Summary

Readers who are interested in a more detailed summary of the discussed chapters are referred to the abstracts at the beginning of each chapter.

For a long time it was thought that pure carbon only existed in two allotropes (forms). The first allotrope is known as graphite and consists of ‘loose’ layers of carbon. The second allotrope is diamond, which is formed by a three dimensional network of carbon atoms. In the 1985 a third allotrope of carbon was discovered, the so called fullerenes (see figure 1). Fullerenes are molecules consisting of carbon, forming a closed sphere structure. These spherical structures are built up out of hexagons and pentagons. Each fullerene, per definition, consists out of 12 pentagons, since a sphere containing \( n \) hexagons can not be closed otherwise (Euler’s theorem). Besides, for energetic reasons, every pentagon has to be surrounded by hexagons (Isolated Pentagon Rule). The smallest sphere fulfilling these conditions is \( C_{60} \). This molecule, consisting of sixty carbon atoms, forms a perfect soccer ball structure. \( C_{60} \) is also known as Buckminster Fullerene, named after the architect Richard Buckminster Fuller, who designed geodetic domes. Next to \( C_{60} \), larger fullerenes are also possible. Most known and common are \( C_{70} \) and \( C_{84} \). Because the properties of these fullerenes differ strongly, their areas of application can vary.

Fig. 1: Carbon allotropes. From left to right: diamond, graphite and fullerene.

Fullerenes posses some unique properties, rendering them suitable for many applications varying from medicine to organic electronics. Fullerenes have the exceptional capability of being very good electron acceptors and conductors. It is these properties that have drawn most attention of scientists. Fullerenes are nowadays mainly used in organic solar cells, transistors, and holographic materials. Since fullerenes are highly insoluble they need to be functionalized prior to being used for any application.

This thesis describes the application of fullerenes in different types of organic electronics. By means of chemical modification the fullerene properties were adapted to fit the desired application.
In chapter two as well as chapter three, new fullerenes are presented for use in solar cells. Organic solar cells consist of two components: a light absorbing polymer, which also conducts positive charges, and fullerenes which accept electrons and subsequently conduct electrons. The operating principle of an organic solar cell is as follows: A polymer absorbs sunlight. The energy of this light causes an electron of the polymer to be excited to a higher energetic state. If an electron accepting material (fullerene) is near, this electron can jump to this material. A hole, or positive charge, remains on the polymer, while the electron accepting material is negatively charged. Both charges are now separated. When electrodes are connected to the material, the positive charge can travel to one electrode and the negative charge to the other electrode (see figure 2). We have a current now! Major advantages of organic solar cells are, for example: cheap production, flexibility of the material and environmentally friendly construction.

Fig. 2: Operating principle of a solar cell. Light is absorbed by the polymer forming an exciton (bound + and – charge). At the interface, of the polymer and the fullerene, charges are separated. The charges then move to the electrodes.

The amount of light that is absorbed, is essential for the efficiency of the solar cell. Normally, fullerenes do not absorb much sunlight. Chapter two describes how a modified, larger, C_{84} fullerene (see figure 3) can be used to absorb more light. Although more light is indeed absorbed when the synthesized C_{84} derivative is used in stead of its C_{60} analogue, the solar cell is less efficient. The cause of this low efficiency is the dramatic drop in voltage. Besides, it seems that the conduction
of positive charges through the polymer does not increase when mixed with the fullerene, while this is the case when the $C_{60}$ equivalent is used.

In chapter three, the synthesis of a series of fullerenes, based on $C_{60}$, is described. We attempted to alter the electron accepting properties of the fullerenes. This was accomplished by placing electron donating and electron pushing groups on the fullerene (see figure 3). We succeeded in inducing small changes. The effect of these changes on the application of these fullerenes in organic solar cells was studied. The results of the solar cells indicate that the voltage of the solar cell is directly related to the electron accepting capability of the fullerenes. The voltage of the solar cells increases when the electron accepting capability of the fullerene decreases.

An important different application of fullerenes is as a semiconductor in transistors. Transistors are often used in electronics and can be considered to be a type of switches, which conduct only under certain conditions. The advantages of organic transistors are once more, cheap production costs and flexibility of the material. Normally speaking, transistors conduct either electrons or positive charges. Fullerenes, however, can conduct both charges (ambipolar) and are therefore highly interesting as material for transistors. The biggest challenge is the instability of fullerenes in air. In chapter four, two different attempts are described to achieve air stable transistors with fullerenes. The first method uses the, in chapter two described, $C_{84}$ derivate. The charged form of this molecule is so stable that it does not undergo redox reactions with water and oxygen. We show here, the first, air and light stable, ‘ambipolar’ organic transistors. The second way to generate air-stability is by introducing fluor atoms in the molecule. Fluor atoms are water repellent. The synthesis of a number of fluorine containing fullerenes is described. The transistors fabricated with these fullerenes however, were not stable in air.
The last application of fullerenes that is investigated in this thesis is holograms. Holographic techniques can be used for medical diagnostic purposes. Because skin is transparent for light of certain wavelengths (infrared), light can be reflected by particles underneath the skin. Light, which is only reflected once, can be used to obtain information about the tissue it was reflected from. This light has to be detected however. This is possible with holograms (see figure 4). The holographic film material consists of three components: a polymer (matrix) which absorbs light and conducts positive charges, nonlinear optical chromophores which orientate in an electric field, and 'so-called' sensitizers (fullerenes) which trap negative charges. When a laser beam combines with light reflected from the tissue, an interference pattern is formed. This means that both light beams either enhance or dim each other. Charges are formed, in the polymer of the holographic material, at the point where the beams enhance each other. The sensitizers (fullerenes) accept a negative charge (as in solar cells). Since there are very few sensitizers, the negative charge is trapped and can not move through the material. The positive charges, however, are able to move through the polymer matrix. This 'separation' of positive and negative charges creates an electric field in the material. The chromophores orientate according to this field. When this happens the diffractive index of the material as a whole is changed. This means that light, which falls on the material, is reflected in a different way. This change can be detected and will ultimately give an image of the tissue at a certain depth. Writing, reading and erasing of such a hologram can be achieved at such high speeds that video images can be produced! In chapter five the influence of the electron accepting capability of the fullerenes on the build up speed of the hologram is studied. We show here that the build up speed increases (up to 4x) with better electron accepting fullerenes. Next, we show a second way of making holographic materials. Instead of using a polymer matrix which conducts positive charges, fullerenes can be used as a matrix which conducts negative charges. Both the synthesis of specially designed fullerenes and the holographic tests are described in this chapter.
We show by means of mass spectrometry that giant ring structures are formed, containing up to 18 fullerenes. These structures are the first real examples of pearl-necklace structures.

Fig. 6: Examples of pearl-necklace structures with \( n=1,2,3 \).