Thus the PSi is quite an ideal transducer for the monitoring of biochemical species. Due to the sensing mechanism, these kinds of devices are not able to identify the components of a complex mixture. In order to enhance the sensor selectivity through specific interactions, some chemical or physical strategies are employed to modify the PSi surface. Moreover the hydrogenated PSi surface shows high thermodynamical instability which constitutes a severe drawback in most applications. A very promising approach to overcome these difficulties is in fabricate hybrid systems based on the infiltration of polymers in the nanometric pores of the PSi matrix. As an alternative to the conventional chemical surface modification strategies, we have infiltrated biocompatible multi-block copolymers based on PCL and poly(oxyethylene), containing pendant functional groups regularly spaced along the chain, which can be used as passivation coatings of PSi. The reactive groups are available for the attachment of chemical substances and their distribution, as well as the hydrophilic/hydrophobic ratio, can be modulated according to the molecular weight of both segments. In this way, we can change and modulate the physical and chemical properties of the porous silicon surface.

S20-526

The use of Diffusion Induced Phase Separation (DIPS) technique for the preparation of biodegradabile scaffolds for angiogenesis

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A critical obstacle in tissue engineering is the inability to maintain large masses of living cells upon transfer from the in vitro culture conditions into the host in vivo. Capillaries, and the vascular system, are required to supply essential nutrients, including oxygen, remove waste products and provide a biochemical communication “highway”. For this reason it is mandatory to manufacture an implantable structure where the process of vessel formation -the angiogenesis – can take place. In this work PLLA scaffolds for vascular tissue engineering were produced by means of a Diffusion Induced Phase Separation (DIPS) technique. The scaffolds, with a vessel-like shape, were obtained performing a DIPS process around a nylon fibre whose diameter was 700 µm. The fibre was first immersed into a 4% PLLA dioxane solution and subsequently immersed into a second bath containing distilled water. The covered fibre was then rinsed in order to remove the excess of dioxane and dried; finally the internal nylon fibre was pulled out so as to obtain a hollow biodegradable PLLA fiber. A SEM analysis revealed that the scaffolds attained have a lumen of ca. 700 µm. The internal surface is homogeneous and with micropores 1-2 µm large. Moreover, a cross section analysis as shown an open structure across the thickness of the scaffold walls. A cell culture of endothelial cells was performed into the scaffolds. The result showed that cells are able to grow within the scaffolds and after 2 weeks they begin to form a “primordial” vessel-like structure.

S20-721

pH-sensitive Nanoparticles Formed by Interchain Hydrogen Bonding

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Polymer - polymer complexes are formed by association of various macromolecules. Complex formation and its stability are often sensitive to the environment which makes the complexes good candidates for applications in biotechnology and in biomedicine. Of interest here are the complexes formed via hydrogen bonds between diblock copolymers of model hydrogen donors such as polycarboxylates and hydrogen acceptors such as poly(ethylene oxide) and complexes formed by hydrogen bonding between partly ionized polycarboxylates. Therefore, the aqueous solution properties of poly(methacrylic acid-block-ethylene oxide) (PMAA-b-PEO) copolymers and partly ionized polycarboxylates were investigated by static and dynamic light scattering methods. Well defined pH-sensitive micelles formed by interchain hydrogen bonding of PMAA-b-PEO copolymers were prepared and investigated at pH < 5. Both the molecular weight Mw and hydrodynamic radius Rh of micelles increase with decreasing pH of solution, displaying an asymptotical tendency at low pH values. The observed micelles (10 – 40 nm) are well defined nanoparticles with narrow size distributions (polydispersity Delta Rh/Rh < 0.05, Delta Rh is a half-width) comparable with regular diblock copolymer micelles. The critical micellar concentrations are slightly below c = 0.1 mg/mL. The micelles are negatively charged and their time-stability is lower than that of regular copolymer micelles based purely on hydrophobic interactions. The structure of the micelles was proposed.The investigation of stable nanoparticles formed by complexation of partly ionized polycarboxylates (e.g., poly(2-ethyl acrylic acid) is in progress. The irreversible formation of stable well defined nanoparticles was observed on heating of aqueous solution of the polyelectrolytes. The results will be included in the contribution.

S20-811

Dynamic Creep and Fatigue Performance of Thermoplastic Elastomers as Biomaterials

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The use of polymers has becoming more relevant in medical applications for the variety in processing and enhanced material properties of polymers.To be biomaterials, it is of medical importance that polymers must be biocompatible and do not elicit negative responses from the body after implantation.Furthermore during the course of usage,these biomaterials must sustain dynamical loadings due to our body movement so that they do not degrade and fracture to cause inflammation.This work looks into the dynamic creep and fatigue performance of a new class of thermoplastic elastomers-a poly (aliphatic/aromatic-ester)(PED) multiblock copolymer that has a physical network with semi-crystalline hard segments.The biocompatibility of PED has already been established under in vitro and in vivo conditions in earlier works.To enhance the material stiffness, the PED studied herein will be subjected to various dosages of e-beam curing to introduce covalent bonds between polymer chains and hence form a cross-linked network structure.These pristine and e-beam cured PEDs will be evaluated using the classical quasi-static tensile and creep testing, as well as, the dynamic hysteresis measurement.To relate the mechanical behaviour to the network structure and degree of crystallinity of PEDs, research effort is made to study the thermal and thermo-mechanical responses of these materials using differential scanning calorimetry and dynamic mechanical analysis, respectively.With the aid of light and electron microscopy, an investigation of the surface morphology will be undertaken to examine the change of fracture characteristics of PEDs for different cross-link densities after testing.To benchmark the dynamic creep and long-term performance of the PEDs, commonly known biomaterials, like silicone rubbers and thermoplastic polyurethanes, shall be adopted as references to elucidate the influence and effectiveness of material network structure on these soft materials.

S20-841

Novel bottom up approach to the design of multifunctional scaffolds

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Advanced tissue engineering approaches rely upon the employment of biomaterials that integrate biodegradable scaffolds with growth factor delivery platforms to guide cellular events and enhance tissue neogenesis [1]. Therefore, novel strategies to biomaterials design that can contemporary provide adequate microstructural properties and controlled sequestration of bioactive molecules in a spatial-temporal controlled manner are required. Along this line, here we proposed a novel bottom up approach for the realization of bioactive scaffolds with controllable pore size and interconnection, combined with protein-loaded polymeric microcarriers acting as local chrono-programmed point source generation of bioactive signals. Bioactive scaffolds are obtained through two different methods, the thermal and the chemical assembly of protein activated poly ε-caprolactone (PCL) microspheres, prepared by double emulsion, and larger protein free PCL microspheres, obtained by single emulsion. It is shown that the pore dimension, interconnectivity and mechanical compression properties of the scaffold could be predefined by an appropriate choice of the size of the protein-free microparticles and process conditions. Protein-loaded microparticles were successfully included within the scaffold and provided a sustained delivery of a model protein (BSA). These novel matrices offer the unique possibility of concurrently modulate and control the size and extension of the porosity, mechanical properties and the spatial-temporal distribution of multiple bioactive signals. Furthermore, a comparison between the two different sintering methods will be provided.[1] Ikada Y. Challenges in tissue engineering. J R Soc Interface 2006;3:589–601.

S20-843

A predictive model for time-dependent protein concentration profiles within single biodegradable PLGA microspheres

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Protein release from biodegradable poly(lactide-co-glycolide) (PLGA) microspheres (MS) in an aqueous milieu occurs by a complex diffusion-degradation mechanism initiated by water invasion in MS which triggers protein solubilization/release and polymer autocatalytic degradation. The determination of the interplay between protein transport/delivery and PLGA degradation/erosion is necessary to engineer release kinetics. Thus, the aim of this work was the development of a mathematical model describing protein radial profiles as a function of time to obtain a relation between local transport parameters and protein release kinetics within degrading MS. PLGA MS encapsulating a fluorescent model protein, namely rhodamine-labelled bovine serum albumin (BSA-Rhod), were prepared and the time evolution of BSA-Rhod radial distributions within MS during release in collagen determined by Confocal Laser Scanning Microscopy (CLSM). BSA-Rhod concentration was divided into two populations: an immobile fraction, enclosed in unopened macropores, and a mobile fraction, diffusing through MS after exiting the eroding macropores. BSA-Rhod profiles showed that protein concentration mainly decreased in MS center, while remaining essentially unchanged in the periphery. Results suggest that the release mechanism is governed by macropore opening, which is favoured in MS center, and the subsequent quick diffusion of free BSA-Rhod through the highly hydrated microchannel network of MS, and show the existence of a degradation front moving from outwards from MS centre. Taken all together, the data suggest different degradation patterns along MS radius. The model helps elucidating the complex interplay between transport and degradation patterns governing protein release from biodegradable MS as it can describe transport phenomena at the single MS level, and represents a valuable tool to design ab-initio release profile from polymeric MS.

S20-978

Reduction of bacterial adhesion onto biocompatible polymers via plasma processing

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Polymers are vastly used in biomedical devices due to their unique mechanical properties. However, the widespread application of such class of materials is sometimes restricted by poor biocompatibility. Several studies have shown that the performance of an artificial material when in contact with a biological environment is mainly determined by the properties of its surface. Owing to that, a great effort has been devoted to the development of materials with surfaces more appropriated to a certain application. In this sense, plasma processing is particularly interesting because it enables to modify the characteristics of a surface without affecting bulk properties. In addition, the results are strongly influenced by the conditions of the treatment. Therefore, adjusting the plasma parameters it is possible to tailor the surface properties to best fulfill the requirements of a given application. In this work, polyurethane and silicone coupons have been subjected to glow discharge plasmas. It has been investigated the influence of different plasma conditions on the adhesion of Escherichia coli and Pseudomonas aeruginosa onto the polymers. The wettability and free surface energy have been characterized by contact angle measurements while the surface roughness has been evaluated through profilometry. Attenuated total reflection Fourier transform infrared spectroscopy (ATR-FTIR) has been applied to explore chemical modifications on the surface after plasma exposures. Bacterial adhesion has been quantified through in vitro experiments by the determination of the number of colony forming units on Petri dishes with agar nutrient. It has been observed that the surface roughness played an important role on the microbe adhesion. Furthermore, the results have shown that the amount of adhered bacteria is dependent on the surface contact angle. The more hydrophobic the surface, the smaller amount of microorganisms attached to it.

S20-995

DESIGN OF BIODEGRADABLE SCAFFOLDS VIA GAS FOAMING AND MICROPARTICULATE TEMPLATING

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The regeneration of biological substitutes for damaged tissues and organs is one of the most important challenges in tissue engineering. For the regeneration of new tissues in vitro and/or in vivo, one of the strategies is to guide the distribution and organization of cells in 3D by using biocompatible and biodegradable scaffolds. The scaffold must be characterized by a well definite porous network and highly interconnected porosity, that need to be optimized with the opportune selection of the processing techniques. From this point of view, CO2 foaming has been attracting great interest since this process can be applied to a large class of biodegradable polymers and avoid the use of organic solvents that may damage transplanted cells and inactivate the biological signals incorporated into the polymeric matrix. Although the gas foaming allows a fine control over the extension of porous networks of the scaffold, the achievement of high degree of interconnectivity is often impaired by a combination of rheological and processing limitations. To overcome these limitations, in this study we combined gas foaming with NaCl microparticles templating. The modulation of the viscoelastic properties of the microparticulate composites and the fine control of the gas foaming parameters allowed the preparation of highly interconnected poly(ε-caprolactone) scaffolds in a wide range of processing conditions and characterized by different porosity, pore size and degree of interconnection. In addiction, by the control of the microparticles spatial distribution, this technique provided the possibility to create and tailor gradients of porosity and pore size within the porous architecture of the scaffolds suitable to reproduce the spatial organization of cells and extracellular matrix into the native tissue.

S20-1013

Electrospun Cellulose Acetate Fiber Mats Containing an Herbal-Zingiber officinale -Extract and Release Characteristic of Herbal Subtance

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Electrospinning is a straightforward method of producing fibrous polymer mats with fiber diameters in the range of ca. 0.05 microns to several tens of µm . Such materials may be useful for many applications in medicine such as wound dressings and scaffolds for tissue engineering. Cellulose acetate (CA) is the acetate ester of cellulose. CA has been fabricated as semi-permeable membranes for separation processes and fibers and films for biomedical applications. Ginger rhizome (Zingiber officinale) are extracted with dichloromethane-methanol (1:1 v/v), which contained a mixture of active compounds, 6-, 8- or 10-gingerol and 6-shogaol. Extracts have been reported to have effect as an anti-inflammatory , antioxidant , antithrombotic and anticancer agent are selected as model drugs. In this work, the use of mats of electrospun cellulose acetate nanofibers as carriers for delivery of ginger extract is evaluated. The morphology of ginger extract loaded electrospun cellulose acetate fiber mats are determined by Scanning electron microscope (SEM). Degree of swelling and weight loss of both the neat and the ginger extract-loaded electrospun CA fiber mats as well as those of the neat and the drug-loaded as-cast CA films are measured after the samples are submerged in an acetate buffer solution at 32 °C for 24 h. Moreover, the release characteristics of gingerol and its derivatives from the ginger extract-loaded electrospun CA fiber mats and the corresponding as-cast films are investigated by total immersion method as well as transdermal diffusion through a pig skin method.

S20-1210

On the release of active molecules from hydrogels based tablets

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Controlled drug delivery is a key topic in pharmacology and many researchers over the entire world are currently involved in related studies. The most frequent route of drug administration is the oral one, i.e. the assumption of drug through the mouth. To assume a drug through the mouth is simply and very effective, due to the large exchange available area for the drug assimilation in the gastro-intestinal tract (GI). To avoid the need for administration repetitions, and to maximize the time interval between two administrations, the best would be a system able to provide a constant flux of drug in the dissolving physiological compartment, i.e. to realize a well-defined profile of drug release. The systems based on drug dispersed into hydrogel swellable matrices seem to be able to fulfill this requirement. The use of the HydroxyPropyl MethylCellulose (HPMC), a bio-compatible hydrogel, was explored in this study, to clarify the transport phenomena which take place after the tablet swallowing: the matrix hydration with subsequent polymer plasticization, the increase in water and drug diffusivities, the drug diffusion toward the dissolution medium, the tablet erosion. We designed and carried out several experiments to analyze these phenomena: 1) the hydration of pure HPMC tablets, allowed 1.a) only along the radial direction by confining the tablets between two glass slabs ("radial" tests) and 1.b) by the entire tablet surface area ("overall" tests); 2) the hydration and the drug release from tablets loaded with Diclofenac Sodium or with Theopylline, both by 2.a) the radial tests and by 2.b) the overall tests. Observed phenomena were also described by proper models. The general framework of the research is presented here, and a critical evaluation of the full set of experimental and modeling results is carried out.

S20-1344

Material property changes vs. release of phthalates from in vitro and in vivo aged biomaterials made of PVC

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Materials used in biomedical devices are exposed to the body environment and in many cases a biofilm is formed on the surface. There is continuing debate in the literature regarding these materials and their long term stability. Over the years, several studies on polymeric biomaterial have described the effect of migration and loss of additives. After the loss of additives the polymers may demonstrate increased embrittlement and an overall reduction of the mechanical stability. At a molecular level, changes in the chemical structure may also be monitored. In Sweden there are more than two hundred people living with permanent tracheostomy. Since 1998 the Division of Anesthesia- and Intensive Care, Danderyd University Hospital is the main centre in Sweden for tracheostomy patients with complicated case histories. At Danderyd University Hospital standard tubes of material such as polyvinylchloride (PVC), polyurethane (PUR), silicone and sterling silver are modified. These custom designed tracheal tubes are made to give each patient optimal function and comfort, in order to breathe and speak effortlessly. Most of the manufactures recommended frequent changing of the tubes, but they do not give a life span of the tubes. In Sweden the time limit for a tracheal tube implant is 30 days after which the tubes are taken out and inspected. If nothing appears to be wrong, the tubes are again inserted into the trachea. The trachea constitutes a very complex and harsh environment, which is very demanding for the material in terms of durability and bicompatibility. The presentation addresses part of an ongoing study about the long-term properties of biomaterials used in tracheal implants1. The changes in polymeric properties is presented and discussed with relation to the release of phthalates from PVC tracheal tubes. 1.G. Björling, S. Axelsson, U-B. Johansson, M. Lysdahl, A. Markström, U. Schedin, R. E. Aune, C. Frostell and S. Karlsson: Clinical use and material wear of

S20-174

Surface modification of cellulose by using chitosan

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The research consists of synthesis of chitosan from various sources and modify the same using quaternary ammonium salts for improving antimicrobial activity in cellulosic polymers. Chitosan is a naturally occurring, bio-degradable, non-toxic, non-allergenic bio¬polysaccharide derived from chitin, found in abundance in nature. The main focuses of investigations is aimed at excellent antimicrobial activity demonstrated by chitosan. In this context, certain quaternary ammonium derivatives of chitosan have been shown in the past to be highly effective antibacterial materials. In the present study, substituted chitosan derivatives were synthesized and further quaternized to produce water soluble derivatives at neutral pH. The antibacterial assessments of the quaternized derivatives of substituted chitosan were performed on liquid cultures of E. coli and S. aureus and results expressed in terms of Minimum Inhibitory Concentration (MIC). Three synthetic routes, viz. Bosch reduction, γ-lactone addition and an anhydride addition have to be used to obtain hydrophobic as well as strongly and weakly acidic organic groups. In the case of Bosch reduction aromatic aldehydes are used to obtain targeted substitutions.Also n-Octyl aldehyde in the presence of dimethyl formamide and γ-Octanoic lactone addition resulted in very low levels of substitution. The derivatives obtained using this method provided excellent antibacterial activity (MIC=16µg/mL, compared to 128 µg/mL for the unsubstituted quaternized chitosan). γ-Lactone addition and anhydride addition are simple methods of synthesizing chitosan derivatives with potential applications in industry. Chitosan derived gels have been proposed using an aromatic, rigid (terephthaldicarboxaldehyde) and an aliphatic, flexible (glutaraldehyde)cross-linkers and their quaternized derivatives with low levels of cross-linking. Thus the research aims for modification of textiles using chitosan for antimicrobial properties.

S20-180

BIOACTIVE BIODEGRADABLE POLYMERS PROCESSED VIA TWIN SCREW EXTRUSION

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Bioactive biodegradable copolymers have been the subject of extensive research in the past decade. Here the affects of bioactives and copolymers on the primary matrix are studied.Various blends of the polymers; Poly ε-caprolactone [PCL], Poly l-lactic acid [PLLA], Polyethylene Glycol [PEG] were shear blended with Nalidixic Acid [NA] and melt pressed into plaques, to determine changes in their drug release, morphology, topography and thermal and mechanical properties. Release of NA was seen to increase with the addition of copolymers in the PCL matrix with maximum release at 55% in a blend containing 5%w/w each of PLLA, PEG and NA. Rheograms showed higher viscosity of PCL blends with increasing content of PLLA and NA, due to the filler effect. A lower viscosity occurred with the addition of PEG due to its Newtonian behaviour. FTIR spectrums showed the major chemical bonds within the blends appeared unaffected by addition of copolymers but that a polymerisation reaction occurs between the PCL and the NA. The C=0 allowed for some degree of miscibility between the PCL and PLLA, which was also noted by the decrease in thermal conductivity from 0.26 to 0.15 Wm-1°C. Mechanical properties were greatly reduced by the copolymers, with the Young's Modulus decreasing by 15% and the Elongation at Break from 780 to 10%. This reduction in mechanical strength is due in majority to the addition of the brittle PLLA.

S20-185

Study on synthesis of nanomaterial based on nanosilver and shape memory polyurethanes using in biomedical applications

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The structure-property relationship in elastomeric polyurethanes based on 4,4-diphenylmethane diisocyanate (MDI) ,1,4-butanediol (BD) and nanosilver was studied. An ether type polyol was used to synthesize of polyurethanes and the effect of appropriate programming on their microstructures and shape memory has been studied. Differential scanning calorimetry (DSC), dynamic mechanical analysis (DMA) and Fourier transform infrared spectroscopy (FTIR) and invitro tests have been used to evaluate performance of these kind of polyurethanes as an biomedical device.The degree of phase separation , final bulk mechanical properties and bio function is dependent to processing conditions , materials and also thermal history that they have experienced.

S20-258

Nanomechanical Properties of UHMWPE irradiated with N

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Inadequate mechanical surface properties limit the service life of ultra-high molecular weight polyethylene (UHMWPE) hip and knee joint components. It is well known that in vivo wear resistance of UHMWPE can be improved by crosslinking but concomitantly its fatigue strength is reduced. In an own previous work, it was shown that irradiation with 33MeV N ion beam at fluences ranging from 5x1010 to 1x1014 ions/cm2 induces a highly crosslinked structure only in the outer layers of the material. Thus it appears that by means of this irradiation technique it may be possible to generate suitable surface modifications without affecting the desirable bulk properties of UHMWPE. The motivation of this work was to examine nanoscale mechanical properties of the outer layer region of N2+ beam irradiated UHMWPE using depth-sensing indentation. Reduced elastic modulus and universal hardness were determined applying trapezoidal loading functions with a Berkovich tip by means of the Oliver-Pharr approach. The apparent friction coefficient was evaluated from nanoscratch tests carried out with a 20 µm conospherical tip under constant load. The irradiated samples exhibited better performance than unirradiated ones, ie. higher elastic modulus and hardness, and lower friction coefficient, suggesting a better wear resistance. Fortunately, changes in nanomechanical properties were solely confined to the surface layer. Indeed, these properties showed up to be dependent on fluence under the irradiation conditions used. UHMWPE irradiated with 1x1012 ions/cm2 exhibited a four-fold increase of hardness, a three-fold increase of elastic modulus and an one point five-fold decrease of friction coefficient.

S20-317

New photopolymerizable dimethacrylates with potential for dental applications

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Starting from the improved properties of some urethane acrylate– based composites [1], intensively investigated as esthetic restorative materials besides acrylic ones, and the fact that, acid- functionalized urethane dimethacrylates have been not reported, our group initiated a research on this subject [2] in order to follow the effects of chemical structure of some oligomers on the photocuring process in some dental formulations. As reference co- monomer was used bis GMA or a new derivative of bisGMA with aim to reduce the polymerization shrinkage. Therefore, a series of light- cured hybrid composites containing a higher loading of different fillers embedded into an organic matrix of these monomers functionalized with carboxylic/phosphate groups to confer them a good adhesion [3], were prepared and characterized. A study concerning photobehavior to polymerization initiated of a dual system (amine/ CQ/BP) is accompanied of data obtained via fluorescence technique that allows in situ monitoring the photopolymerization of multifunctional dimethacrylates to form a polymeric network. For this, a molecular probe (piren, anthracen)[4] was added in each dental formulation following the response of the probe's emission to changes in its environment during photopolymerization. A comparison between these data reveals that depending on the nature of fluorophore, a fluorescence quenching effect can be observed in our systems. Other properties as adhesiveness, polymerization shrinkage, mechanical characteristics, and water behavior were also evaluated in order to select the best compositions. Acknowledgements The authors thank to the board of MATNANTECH program for the financial support (Projects Nr. 30/2005; 504/2006) [1]. Kwon Y.H. et al. J. Prosthet. Dent. 88, 396 (2002) [2]. Buruiana E.C. et al. J. Polym. Sci. Part A: Polym. Chem. 45, 1956 (2007) [3] .Wan Q. et al. Biomacromolecules, 2, 217 (2001) [4]. Buruiana

S20-383

BIODEGRADABLE NYLONS. HYDROLITIC DEGRADATION OF AABB-POLYAMIDES DERIVED FROM L-ARABINOSE AND D-XYLOSE

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Commercial nylons have good mechanical properties but they present low susceptibility to biodegradation, even to hydrolytic degradation. In order to improve the degradability of nylons as nylon 6 and nylon 66, some chemical modifications have been made to obtain more hydrophilic analogous. However, these modified nylons were not degradable in a noticeable extension.We now describe the hydrolytic degradation studies of some carbohydrate-based polyamides of the AABB type, which are nylon-n,n analogs. This type of polyamides are obtained by the polycondensation reaction of diamines with active esters of dicarboxylic acids. Starting from the naturally occurring and easily available sugars L-arabinose and D-xylose, we had prepared the appropriate monomers for the polycondensation reactions.The corresponding polyamides, in which both monomers were sugar-based, are designated as PA-SuSu; and PA-mSu and PA-Sun if the sugar-based monomers are only the dicarboxylic acid or the diamine units, respectively. The L-arabino and xylo moieties are designated as Ar and Xy. The thermal properties and crystal structures of these carbohydrate-based polyamides depended on their constitution and on the configuration of the carbohydrate-based moiety.In the present communication, we conclude that aregic polyamides of the AABB-type based on L-arabinose and D-xylose, underwent degradation in aqueous media by a simple hydrolytic mechanism of the amide function. In general, those polyamides being amorphous and highly hygroscopic materials, such as PA-SuSu and PA-Sun, were the most easily degraded, even in water, at neutral pH, at temperatures near to their glass transitions. Furthermore, the less crystalline xylitol-based polyamides were more sensitive to the hydrolysis than the more crystalline L-arabinitol-based ones.

S20-384

PBT ANALOGS POLYESTERS DERIVED FROM GALACTARIC AND L-ARABINARIC ACIDS

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Poly(butylene terephthalate), PBT, is a semicrystalline thermoplastic, which is mainly produced by polycondensation of dimethyl terephthalate with 1,4-butanediol. PBT has a moderately melting temperature of around 225 °C and crystallizes very fast from the melt. This exceptional thermal behavior together with its excellent mechanical properties made PBT the material of choice for durable goods that are manufactured by injection molding. This polyester, together with PET, is the genuine representative of the aromatic polyester family. It is a well stable material, scarcely affected by environmental conditions. Since it absorb very little water (less than 0.1%), it is highly resistant to degradation in aqueous media. It is also very reluctant to be degraded by microorganisms. Nowadays, great efforts are being done in order to render aromatic polyesters more hydrophilic and susceptible to biodegradation with the aim of making it usable in temporary applications, mainly in the biomedical field. It is known that all polyesters degrade eventually, hydrolysis being the dominant mechanism. However, degradation rates may range from weeks for certain aliphatic polyesters to decades for PET and PBT. Copolymers including aliphatic and aromatic counterparts are currently under investigation to develop biodegradable materials able to satisfy new demands. We have recently described the synthesis, characterization and some properties of homopolyesters and copolyesters analogous to PBT and PET based on L-arabinitol and xylitol.In this communication we present the synthesis of new homo- and co-polyesters analogous to PBT, where 2,3,4,5-tetra-O-methyl-galactarate or 2,3,4-tri-O-methyl-L-arabinarate units substituted the terephthalate units. The obtained polymers show medium values of Mw, in the range 10,000 to 34,000. We have also studied the microstructure of these polyesters and some properties as their hydrofilicity, thermal properties and crystal structure.

S20-389

CHITIN-CONTAINING MATERIALS FROM NATURAL FUNGI

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A new simplified technology for isolation of chitin-glucan and chitin-glucan-melanin complexes from biomass of native higher fungi has been developed. The complexes of biopolymers obtained from fungi belonging to classes Basidiomycetes and Ascomycetes were studied in terms of processing characteristics, sorption properties, and structural parameters by means of thermogravimetry, pyrolysis gas chromatography, FTIR spectroscopy and X-ray diffraction in comparison with the crabshell chitin. The chemical and structural changes occurring in the successive stages of the treatment were examined. It was shown that new method allows preserving native properties of the material in the process of chitin isolation. The application of the domestic detergent enabled one to substantially decrease the number of treatment with acids and alkalis and to markedly reduce their concentrations. The Arthrodpoa chitin was found to be both deacetylated and deaminated in the course of isolation. The received results indicate that native higher fungi can be used as a potential source of raw materials for chitin and D-glucosamine production. The availability of chitin-containing materials with desired selective properties by control of component ratio in heterogeneous structure (chitin-glucan-melanin) selection of fungi kind and conditions of processing was shown. Comparisons of sorption properties of biopolymers in relation to water and ions of heavy metals (Ni2+ and Cd2+) with properties of crab chitin and cotton cellulose have been elicited. Influence of the nature of metal, type of fungi, and particle size of adsorbent, conditions of processing on sorption properties was discussed. It was shown that sorption characteristics of the received biopolymers in relation to water and Ni2+ and Cd2+ exceed that of cotton cellulose and crab chitin.

S20-413

Preparation of High-Strength and High-Modulus HA/UHMWPE nanocomposites for Cortical Bone Replacement

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Cortical bone accounts for 80% of the total bone mass of an adult skeleton and it plays a load-bearing role in human body, with over 150 MPa in tensile strength, more than 17 GPa in tensile modulus and around 2.0% in tensile strain. Metal alloys are currently used for cortical bone replacement due to their good mechanical properties. But the mismatch between bone and metal alloy can cause “stress shielding” of the bone. Besides, those meal alloys are also too heavy. Since Bonfield introduced hydroxyapatite (HA) reinforced high density polyethylene (HDPE), there have been numerous works done to develop polymer composites with bioactivity. But these composites are just suitable for soft bone substitution applications due to their relatively low strength and stiffness.In this study, hydroxyapatite (HA) reinforced ultra high molecular weight polyethylene (UHMWPE) nanocomposites were fabricated by twin-screw extrusion using the masterbatch method. HA aggregates were broken down into nano size particles by applying an effective grinding technique followed by ultrasonication. Paraffin oil was used as a swelling solvent during the extrusion. The electron micrographs results indicated a homogeneous dispersion of nano-hydroxyapatite (n-HA) in the polymer matrix. The composite with n-HA weight percentage of 10% (volume percentage 3.2%) exhibited good bioactivity in vitro after hot drawing. After immersing in SBF for 7days, the sample formed a thick Ca-P layer on the surface. The hot drawn composite showed 534.8 15.8 MPa in strength and 19.7 2.2 GPa in Young’s modulus, and a strain at break of 6.8%, that are comparable to human cortical bones.

S20-423

Blending PLLA with PLA so as to tune the biodegradability of polymeric scaffolds for soft tissue engineering applications

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Foams for tissue engineering were prepared via thermally induced phase separation (TIPS). Poly-L-Lactic Acid (PLLA) and blends of PLLA with PLA in different proportions were used (100/0, 90/10, 75/25, 50/50, 0/100 PLLA/PLA wt/wt) starting from ternary systems where dioxane was the solvent and water the non-solvent. Morphology was evaluated by Scanning Electron Microscopy (average pore size and interconnection) and the void fraction was measured by means of Hg porosimetry. The foams' apparent density was evaluated (porosity ranges from 87% to 92%). Biodegradability was estimated in a body mimicking fluid. Results show that structure and morphology (in terms of average pore size and distribution, interconnectivity and mechanical properties) depend upon the combination of the operating conditions adopted for the process (polymer concentration, solvent/non-solvent ration, demixing temperature and time). Furthermore, by blending PLLA with PLA it is possible to tune the biodegrability of the foam, thus addressing the possible applications in different fields of soft tissue engineering.

S20-432

Polyalkenoates modified with photopolymerizable functions useful in formulating dental materials

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As is well-known, the glass polyalkenoates or glass-ionomer cements (GICs) continue to be widely used in restorative dentistry and not only [1]. Such materials currently used in GIC are water-based materials, that involve an inorganic glass powder of varying particle size/shape/nature, a polyacid, usually polyacrylic acid/copolymers, water, and tartaric acid, capable to modify the cement-forming reaction for improving the properties [2]. The first commercial GIC consisted of a high-fluoride glass powder and a copolymer of acrylic and itaconic acid that reacted in the presence of water to couple the organic and inorganic phase together. To improve the mechanical properties of such biomaterials, like impact resistance, flex strength, fracture toughness and resistance to water, a preparative variant proposed the incorporation of polymerizable groups as part of the liquid component, as HEMA or Bis-GMA [3].The purpose of our study was to obtain classically and under microwave-accelerated synthesis two acrylic copolymers, namely, poly (acrylic acid-co-N-vinylpyrrolidone) and poly (acrylic acid-co-itaconic acid-co- N-vinylpyrrolidone) to be further modified. Thus, based on the reaction of the acid copolymer with HEMA or urethane methacrylate monomer, new acid copolymers have been prepared and characterized. The properties of these copolymers, formulated with glass powders were also examined by spectral, thermal methods and mechanical ones. Previously, our group initiated researchers concerning the photopolymerizable acid urethane oligomers for dental formulations [4].AcknowledgementsThe authors thank the Ministry of Research and Education and the board of RELANSIN program for the financial support (Project no. 132/2006) References1. B. Culbertson J. Dent. 2006, 34, 5562. M. Towler et al. Bio-Med. Mat. Eng. 2004, 14, 565.3. B. Culbertson et al. Polym.Prepr. 1997, 38, 127.4. E.Buruiana et al.J. Polym. Sci. Part A: Polym. Chem. 2007, 45, 1956.

S20-458

New polymers with potential pharmaceutical application

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As is known macromolecules with hem.-dichloromethylene fragments have biological activity. For the synthesis of polymers containing such type of sections we involve vinyl-hem.-dichlorocyclopropanes to the free radical polymerization and copolymerization.Was founded that in the structure of homo- and copolymers hem.-dichlorocyclopropane fragment is missing. The base structure of section was cis-/trans- 1,1-dichloro-pentene-3. In addition the chain was prolongated by 1,1- dichloromethyl radicals.In the report is discuss the mechanism of homo- and copolymerization, activity of vinyl-hem.-dichlorocyclopropanes in the reactions with monomer of different structures.

S20-464

Chitin and Polyurethane Blends: Specific Interactions, Thermal Properties and Crystallization Behavior

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Chitin is a very abundant natural polymer with well known healing properties, but its application has been hindered by its stiffness. Blends of chitin and a biodegradable, polycaprolactone based polyurethane were explored, aiming to prepare a bio-compatible and degradable material for drug release membranes. The morphology, mechanical and thermal properties were studied as the first step in the project. Cast films of the blends with systematic variation in the components ratio, varying from 10% to 70% polyurethane/chitin (w/w) were analyzed by TGA, DSC, DMTA, SEM and Xray diffraction. An increase in the glass transition of the polyurethane was observed, indicating the occurrence of a specific interaction between the two components, probably through hydrogen bonds. Up to 40% (polyurethane/ chitin) the continuous phase is the chitin, above that the matrix is polyurethane. When chitin is the matrix, the polyurethane domains retain a certain degree of crystallinity, but in the reverse situation no crystallinity of chitin was detectable. The upper limit of 70% polyurethane/chitin was set due to miscibility reasons: above that the components did not form homogeneous materials, but coarse phase separated ones.

S20-494

Selective Solid Phase Extraction for Methoclopramide Made by Molecular Imprinted Polymers

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Molecular imprinting technique is a powerful method for preparing synthetic recognition sites with predetermined selectivity for various substances. Molecularly imprinted polymers (MIPs) are suited as antibody-like materials due to their long-term stability, chemical inertness and insolubility in water and most organic solvents. In this work, the molecularly imprinted polymer was synthesized by precipitation polymerization, using methoclopramide hydrochloride as a template molecule, methacrylic acid (MAA) as a functional monomer and ethylene glycol dimethacrylat (EGDMA) as a cross linking agent. Metoclopramide hydrochloride is a medicine that increases the movements or contractions of the stomach and intestines. Solid-phase extraction (SPE) columns packed with materials based on molecularly imprinted polymers (MIPs) were used to develop selective separation and preconcentration for methoclopramide from aqueous solutions. Methoclopramide was loaded on the MIP-SPE cartridge using different loading conditions (solvents, pH values). Washing and elution of the methoclopramide bound to the MIP was studied utilizing different protocol. The extraction protocol was successfully applied to the direct extraction of methoclopramide from real sample such as urine and plasma. References: [1] L.I. Andersson, J. Chromatogr. B 739 (2000) 163.[2] B. Sellergren, Anal. Chem. 66 (1994) 1578.

S20-497

Preparation of a molecularly imprinted polymer based solid-phase extraction of betamethasone acetate from human plasma

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Molecularly imprinted polymers have been applied as selective sorbents in several analytical techniques, including liquid chromatography, capillary electrophoresis and capillary electrochromatography, solid-phase extraction, and 'immunoassay'.An advantage of this type of sorbent is the possibility to synthesize polymers with selectivity pre-determined for a particular analyte. A method using a molecularly imprinted polymer (MIP) as the selective sorbent for solid-phase extraction (SPE) has been developed. The MIP was synthesized using betamethasone acetate as a template molecule , methacrylic acid (MAA) as a functional monomer, ethylene glycol dimethacrylate (EGDMA) as a cross linker and 2,2-azobisisobutyronitrile)AIBN) as a initiator.Betamethasone acetate is a moderate-potent glucocorticoid steroid with anti-inflammatory and immunosuppressive abilities, used especially where water retention is undesirable. Solid-phase extraction (SPE) columns packed with materials based on molecularly imprinted polymers (MIPs) were used to develop selective separation and preconcentration for betamethasone acetate from aqueous solutions. Betamethasone acetate was loaded on the MIP-SPE cartridge using different loading conditions (solvents, pH values). Washing and elution of the betamethasone acetate bound to the MIP was studied utilizing different protocol. The extraction protocol was successfully applied to the direct extraction of betamethasone acetate from plasma. References: [1] A . Martın-Esteban, Fresenius J. Anal. Chem. 370 (2001) 795.[2] M .T. Muldoon, L.H. Stanker, Anal. Chem. 69 (1997) 803.[3] C . Berggren, S. Bayoudh, D.C. Sherrington, K. Ensing, J. Chromatogr. A 889 (2000) 105.

S20-499

Synthesis and characterization of molecularly imprinting polymers for carbamazepine and determination of drug release profile

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Molecular imprinting technique is a powerful method for preparing synthetic recognition sites with predetermined selectivity for various substances. Molecularly imprinted polymers (MIPs) are suited as antibody-like materials due to their long-term stability, chemical inertness and insolubility in water and most organic solvents. In this work, the molecularly imprinted polymer was synthesized by precipitation polymerization, using methoclopramide hydrochloride as a template molecule, methacrylic acid (MAA) as a functional monomer and ethylene glycol dimethacrylat (EGDMA) as a cross linking agent. Metoclopramide hydrochloride is a medicine that increases the movements or contractions of the stomach and intestines. Solid-phase extraction (SPE) columns packed with materials based on molecularly imprinted polymers (MIPs) were used to develop selective separation and preconcentration for methoclopramide from aqueous solutions. Methoclopramide was loaded on the MIP-SPE cartridge using different loading conditions (solvents, pH values). Washing and elution of the methoclopramide bound to the MIP was studied utilizing different protocol. The extraction protocol was successfully applied to the direct extraction of methoclopramide from real sample such as urine and plasma. References1.Makoto, K., Takeuchi, T., Makawa, T., Molecular imprinting from fundamentals to application, Willey-VCH, 2003.2.Alvarez, c., Yanez , F., imprhnted soft contact lenses az norfloxacin delivery systems, J. controlled release, 2006, 113 236-2443.Slinchenko, O., Rachkov, A., Miyachi, H., Ogiso, M., 2004, Biosens.Bioelectron, 20, 1091-1097

S20-504

Preparation of Antibiotics Encapsulated Microfibers by using Electrospinning

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Poly(D,L-lactide-co-glycolide) (PLGA), Polycaprolactone (PCL), alginate and gelatin have been extensively investigated biocompatible polymer for various applications. We prepared antibiotics particles loaded biodegradable microfibers by using electrospinning. Electrospinning is a well-known method for obtaining polymer fibers with diameters as low as a few nano/micrometers and length in the range of several centimeters. Antibiotics with PCL or PLGA is emulsified in alginate or gelatin solutions which are injected using singe needle by high voltage. Diameters of fiber could be controlled by various voltages, solution flow rates and solution weight percent. Morphologies and diameters of fibers are confirmed by scanning electron microscope (SEM). Encapsulated antibiotics are observed by using confocal microscopy. We make observation release behavior of antibiotics and degradation times of fibers in PBS at 37 °C.

S20-505

Molecularly Imprinted Solid Phase Extraction for the Selective Spectrophotometric Determination of Verapamil

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Molecular imprinting is a rapidly developing technique for preparing polymeric materials that are capable of high molecular recognition. During the last year, application of molecular imprinting polymers (MIPs) as affinity phase in solid phase extraction and as stationary phase in liquid chromatography is rapidly developed. Solid phas extraction is commonly used for sample clean up and for concentration of analytes at trace levels. It is usually used to clean up a sample before using analytical method to quantities the amount of analyst(s) in the sample. In this work, the MIP was prepared by using methacrylic acid (MAA) as functional monomer, ethylene glycol dimethacrylate (EDMA) as cross-linker, chloroform as porogen and verapamil as template molecule. The results clearly indicate that synthesized MIPs have higher binding ability as compared with corresponding non-imprinted polymers. Verapamil is used to treat irregular heartbeats (arrhythmias) and high blood pressure. It relaxes blood vessels so the heart does not have to pump as hard. Physical and chemical variables were optimized, and the selectivity was appraised using some molecules similar to verapamil. The MIP described was used as a solid phase extraction phase for the extraction of verapamil from various sample matrices including urine and plasma coupled whit spectrophotometer for the analysis of the drug. Reference:1- R.N. Brogdn, P.Benfield, Drugs 51 (1996) 792.2 – A. Beltran , E. Caro ,R.M. Marce, P.A.G. Cormack, D.C. Sherrington, F.Borrull, Analytica Chimica Acta (2007)6.

S20-513

Rheological Analysis of Gelation and Gel dissolution of Poly(N-isopropylacrylamide) (PNIPAA) and Poly(N-isopropylacrylamide co-Acrylic acid) (PNIPAAcoAA) water mixtures

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Poly(N-isopropylacrylamide (PNIPAA) exhibits a well-defined lower critical solution temperature (LCST) in water and cross-linked PNIPAA gels undergo analogous collapse transitions in aqueous solvents. The LCST, and the accompanying changes in polymer conformation, result from a balance between hydrogen bonding and hydrophobic effects in aqueous solutions. The PNIPAA LCST can be controlled by copolymerization with other monomers (i.e. acrylic acid). The addition of hydrophilic monomers typically increases the LCST whereas the incorporation of more hydrophobic units has the opposite effect. The rheological behaviour of poly(N-isopropylacrylamide) (PNIPAA) and poly(N-isopropylacrylamide co-Acrylic acid) (PNIPAAcoAA) in aqueous media is of interest from both theoretical and practical perspectives. These aqueous polymer systems show thermally reversible gelation upon heating (inverse gelation) that is relatively rare and is only reported with modified polysaccharides, such as an aqueous solution of methyl cellulose, the mechanism of which is not yet clearly understood.The gelation and gel dissolution of poly(N-isopropylacrylamide) (PNIPAA) and poly(N-isopropylacrylamide co-Acrylic acid) were examined by rheology and NMR methods as increasing or decreasing temperature.

S20-528

Versatile Sugar Derivatives in the Synthesis of Potential Degradable Hydrophilic-Hydrophobic Polyurethanes and Polyureas

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The ever-increasing diversity of industrial activity is responsible for the discharge of compounds that are toxic or difficult to degrade into the environment. Carbohydrates and vegetable oils are considered as replacements for some petroleum oil-derived polymer precursors. The use of renewable resources (mainly carbohydrates) in rigid polyurethane and polyurea foams has been known to offer several advantages, such as increased strength, improved flame resistance, and enhanced biodegradability. Most of the polyurethanes are used as commodity materials and/or present industrial applications; some of them are biodegradable and/or biocompatible materials. Therefore, biomedical field is one of the areas of greatest interest for their potential applications. The low toxicity, potential biodegradability, biocompatibility, and versatile structures of polyurethanes make them suitable as part of drug administration systems, dermatological dressing, hemocompatible materials as catheters, presenting an important reduction in their thrombogenic character, and also as filters and biomedical instrumentation and device.In the present communication we describe the synthesis of new sugar-based monomers as the 2,3,4-tri-O-benzyl-L-arabinitol, 2,3,4-tri-O-benzyl-xylitol as well as their 1,6-diamino-1,6-dideoxy derivatives. By reaction of these monomers with different diisocyanates we have obtained a series of new polyurethanes and polyureas. The characterization of these polymers has been completed and some of their properties have also been studied, mainly thermal properties and hydrolytic degradability.

S20-698

Preparation and characterization of Polycaprolactone (PCL) modified by insertion of functional groups

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PCL is a biocompatible polymer and is reported to promote cell adhesion. Modification of PCL by insertion of functional groups was proposed in order to modulate properties as hydrophilicity and crystallinity and to create specific interactions with drugs or proteins. In this communication, functionalization with unsaturated monomers bearing functional groups such as anhydride and acid (PCL-g-MAGMA) or tertiary amines (PCL-g-DMAEA) was performed by radical grafting in a Brabender mixer. The functionalized PCLs were characterized through viscosimetry, optical microscopy, differential calorimetry, thermogravimetric analysis and rheology. A severe degradation was detected only for PCL-g-MAGMA, while no significant variation of Mw was found for PCL-g-DMAEA. The overall isothermal kinetics of crystallization of the two modified samples studied with calorimetric and rheological techniques always showed, for a given crystallization temperature, a decrease of the time required for the overall crystallization process respect to the neat PCL. Besides with dynamic scans were determinated thermodynamic parameters associated to the crystallization and fusion processes. Spherulite growth rate was studied by using a polarizing optical microscope; in all samples the growth starts with non-spherical entities and ends up with the spherulites. The characteristic for PCL-g-MAGMA and PCL-g-DMAEA is the ring patterns, already observed for PCL blends. The rheological analysis of the melt showed significant differences in the viscosity and modulus values in the low frequency region for the two grafted samples vs the neat PCL. Moreover, the effect of temperature on the storage and loss modulus profiles was investigated to build a master curve and to evaluate the flow activation energy. The results showed that a master curve can be obtained only for the neat PCL, while the two grafted PCL don't behave as thermorheologically simple flui

S20-733

Synthesis and characterization of molecularly imprinted polymer for a chiral drug

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Various substances, containing one or more chiral centers, are used today in the pharmaceutical and food industries. In particular , in pharmaceutical application when racemic mixtures are administrated as a drug, the stereoselectivity of biological processes often determines that one of the enantiomers represents the more active isomer for a given action, while the other one might be even active in a different way for this reason , different methods were developed in order to produce pure optical isomer. One of these methods is "molecularly imprinted polymers". The technique is based on the molecular memory of the substrate to be recognized (template) and memorized in a polymeric material during its preparation .The following extraction of the template molecule will give specific recognition sites in the polymer .The recognition involve both , the strategic arrangement of functional groups and the formation of cavities having shape complementary to the template. Molecular imprinting is a rapidly developing technique to prepare polymers possessing high recognition properties.Because of these advantage we use this method to study enantioselectivity, with this technique , separation of a variety of racemates has been successfully achieved .The control of particle size suitable for different application such as selectivity and separation. Uniform molecularly imprinted microspheres and nanoparticles prepared by precipitation polymerization.The aim of this research is to show the ability of "" MIP "" to enantioselectivity of chiral drug (pesudoephedrine). To obtain the MIP with uniform size we use precipitation polymerization and then characterize the MIP and study the interaction by the SCM , TGA , DSC , FT-IR , H-NMR , UV-VIS and polarimeter , and compare the MIP of enantiomers and racemates in different solution to show the selectivity of MIP.References:[1] Richard J . Ansell, Advanced Drug Delivery Reviews 57 (2005) 1809-1835[2]L.Donato, A

S20-740

Preparation and characterization of biodegradable/biocompatible ABCBA pentablock copolymers

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New biodegradable/biocompatible ABCBA pentablock copolymers, poly (L- Lactide)-b-poly (p-Dioxanone)-b-poly (ethylene glycol)-b-poly (p- Dioxanone)-b-poly (L-Lactide) (PLLA-b-PPDO-b-PEG-b-PPDO-b-PLLA) were synthesized and characterized. First PPDO-b-PEG-b-PPDO was synthesized by a coordination-insertion ring-opening polymerization of DON monomer, which in the mechanism PEG acts as the macro initiator and stannous octate as the catalyst. In the second step, L- Lactide monomers as the other blocks add to this. The resulting copolymers were composed of hydrophilic and hydrophobic segment, which hydrophobic PLLA blocks located in ends of these pentablock copolymers. Chemical composition and micro structural variation of the resulting copolymers were investigated by and and thermal properties by differential scanning calorimetry (DSC) and thermal gravimetric analysis (TGA). The intrinsic viscosity of copolymers was measured in chloroform solution. Reference: 1.Duda A, Biela. Polym. Degrade.Stab.59, 215(1998) 2.Bero M, Dobrzynski P, Polym.Bull. 42,131(1999) 3.J.T. Hong, N.S.Cho, H-S. Yoon, J.Polym. Sci. Polym. Chem 42,2790(2005)

S20-796

Controlled delivery of plasmid DNA by cationic PCL based biomaterial

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The two major approaches to the preparation of synthetic vectors for gene therapy involve the condensation of polyanionic nucleic acids with polycationic polymers (polyplexes) such as polyethylenimine (PEI), dendrimers, and peptides and/ or with cationic lipids (lipoplexes). Although such synthetic vectors have many advantages over viral vectors, including lower immunogenicity, ease of large-scale preparation, and unlimited packaging capacity, poor gene transfer efficiencies have to date precluded their use. Many attempts have been made to improve the efficacy and selectivity of synthetic vectors.This report investigates the effects of delivery of plasmid DNA encoding for CPT-1A/EmGFP chimera from a polymer based on polycaprolactone (PCL) and an alchylammonium-substituted acrylic acid (NAC). PCL (Mw=50,000-60,000 Da) has been functionalized with NAC in a Brabender mixer, in presence of benzoyl peroxide. Temperature, screw rate, and time of reaction were varied in order to obtained a suitable extent of functionalization, The melt reaction was stopped before the start of degradation processes of the polycaprolactone substrate, which would be indicated by a decrease in melt viscosity. The extent of functionalization was assessed by ATR-FTIR, 1H and 13C NMR. Thermal properties of the polymer were studied by DSC and Thermogravimetry. PCL-g-NAC film was obtained by casting from chloroform solution, sterilized by UV ray and soaked in DNA plasmid solution for 4h to reach a maximum adsorption of DNA on the surface. The cationic polymer formed firm noncovalent complexes with DNA. In addition, such cationic polymer-DNA complexes were capable of transfection of murine fibroblasts in vitro with lower cytotoxicity than that of commonly used cationic lipid-based gene-delivery systems (lipofectamine).

S20-804

Development of polyvinyl alcohol-glycine composite membranes based transdermal iontophoretic drug delivery system

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The objective of the present study was to evaluate the feasibility of using prepared polyvinyl alcohol (PVA) and glycine (Gly) blended composite membranes as a transdermal patch for the delivery of glycine by iontophoresis. PVA-Gly membranes, with different PVA:glycine ratios (1:0.2, 1:0.4, 1:0.6, 1:0.8 and 1:1 w/w), were prepared using the conventional solution casting method. Iontophoresis is the application of a small electric current, which enhances the delivery of ionized as well as unionized drug molecules through any synthetic or biological membrane, like skin. In-vitro iontophoretic diffusion experiments were done using a two-compartment horizontal glass diffusion cell, with Ag/AgCl electrodes. Pulsed direct current (1 KHz frequency) was applied by a custom designed portable current delivery device. Although continuous current (dc) was observed to be more potent for glycine delivery than pulsed current, pulsed dc was used to avoid the polarization effect caused by continuous current. Increasing the glycine content in the membrane or increasing the duration of current application resulted in enhancement of drug flux. Enhancement of drug flux was observed on pretreatment of the skin with 75 % alcohol. Presence of competitive ions during iontophoresis resulted in the decrease in drug flux. Among the different membranes used for the study, the membranes of PVA:gly ration of 1:0.8 & 1:1 showed almost similar results with maximum drug flux.

S20-844

Molecularly Imprinted Polymer Nanoparticles Prepared by Miniemulsion Polymerization

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Cross-linked polymer nanoparticles for primidone (antiepileptic drug) have been prepared by miniemusion polymerization using methacrylic acid (MAA) or methyl methacrylate (MMA) as a functional monomer and trimethylopropane trimethacrylate (TRIM) as a cross-linker agent. This technique enables one to exercise much better control over the size and shape of the particle and also to narrow significantly the size distribution. Nano scale diameter of molecular imprinted polymers (MIPs) assures a high specific surface area and also improves the accessibility of the binding sites. These are the major advantages of this technique.In this study, we focused on obtaining MIPs in the final morphology of nano beads, and this MIPs were analyzed by scanning electron microscopy.The release of drug by MIPs with different ratios of MAA and MMA was investigated. Three factors regulated the release properties of MIPs: the amount of MAA units in MIP, the dimension of particles, and the hydrophilicity of particles.The load of drug and binding capacity of MIPs were analyzed by UV spectroscopy. The results showed that the load of drug is proportional with MAA amount, and also the imprinted nanoparticles can be considered promising systems able to release and afterwards rebind template molecules with out the need of drastic extraction procedure.References [1] Dorothea Vaihinger, Katharina Landfester, Iris Krauter, Herwig Brunner, Gunter E. M. Tovar, Macromol. Chem. Phys. 2002, 203, 1965-1973. [2] Huai You Wang, Ji Gang Jiang, Li Ying Ma, Yan Ling Pang, Reactive and Functional Polymers. 2005, 64, 119-126. [3] G. Ciardelli, B. Cioni, C. Cristallini, N. Barbani, D. Silvestri, P. Giusti, Biosensors and bioelectronics. 2004, 20, 1083-1090.

S20-940

Electrospun PLGA-organoclay Nanocomposite Scaffold; Morphology, Mechanical, and Biodegradation Characteristics

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Tissue engineering involves fabrication of three dimensional scaffolds to support cellular in growth and proliferation. These scaffolds should be similar to native extra cellular matrix(ECM) in terms of both chemical composition and physical structure. Elecetrospun polymer nanofiber scaffolds are the best candidates for ECM mimetic materials since they mimic the nano scale properties of ECM. PLGA (poly lactide co glycolide) is a suitable synthetic biodegradable polymer for tissue engineering application. In this study, neat PLGA and PLGA /organoclay (OMMT) nanocomposite were electrospun and the effect of OMMT on the morphology, mechanical and biodegradation properties of nanofiber scaffolds were investigated. The average diameters of the nanofibers were ranged from 500 to 1000 nm. The morphology of nanocomposite was exfoliated. The nanocomposite fibrous scaffold exhibited better mechanical properties and structural integrity during biodegradation process. The shrinkage of the nanocomposite scaffold in cell culture medium was decreased in compare to neat polymer counterpart.

S20-944

ON THE SYNTHESIS AND CHARACTERIZATION OF POLY(ASPARTIC ACID)

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Polyaspartic acids (PAA) belongs to polyamine acids that are an important class of synthetic polymers produced by chemical polymerization of the same amino-acid building blocks found in naturally occurring proteins. They occur in nature as polypeptide sequences, i.e. in mollusk shells or snails, regulating the calcium balance of these organisms. Due to the carboxylate groups, polyaspartate has similar properties to the polyacrylates. The polyaspartate polymers are analogues of natural proteins, particularly the aspartate-rich proteins from oyster shells that play a key role in regulating mineralization. Since the polyaspartate compounds are derived from natural precursors, they are biodegradable and they can be used to replace petroleum-derived polymers such as polyacrylate and polyacrylamide. Because of their unique mineralization and ionic properties, there exist possible applications for the polyaspartate materials. In the medical domain it is possible its application as superabsorbents, biomedical devices for prevention of pathological calcification, microencapsulation for drug delivery, surface coatings for implants to promote crystallization, tartar control agents. PAA can be produced by thermal polymerization of l-aspartic acid. The thermal polymerization of l-aspartic acid with or without catalyst leads to the formation of succinimide units, and the resulting PAA after hydrolysis of polysuccinimide is composed of 70% of β-amide and 30% of α-amide units. In the paper it is presented the synthesis and characterization of the poly(aspartic acid) from the viewpoint of zeta potential as a function of pH and conductivity, with some considerations on the changes induced by the preparation procedures and in direct connection with its final application as a biomaterial. The rheological behavior in dilute solution is also evidenced. Acknowledgement: Financial support for this work is supported by Romanian Ministry of Research and Education through GRANT no. 10 / 2

S20-945

AN APPRAISAL OF INTERPOLYMER COMPLEXATION OF POLY(ASPARTIC ACID) WITH POLY(ETHYLENE GLYCOL)

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Polymer complexes are macromolecular structures formed by the noncovalent association of polymers that have an affinity for one another. Interaction between two different macromolecular species may lead to the formation of an inter-macromolecular complex which essentially possesses properties entirely different from those of the component polymers. The major classes of polymer complexes are stereocomplexes, polyelectrolyte complexes, and hydrogen bonded complexes. Among these complexes, the hydrogen bonded complexes have attracted attention because of their special character; many biological phenomena such as enzymatic processes, supermolecular assemblies in virus shells, and muscle contraction are related to intermacromolecular complexes governed by hydrogen bonds.In this study polymer complex of poly(aspartic acid) and poly(ethylene glycol) were prepared, as matrices for subsequently doping with antimicrobial substance, with biomedical applications. The formation of hydrogen bonded complexes between poly(aspartic acid) and poly(ethylene glycol) has not been yet reported.Poly(aspartic acid) (PAs) as a biodegradable water-soluble alternative to polycarboxylates, is an environmentally friendly chemical used as dispersant, detergent builder and in biomedical applications.Polyethylene glycol (PEG) is a hydrophilic nonionic polymer used in many biochemical, cosmetics, food, and pharmaceutical products, due to its nontoxic character, and mild action on the biological activity of cell components.Complexation in aqueous solution by the formation of branched structures between polyacid molecule and polyether chains was evidenced by laser light diffusion in zeta potential estimation and by dynamic rheology. DSC analysis there was also used to underline the complexation in the system.Acknowledgement: Financial support for this work is supported by Romanian Ministry of Research and Education through GRANT no. 10 / 2005

S20-954

BLENDS WITH BIOMEDICAL APPLICATIONS

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The preparation of an intermolecular complex between poly(aspartic acid) – a nontoxic, biocompatible and biodegradable polymer – poly(vinyl alcohol) – a synthetic polymer with film-forming properties – and an antiinflammatory drug (diclofenac) is investigated. Poly(aspartic acid) belongs to polyamine acids that are an important class of synthetic polymers produced by chemical polymerization of the same amino-acid building blocks found in naturally occurring proteins. Dynamic rheology, zeta potential values, and the pH influence upon zeta potential and dimension size, are used to evaluate the complex shape for its future biomedical applications. Zeta potential measurements are very powerful tools in many areas of pharmaceutical science. Also, a combination of the measurement of zeta potential and size has been shown to provide an insight into mechanisms of dispersion stability. The stability of particle dispersion depends upon the balance of the repulsive and attractive forces that exist between particles as they approach one another. The magnitude of the zeta potential of the particles gives us the measure of the particle interaction. Also, zeta potential measurements give information about the overall surface charge of the particles and how this characteristic is affected by changes in the environment (e.g. pH, presence of counter-ions, and adsorption of proteins). Charge shielding by hydrophilic groups can be used to predict the effectiveness of the barrier function against opsonisation in vivo. For demonstrate in vitro the biomedical applications of the blend it was study the kinetic of the drug release from the blends as film.

S20-1156

Preparation and Characterization of Herceptin-conjugated Chitosan Nanoparticles for siRNA Delivery

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Targeted cancer therapy is currently the leading method to treat cancers around the world. HER2-targeting cancer therapy is among the best models used in cancer researches and even in clinical trials. The deregulation of the HER2 protooncogene has been demonstrated in a wide variety of human cancers and found to be associated with the progress of malignancy and metastasis in animal models. The HER2-mediated malignant phenotypes of many human cancer cells can be repressed by targeting HER2 showing that HER2 itself is a good target for developing anti-cancer drugs.On the other hand, the short double-stranded RNA species, called small interference RNA (siRNA), can be used to silence the gene expression in a sequence-specific manner in a process that is known as RNA interference (RNAi). It has been demonstrated that siRNA can trigger RNAi to silence target genes in mammalian cells. Furthermore, RNAi-based therapies have been successfully implemented in a variety of disease models, including viral infection, hepatitis, cancer, ocular neovascularization, and neurodegenerative diseases. Therefore, the RNAi-based therapy has become a useful method for the analysis of gene functions and holds the significant possibility of therapeutic applications.Chitosan (CS) is a non-toxic biodegradable polycationic polymer with low immunogenicity. Its cationic polyelectrolyte nature provides a strong electrostatic interaction with negatively charged DNA, and protects it from nuclease degradation. In this study, Chitosan-siRNA nanoparticles were prepared using complex coacervation and crosslinking process. The loading efficiency of siRNA in these nanoparticles was very high (more than 95%). The cancer cells can be targeted and repressed by conjugation of herceptin on the nanoparticles.

S20-1194

DISSOLUTION ENHANCEMENT OF FLAVONOIDS BY INCORPORATION IN GASTRO-RESISTANT MICROSPHERES

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A common strategy to obtain enhanced bioavailability and customized release of slightly soluble and poorly absorbed drugs is their incorporation in controlled delivery systems, such as biocompatible polymers' microparticles.Among the several methods developed for the preparation of such microencapsulated systems, solvent evaporation is one of the most versatile and advantageous. In fact, it requires only mild conditions (e.g. ambient temperature and constant stirring); thus, a stable emulsion can be formed without compromising the activity of core materials.In this work gastro-resistant microspheres, containing quercetin or naringenin flavonoids as drug, were prepared by water in oil (W/O) solvent evaporation method, using cellulose acetate phthalate (C-A-P) or Eudragit L100 as enteric polymers. The formulation and preparation conditions were optimized to obtain high encapsulation efficiency and production yield. The synthesized microspheres were characterized in terms of particle size distribution, drug loading, morphology and in vitro release properties.The performed analyses evidenced that the microspheres were free-flowing and spherical in shape, both quercetin and naringenin were microencapsulated in the amorphous state and the drug content was near to the theoretical one. Preliminary in vitro dissolution studies, carried out using a pH-change method, showed that the samples exhibit a typical biphasic drug release trend, due to the pH dependent solubility of the enteric polymers used. In particular, the samples have a fairly gastroresistance followed by an about complete release in simulated intestinal fluid.

S20-1195

POLY(VINYL ALCHOHOL)/POLY(ETHYLENE GLYCOL) BLENDS AS DRUG CARRIERS FOR FLAVONOIDS

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Drug delivery systems play an increasingly important role in biomedical and pharmaceutical markets since they offer numerous advantages compared to conventional dosage forms, which include reduced toxicity, improved dosage capability and efficacy of the active product. Among the potential candidates as carriers for such applications, polymers occupy an unique position due to their wide range of structures and physicochemical properties. Some of the more interesting polymers for drug delivery applications are poly(vinyl alcohol) and poly(ethylene glycol), henceforth referred to as PVOH and PEG, respectively. In fact, they have unique properties including low toxicity, good biocompatibility and biodegradability and are approved for internal use in pharmaceutical. Moreover, they were hydrophilic and then swell in water or other biological fluids, so allowing the release of the incorporated drugs.In this work, PVOH/PEG blends having different composition were prepared and tested as drug carriers for very slightly soluble and poorly absorbed flavonoids. Aqueous solutions at different PVOH/PEG ratios were prepared and loaded with quercetin or naringenin flavonoids at 20wt%. Then, the solutions were cast onto glass slides and dried at room temperature. The obtained samples were submitted to several analyses (XRD, DSC, TGA, DMA, FT-IR, UV-Vis) in order to characterize their morphology, thermo-mechanical response, swelling behavior and in vitro release properties.

S20-1198

A pilot study on sub-micronic hydroxyapatite and polycaprolactone composite substrates synthesized by wet chemical method

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Biodegradable polymer/hydroxyapatite (HA) composites have potential application as bone graft substitute. Recently, different strategies have been developed to realize (HA)/PCL composites by physically blending commercial HA and the resorbable polymers [1]. The most significant problem occurring during the traditional blending process consists in a non-homogeneity distribution of ceramic phase in the polymeric matrix dramatically reducing the effective bioactive potential of the composite. The aim of this work is to develop novel HA/PCL composites at room temperature by wet chemical methods at room temperature. The chemical synthesis of the HA powder based the chemical interaction with a good solvent for the polymer enables to define a novel pathway to generate homogeneous composites materials preserving the micro-structural features of HA crystals.Composite of HA/PCL was prepared at room temperature by using Ca(NO3)2•4H2O and H3PO4 as precursors and tetrahydrofuran was used as the solvent. Poly(ε-caprolactone) (Mw=65000) was used as biodegradable polymer. NH4OH was utilized to adjust the pH of the reaction and the pH changes occurring during the reaction were observed for 24hrs using a pH meter. A consistent morphological investigation have been performed by Scanning Electron Microscopy (Leica 420), X-ray energy dispersive spectroscopy (EDAX) and qualitative estimation of Ca/P ratio. FITR spectra by Nicolet 5700 was used to study chemical interaction between the components. Phase analysis of the composites was conducted using X-ray diffraction. Proposed technique allows obtaining bioactive composite materials with a wide distribution of a single phase of stoichiometric HA, homogeneously dispersed in the polymer matrix resulting a promising alternative to the traditional physically blended composite scaffolds in the bone tissue engineering applications.References[1]Guarino V., Causa F., Ambrosio L.- Expert Rev.Med. Devices4(3) (2007)

S20-1206

Aminolysis of poly-epsilon-caprolactone: a route to surface bioactivation of polymeric scaffolds

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Poly-epsilon-caprolactone (PCL), along with its blends or composites, is widely used in drug delivery and regenerative medicine area. This popular biomaterials for tissue-engineering scaffold often has suboptimal properties when analyzed for cell attachment and growth. As an example, even though some studies have reported that osteoblast-like successfully grow on PCL, many cell lines (such as primary osteoblast) show scarce adhesion [1].The bioactivation of PCL material to promote cell attachment by binding of adhesion receptors thus represents a promising area of interest.Short amino acid sequence such as arginine-glycine-aspartic acid (RGD) has been widely used as recognition motif to mediate cell attachment [2]. Over the last decade, several biomimetic approach have been developed to immobilize short peptide such as RGD on polymeric surface.The aim of this work is to set-up a procedure to covalently immobilize RGD-based peptide sequence to functionalized PCL surface. The procedure consists of three steps: aminolysis of PCL substrate, grafting of a tether, peptide immobilization. Over other methods of immobilizating a bioactive molecules on polymer surface, the covalent immobilization provides a stable bond extending peptide half-life, prevent its metabolism and allow for a continued bioactivity of in-dwelling devices.As result, RGD sequences were covalently immobilized on PCL surface though grafting of a tether after polymer amynolisys. Proposed approach allows for a spatial distribution and as well as peptide immobilization control on polymer surface. We also demonstrated that this bioconiugation is compatible with three-dimensional PCL substrates to be used as scaffold for tissue engineering.
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S20-1211

Micellization and gelation behaviour of a PEO-PPO-PEO co-polymer (F127), investigated by means of calorimetric and dielectric measurements

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The block co-polymers PEO-PPO-PEO constitute a class of bio-compatible surfactants known as "Pluronics". Their amphiphilic nature is due to the PEO water soluble chains, and to the hydrophobicity of the PPO segment. Their water solutions exhibit complex phase behaviour with temperature. Starting from temperatures close to 0°C the affinity between water and PEO segments decreases upon heating, and the single chain of polymer in solution (the "unimers") can aggregate building up micelles, the phenomenon being the "micellization". Further temperature increase can cause a close packing of the micelles, building up to a soft gel with interesting properties for biomedical applications, the phenomenon is noted as "gelation". The Pluronic F127 (PEO100PPO65PEO100), whose solutions of proper concentration (around 20% w/w) give a gel at body temperature (37°C), is of particular interest in biomedical applications. E.g., its use is under consideration to cover the stents used in percutaneous transluminal angioplasty. To do this, a liquid solution (at 5°C) has to be pumped into a catheter and it has to gelate only in the proper position in the cardiac artery. To predict the solidification behaviour both the thermodynamics and the kinetics of the micellization and gelation have to be known. In this work, the micellization and gelation temperatures of F127 water solutions were carefully determined by means of DSC studies and of dielectric constant measurements, to obtain the micelle volume fraction and thus the gelation (which occurs when the volume fraction overcome the simple cubic volume density of 52.3%). Several Pluronic concentrations were analyzed, and the dielectric measurements were repeated for several temperatures, allowing to the sample enough time to reach equilibrium at the temperature tests. Thus, the thermodynamics of micellization and of gelation for water solutions of F127 has been determined over a wide range of temperature and solution concentrations.

S20-1238

Physico-Chemical Analysis of Radiation- Induced Modifications in Radiation Dosimetry Polymer Gels

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In order to minimize the dose to nearby critical organs, clinical applications of advanced radiation therapy techniques continue to progress rapidly, and applications of three dimensional conformal radiotherapy, intensity modulated radiation therapy and stereotactic radiosurgery have been carried on. These techniques have created an urgent need for three-dimensional (3D) dosimetry with high resolution and assured quality. However, conventional dosimetry detectors do not meet this requirement because they are limited in the measurable dimensions. The potential advantages of Magneting Resonance Imaging (MRI) gel dosimetry were recognized two decades ago. However, it did not become a practical tool because of the blurring of dose distributions and other problems. Many researchers worked to improve gel dosimetry, and complex dose distributions and accuracy using radiation-sensitive polymer gels have been demonstrated. Not all polymer gels using MRI have linearity in the dose range 0-1 Gy, and, also these polymer gels are expensive to synthesize and their quality is difficult to control. In order to overcome these problems, we prepared a polymer gel with high-radiation sensitivity, which is synthesized at low cost and with easy quality control. In addition to MRI several techniques can also be used to monitor the changes in the physical and chemical properties of gel dosimeters upon irradiation. Due to a lack of investigation of the fundamental physico-chemical changes taking place in these materials postirradiation it is necessary to undertake fundamental measurements on these systems to be able to develop gel dosimeters that will be of use in the field of radiology. This contribution aims to report the preparation and the effect of high energy radiation on the structural organization and physical properties of a polymeric gel in order to find a correlation between radiation dose absorbed and physico-chemical behavior of the analysed material

S20-1257

Controlled release of antifungal Miconazole nitrate from crosslinked poly(vinyl alcohol)hydrogel

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Poly (vinyl alcohol) hydrogel was prepared by the reaction between 99% hydrolysed poly (vinyl alcohol) with gluteraldehyde as a crosslinking agent . The hydrogel was loaded with antifungal Miconazole nitrate . The swelling ratio of the polymer antifungal delivery system was determined in Simulated Gastric Fluid SGF , Simulated Intestinal Fluid SIF and distilled water . The release rate of Miconazole nitrate from hydrogel was studied by using U.V. technique at constant temperature (37 oC) in SGF and SIF . The mechanism of degradation for the polymer antifungal delivery was also determined by bulk erosion mechanism . Finally the inhibition zone diameters for Miconazole nitrate , polymer miconazole nitrate delivery and polymer alone were studied against three types of fungi , Candida albicans , Cryptococcus neoformans and Fusarium oxysporum .

S20-1271

Study of Polymeric Composites for Medical Applications

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For decades, the biocompatibility and physicochemical properties of polyurethanes have been drawing attention of developers of composite materials widely used in medicine. Among the variety of their applications, polyurethanes have also performed well as drug carriers, especially when continuous drug delivery into a body was required. Such depot forms are essentially a polymeric base (matrix) into which low-molecular bioactive substances are introduced. Besides the extended drug delivery, advantages of the depot forms also include reduced side effects The present work compares drug-filled polyurethane (PU) composites synthesized by the prepolymer technique from oligoglycols of varied molecular weight and toluylene diisocyanate (a mixture of 2,4 and 2,6 isomers at a mass percentage ratio of 80 to 20). PU composites filled with three drugs were studied: cephasoline, antaxon, and piroxicam. Using IR spectroscopy, it was shown that the relative intensity of absorption band at 2,280 cm-1 (stretching vibrations of –N=C=O) was the same in the spectra of filled and non-filled prepolymers. However, introduction of a drug into PU matrix resulted in physical modification of the latter in all variants of composites studied. This, in turn, resulted in extended therapeutic effect of the PU/drug system. The wetting ability of PU matrix was shown to have a significant impact on the polymer degradation progress and the pattern of drug release from the polymer. The drug release dynamics was studied in vitro using reverse-phase HPLC. Cephasoline and antaxon were steadily released into model medium for 7 and 10 days, respectively. In vivo, PU composites with antaxon were tried on rats with alcohol dependence, and PU composites with with piroxicam, on rats with inflammation. In the former case, the therapeutic effect was observed for 3 days, and in the latter case, for 5 days.

S20-1301

Effect of ionizing radiation on polysulfone membrane fouling due to protein filtration

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Membrane separation processes have successfully been applied to biotechnology industry, but membrane fouling during filtration of proteins is still an important problem since it causes a strong decrease of permeate flow, which reduces the productivity of the process and the polymeric membrane life due to cleaning procedures (1). Temperature, hard chemicals, UV light and gamma-radiation are used for cleaning/sterilizing membranes in biotechnological and medical applications (2), but they can modify the polymer matrix and affect the barrier behaviour associated to a given membrane. Moreover, an adequate description of fouling mechanism (i.e. in membrane pores or on the membrane surface) is important to create favourable hydrodynamic conditions to minimize fouling. This work studies the effect of ionizing radiation (dose of 10 J/kg from a 60Co therapeutic unit) on a porous polysulfone membrane in connection to its fouling behaviour by the protein bovine seroalbumine or BSA. Time evolution of permeate flow, protein retention and water permeability for native (GRM) and irradiated (GRM-Ir10) samples were determined; clean and fouled samples (both untreated and irradiated) were also characterized by streaming/zeta potential at a given NaCl solution and different pHs. All these results can provide information on the main membrane fouling mechanism and to see differences associated to membrane irradiation. (1) M. Mulder, Basic Principles of Membrane Technology. Kluwer, Dordrecht, 1992.(2) F. Aucella, M. Vigilante, G. Gatta, E. Grandone, D. Colaizzo, M. Margaglione, S. Modoni, C. Stallone, Artificial Organs, 26 (2002) 543.

S20-1302

Modification of transport parameters trough different cellulose membranes as a consequence of protein fouling

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Cellulose is a natural polymer commonly used in manufacture of membranes for different separation processes: regenerated cellulose (RC) is mainly used as a material for dialysis membranes, while cellulose derivatives as cellulose acetate and cellulose nitrate are used for microfiltration and ultrafiltration, whereas cellulose triacetate shows good properties for desalination applications (1). Regenerated cellulose (or cellophane) is a polymer very hydrophilic but it is not water-soluble, due to its structure RC allows the diffusion of ions and low molecular weight solutes but it avoids the pass of proteins or macromolecules of high molecular weight, which is a basic requirement for hemodialysis membranes. This work presents the effect of protein adsorption in some characteristic membrane parameters (diffusional permeability, ion transport numbers and electrical resistance) determined for different RC membranes after being in contact with a solution containing the selected protein. Membrane parameters were obtained from salt diffusion, membrane potential and impedance spectroscopy measurements carried out with the membranes in contact with NaCl solutions at different concentrations. A comparison of the results obtained with pristine and protein-fouled samples allows the estimation of modifications in the membrane characteristic parameters associated to protein deposition and its effect on transport across cellulose based dialysis (or hemodialysis) membranes. (1) M. Mulder, Basic Principles of Membrane Technology. Kluwer, Dordrecht, 1992.Acknowledgements: Project MAT2007-65065

S20-1323

Morphological changes during plastic deformation of PHB, PHBV and PHB/PCL compressed moulded samples suitable for tissue engineering

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The formulation of a suitable scaffold able to facilitate the several biological processes needed for skin reconstruction is a major issue for groups developing dermal and epidermal substitutes. The challenge is to develop materials and tissue engineering technologies to produce a complete skin equivalent consisting of both a dermal and an epidermal layer, an entirely autologous product, designed for the treatment of acute skin wounds and chronic skin wounds.The biocompatibility of poly(3-hydroxybutyrate) (PHB), poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) and poly(ε-caprolactone) (PCL) has been studied profusely studied. PHBV thin films used for retinal pigment epithelium cell culture, implants for osteomyelitis therapy and PHBV matrices for bone tissue engineering are examples of applications of PHA as tissue engineering materials. The purpose of this contribution is to show the morphological changes produced in the amorphous matrix and the spherulites of PHB, PHBV and a blend of PHB and PCL under tensile tests until failure. To achieve this goal, specimens with different spherulitic structures were produced by compression molding. A new etching agent to reveal details in the spherulitic structure is presented. Micro-tensile tests allow to follow morphological changes during uniaxial loading by optical microscopy and to determine the different crack propagation mechanisms.Crazing and shear bands are the strain mechanisms operating within the amorphous matrix of PHBV. In the closely packed spherulitic morphology, crack propagation may be: along the circumferential planes of the spherulites, trans-spherulitic along radial or circumferential planes or through the sharp boundaries between adjacent spherulites.

S20-1330

Thermal, mechanical and morphological characterization of Poly(lactic acid) - polyadipate films during aging

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Poly (lactic acid) (PLA) is a bio-based thermoplastic aliphatic polyester which has been extensively investigated during the last decades as an alternative to polymers obtained from petroleum (1-2). However, its high brittleness limits PLA uses in flexible films and sheets. This drawback can be overcome by mixing the polymer with environmentally friendly plasticizers. PLA (Hycail CML) was melt-blended with three commercial polyadipates based on adipic acid and 1-3 propanediol with different molar masses between 1700 and 3400 g/mol. Blends were prepared in mass ratio varying from 10 to 20% (w/w) and processed into films by compression moulding. Compatibility of blends after processing was studied, but also its morphological stability to assure that the blends remain miscible during the foreseen shelf-life of the material (1). The obtained films were kept at 25ºC and structural, thermal, mechanical and thermo-mechanical properties were studied at different storage times in order to evaluate the stability of the samples. DSC analysis revealed that the incorporation of polyadipates reduced the glass transition temperature of PLA in all cases, but this effect was more pronounced for the plasticizer of lower molar mass. The addition of polyadipates was also characterized by an increase in PLA chains mobility which induced crystallization during heating. X-ray Diffraction showed that all the quenched blends were mainly amorphous but some of them developed crystallinity during aging. Phase separation was observed with time for some of the films at the higher plasticizer concentration. Mechanical and thermo-mechanical properties showed an increase in ductility with the plasticizer content but there was a reduction in such properties upon aging due to matrix densification and crystallization of PLA chains.(1) I. Pillin, N. Montrelay, Y. Grohens, Polymer, 2006, 47, 4676.(2) V.P. Martino, R.A. Ruseckaite, A. Jiménez, J. Thermal Anal. & Calorimetry, 2006, 86, 707

S20-1356

Comparative study of diagnostic and therapeutic medical ultrasound gels

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The medical ultrasound gels are usually viscous, non irritating, bacteriostatic, conductive thus recommended for ultrasonic diagnosis, ultrasonic therapy, ECG, EEG, EMG, defibrillation etc. It is also reported that ultrasound conductive gel has various applications like: breast area, testicles, thytoid, carotid artery, superficial structures etc. The aim of our main object on the research is to improve / control the viscoelastic properties and conductivity of gels. The present paper discusses about the fluid characteristic of three different kinds of commercially available medical ultrasound gel i.e. ULTRASONIC GEL, SORTIS SONO GEL and US-GEL PROFESSIONAL. According to the responses about properties of these gels, many specialists (doctors, pharmacists) commented that the SORTIS SONO GEL has good sound transmission, distinctive image display and easily wipe out the left over coupling agent with tissue paper. When investigated in our laboratory, all these three gels have shown very interesting results in case of conductivity as well as viscoelastic properties. Although, these gels have shown good conductivity, their bacteriostatic property is unlike. Bacteriostatic properties of the gels have been investigated in presence of Escherichia coli and Staphylococcus aureus. It was noticed that SORTIS SONO GEL has reasonably good antibacterial property compared with US-GEL PROFESSIONAL but ULTRASONIC GEL has shown negative response.

S20-1384

Characterization of nano-structured poly(D,L-lactid acid) and poly(L-lactide acid) nonwoven mats produced via electrospinning

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The applications of poly(D,L-lactic acid), PDLLA, and poy(L-lactide acid), PLLA, nanofibers mats are implant substrates, skin regeneration and coating and drug release. Many different techniques have been developed to produce nanostructured biodegradable articles such as microspheres, foams, films and fibers. Biodegradable nanofibers can be produced by electrospinning of a polymeric solution. In this work, nano-structured PDLLA and PLLA nonwoven mats were prepared by electrospinning. The effect of solvent on its electrospinning is investigated. The morphology, crystallinity and thermal properties of electrospun PDLLA and PLLA mats were characterized by scanning electron microscopy (SEM), wide-angle X-ray diffraction (WAXD) and differential scanning calorimetry (DSC). The results demonstrated that the solution properties is one of the main parameter in electrospining and it controls the porous structure. The obtained mats are amorphous, contain residual solvents and their average diameter decreases with the decrease of the solution concentration and the increase of the electrical field.