The use of static light scattering for the structure analysis of radiosensitive polymer gels: a literature survey

To cite this article: E Vanden Bussche et al 2004 J. Phys.: Conf. Ser. 3 180

View the article online for updates and enhancements.

Related content
- Stability of polymer gel dosimeters
  Y De Deene, A Venning, C Hurley et al.
- Historical overview of gel dosimetry
  Clive Baldock
- An investigation of the chemical stability of a monomer/polymer gel dosimeter
  Y De Deene, P Hanselaer, C De Wagter et al.

Recent citations
- Handbook of Analysis and Pharmaceutical Quality: Nano-Methods
  Katherine M. Tyner
- Advances in polymeric micelles for drug delivery and tumor targeting
  Uttam Kedar et al.
The use of static light scattering for the structure analysis of radiosensitive polymer gels: a literature survey

E Vanden Bussche¹, Y De Deene¹, P Dubruel², K Vergote¹, E Schacht² and C De Wagter¹
¹ Department of Radiotherapy and Nuclear Medicine, Ghent University Hospital, De Pintelaan 185, 9000 Ghent, Belgium
² Polymer Material Research Group, Ghent University, Krijgslaan 281, 9000 Ghent, Belgium
E-mail: Eline.VandenBussche@UGent.be

1. Introduction

Since Maryanski et al published their first paper on gel dosimetry [1], a lot of research has been done on the subject. After ten years of research a polymer gel (polyacrylamide gelatine gel) is obtained, that is clinically applicable [2]. The preparation of these gel dosimeters is unfortunately difficult and time consuming. The main reason is the removal of oxygen by bubbling nitrogen through the gel.

The removal of oxygen by use of an anti-oxidant as suggested by Fong et al [3] was first thought to be a solution for the oxygen problem. Until now, none of the applied ‘normoxic’ gels was found clinically applicable. An unpublished internal study showed dose rate dependence, temperature dependence and loss of spatial integrity.

In order to find a radiation-sensitive gel with an optimal composition, one should understand the chemical processes that occur in the gel during irradiation. In order to gain more information about these processes, a structure analysis of the radiation sensitive gels should be done. One of the techniques is static light scattering measurements.

2. Methodology

2.1. Basic principle of static light scattering measurements

Static light scattering the angular dependency of the time-mean-intensity of laser light scattered by the particles is measured. The course of the scattered intensity as a function of the detector angle depends on size and structure of the particles.
Rayleigh developed a theory that predicts the angular intensity distribution of light scattered by particles much smaller than the wavelength of light (R < λ/20). In this case the intensity of the scattered light depends only on the size of the particles and not on their structure or the concentration. If the intensity of the scattered light is measured in a plane orthogonal to the polarization plane of the incident light, there will be no angular distribution of the scattered light.

\[
R_\theta = \frac{16\pi^4 R^6}{\lambda^4} \left( \frac{n^2 - 1}{n^2 + 2} \right)^2
\]

(1)

In this expression, \( R_\theta \) is the Rayleigh ratio, \( R \) is the particle size, \( n \) is the relative refractive index of the solute and \( \lambda \) is the wavelength of the incident light.

Figure 1. Basic principle and setup of static light scattering measurements.

Figure 1. Comparison of the angular dependence of the intensity of scattered light for a spherical particle in the Rayleigh region (--) and a large spherical particle (→).
For larger particles, the influence of the particle structure and its concentration should be brought into consideration. In that case, the scattered light intensity for low particle concentrations can be described by:

\[ R_{ex} = K c M_w P(\theta) \]  

(2)

In this expression, \( R_{ex} \) is the excess Rayleigh ratio, \( K \) is the optical constant, \( c \) is the particle concentration, \( M_w \) is the mean molecular weight of the particles and \( P(\theta) \) is the particle structure factor.

If the particle concentration increases the influence of intraparticle optical interference and interparticle interactions increases. In that case the angular dependency of the scattered light can be described by:

\[ \frac{Kc}{R_{ex}} = \frac{1}{MP(q)} + 2A_2c + ... \]  

(3)

in which, \( A_2 \) is the mean second virial constant.

2.2. Fitting of the data

Zimm [4] discovered that in the limit of low scatter vectors and low concentrations, the angular distribution of the scattered intensity becomes independent of the particle shape. By extrapolation of \((Kc/R_{ex})\) to zero concentration and zero angle the average molecular weight, radius of gyration and second virial constant can be estimated.

The structural parameters can be compared with theoretically derived values for several models such as from random coil, homogeneous sphere.

3. Applicability for gel dosimetry

About the structure of polyacrylamide polymers, there is already some information available [5,6]. N,N’-methylenbisacrylamide (Bis), much more hydrophobic as acrylamide (Ac), has the tendency to form clusters. As the polymerization starts, these clusters form cross-linked regions with a high concentration of Bis. As the concentration of Bis and the cluster formation decreases during polymerization less cross-linked side chains are formed that link the different microgel particles.

As a consequence, two phases will exist in the structure: a dilute phase constituted by the side chains and a dense phase constituted by the Bis-clusters. The dense phase will contribute more to the light scattering as the dilute phase.

The separation of the polyacrylamide particles and the gelatin matrix is very difficult. This can be ascribed to the high crosslinked structures of polyacrylamide in which several gelatin molecules are caught. That is why we plan to perform the structure analysis of the polyacrylamide particles inside the gelatin medium. Tian et al [7] investigated the possibility of performing static light scattering measurements with particles embedded in a hydrogel. They came to the conclusion that all parameters could be estimated correctly, except for the second virial constant.
4. Conclusion

Static light scattering (SLS) could be a worthy technique to perform a structure analysis of the polymer structures inside radiation sensitive gels. The information obtained with SLS is a static characterization of the particle structures inside the gel. SLS will be combined with NMR relaxometry and NMR diffusion measurements, which deliver a hydrodynamic characterization of the microstructure of the gels.

Acknowledgement

This study was supported by IWT funds through the specialization fund IWT-SB/31223.

References

[1] Maryanski M, Gore J, Kennan R and Schulz R 1993 NMR relaxation enhancement in gels polymerized and cross-linked by ionizing radiation: a new approach to 3D dosimetry by MRI Magn. Reson. Imaging 11 253–8

[2] Vergote K, De Deene Y, Duthoy W, De Gersem W, De Neve W, Achten E and De Wagter C 2004 Validation and application of polymer gel dosimetry for the dose verification of an intensity-modulated arc therapy (IMAT) treatment Phys. Med. Biol. 49 287–305

[3] Fong P, Keil D, Does M and Gore J 2001 Polymer gels for magnetic resonance imaging of radiation dose distributions at normal room atmosphere Phys. Med. Biol. 46 3105–13

[4] Zimm B 1948 The scattering of light and the radial distribution function of high polymer solutions J. Chem. Phys. 16 1093–9

[5] Baselga J, Llorente M, Hernández-Fuentes I and Piérola I 1989 Network defects in polyacrylamide gels Eur. Polym. J. 25 471–5

[6] Baselga J, Llorente M, Hernández-Fuentes I and Piérola I 1989 Polyacrylamide gels: process of network formation Eur. Polym. J. 25 477–80

[7] Tian M, Tuzar Z and Munk P 1995 Light scattering of particles embedded in soft hydrogel Collect. Czech. Chem. C. 60 1719–27