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Introduction

Polymer Chemistry for the Design of New Materials

On 19 and 20 September 2002, an international symposium '*Polymer Chemistry for the Design of New Materials*' took place in Ghent, Belgium, to pay tribute to the achievements of ***Professor Eric Goethals*** in the field of polymer chemistry on the occasion of his retirement from Ghent University in October 2002. The symposium, organised by the Polymer Chemistry and Materials groups at the Department of Organic Chemistry, Ghent University, was held under the auspices of the European Polymer Federation (EPF), the Belgian Polymer Group (BPG) and the SCI journal *Polymer International*. The aim of the symposium was to give an overview of major advances in the design of new polymer architectures. Fifteen keynote lectures and 65 poster contributions covered the fields of polymerisation mechanisms, design of polymer architectures, supramolecular chemistry and new functional polymer materials. Professor Eric Goethals also gave an overview on 40 years of polymer chemistry at Ghent University. The introduction to this special issue is taken as an occasion to highlight the most important aspects of his outstanding career.

Professor Eric Goethals was born in Ghent on December 6, 1936. He studied chemistry at Ghent University, graduating in 1958. In the same year, he started his PhD research under the guidance of Firmin Govaert, Professor of Organic Chemistry at the same university. He defended his dissertation 'New Sulfur Derivatives of Pentaerythritol' in 1963.

Professor Govaert asked him to start up research on polymer chemistry and, in order to gain some experience in the field, he completed a post-doctorate assignment at the Johannes Gütenberg University at Mainz under the guidance of RC Schulz. Back in Ghent, at the end of 1963, Goethals then explored different topics in polymer synthesis and modification. In particular he studied cyclopolymerisations and modifications of polymers with 'sultones', ie cyclic esters of hydroxysulfonic acids, the sulfur analogues of lactones. A second topic that started in these early years was the polymerisation of four-membered heterocyclic monomers. At that time, it was already known that oxetanes could be polymerised by a cationic mechanism. However, the mechanism of polymerisation was still unclear. This line of research was a most timely one and developed into one of the major fields of research for which the Polymer Chemistry group in Ghent became known: Cationic Ring-Opening Polymerisations (CROP).

In 1968, Goethals made a short visit to two distinguished experts on cationic polymerisation: David Pepper at Trinity College in Dublin and Peter Plesch at Keele University. As a result, the group started to elucidate the mechanism of the cationic polymerisation of thietanes and, somewhat later, of azetidines by means of kinetic measurements. It was shown that the active species in these cationic polymerisations are cyclic onium salts and that the occurrence of a termination reaction, proposed on the basis of the kinetic studies, could be confirmed by direct measurement of the concentrations of cyclic and non-cyclic onium ions as a function of time. In this way, absolute rate constants for propagation and for termination were determined for a large number of ring-opening polymerisations. At that time, polymerizations were either considered as 'living' (ie according to the definition of Michael Szwarc, without termination nor transfer) or 'non-living'. On the basis of a knowledge of the rate constants of propagation and of

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termination, Goethals proposed in 1976 a first attempt to 'rank' polymerisations according to their 'living character', expressed as the ratio k_p/k_t.

In the late 1970s, the group started to study the cationic polymerisation of three-membered heterocyclic monomers: thiiranes, aziridines and oxiranes. It was discovered that, in this kind of polymerisations, cyclic oligomers were generally formed as side products and, in some cases, even as the main end-products. The importance of the presence of substituents on the living character of the polymerisations was demonstrated. This study opened the way to a large number of end-group functionalised poly(*N*-tert-butylaziridine)s yielding telechelics, macromonomers, amphiphilic block-copolymers and *N*-tert-butylaziridine-containing polymer networks.

In 1977 Goethals was asked to organise, with Takeo Saegusa of the Kyoto University, an 'International Symposium on Ring-opening Polymerisation' in the frame of an ACS meeting in New Orleans. Also in 1977, he was invited by the Japanese Society for the Promotion of Science to visit a number of polymer departments at Japanese universities. In 1979, Goethals organised an IUPAC-sponsored international symposium on 'Polymeric Amines and Ammonium Salts', which resulted in the publication of a book of the same title. In 1983, the sixth 'International Symposium on Cationic Polymerisation and Related Processes' was organised at Ghent, recognising the role played by the Ghent group in this field.

Around 1980, the interest of the group shifted from mechanistic studies to the design of new polymer materials. A first example was the development of a new anti-felting polymer for wool. The group then focused on polymerisations of 'easy' monomers such as tetrahydrofuran and cyclic acetals. The Ghent group made a detailed study of the synthesis of various telechelics, macromonomers, star-shaped polymers, block-copolymers and polymer networks derived from these monomers. In 1989, the book 'Telechelic polymers: synthesis, properties and application' edited by Goethals was published.

When the 'living' polymerisation of vinyl ethers was reported by Higashimura and Sawamoto in 1983, Goethals saw the similarity between the mechanism of vinyl ether polymerisation and that of cyclic acetal polymerisation. It was decided to use the expertise on cyclic acetal polymerisation for vinyl ether polymerisation. This led to the development of a number of new methods for initiation and end-capping for cationic polymerisations of vinyl ethers and to the development of new materials based on poly(vinyl ether)s. Materials with shape memory properties, thermotropic polymers and thermo-responsive polymer materials were detailed. One of the key properties in these new materials was the lower critical solution temperature (LCST) properties of poly(methyl vinyl ether).

In 1996 Goethals became head of the Department of Organic Chemistry and, at the same time, was elected representative of the Faculty of Sciences at the Central Board of the Ghent University. During his 35 years of professorship, Goethals has been promoter of more than 70 PhD theses. We would like to take this opportunity to thank him for his hard work and dedication to the University and to the advancement of science.

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