



# Workshop on Solid State Dosimetry in Neutron and Ion Radiation Fields

**28 June – 29 June 2012**

**Johannes Gutenberg-University  
Mainz, Germany**

## **Aims**

- Exchange of experience: Detectors in mixed radiation fields
- New approaches: ESR-dosimeter and radiochromic materials
- Discussion: The perfect dosimeter for mixed radiation fields



## Workshop Abstract

Solid state detectors are widely used for medical dosimetry in neutron and ion beams, however often large correction factors must be applied which depend on the nature of the radiation field. Detector response models are all phenomenological, the exact details of how they function remain unclear. Alanine is established as a very reliable dosimeter for mixed radiation fields, however shows poor sensitivity at medical dose levels and is practically insensitive to neutrons. Adding elements with high thermal neutron cross section could increase sensitivity but may compromise radical stability and tissue equivalence. In the first part of the workshop we will present results obtained with alanine in mixed neutron and ion fields, summarize the current knowledge about alanine and related radical chemistry, and discuss possibilities to enhance neutron sensitivity.

The second part of the workshop we will take a closer look on the latest research in radiochromic detectors based on leucomalachite green dye. The goal is to find ways of increasing the sensitivity and stability of a specific research detector system which will be presented at the workshop.

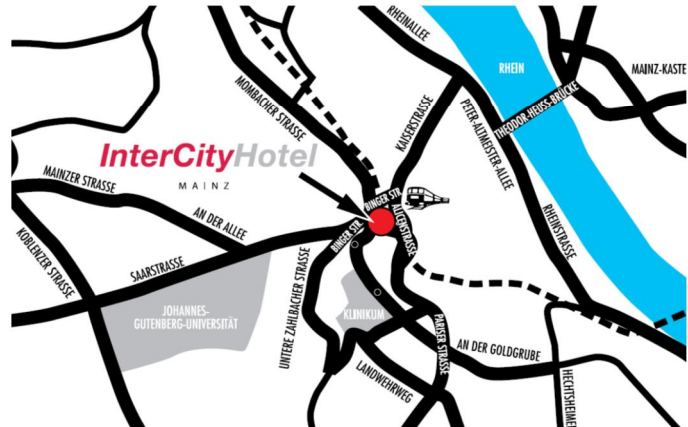
Understanding of the underlying radical chemistry of these detectors could be the key to develop less phenomenological detector response models pursuing the idea of an “ideal” dosimeter for mixed radiation fields.

## Accommodation

InterCityHotel Mainz  
Bingener Strasse 21  
55131 Mainz

## Workshop Venue

Institut für Physikalische Chemie  
Johann-Joachim-Becher Weg 14  
55128 Mainz

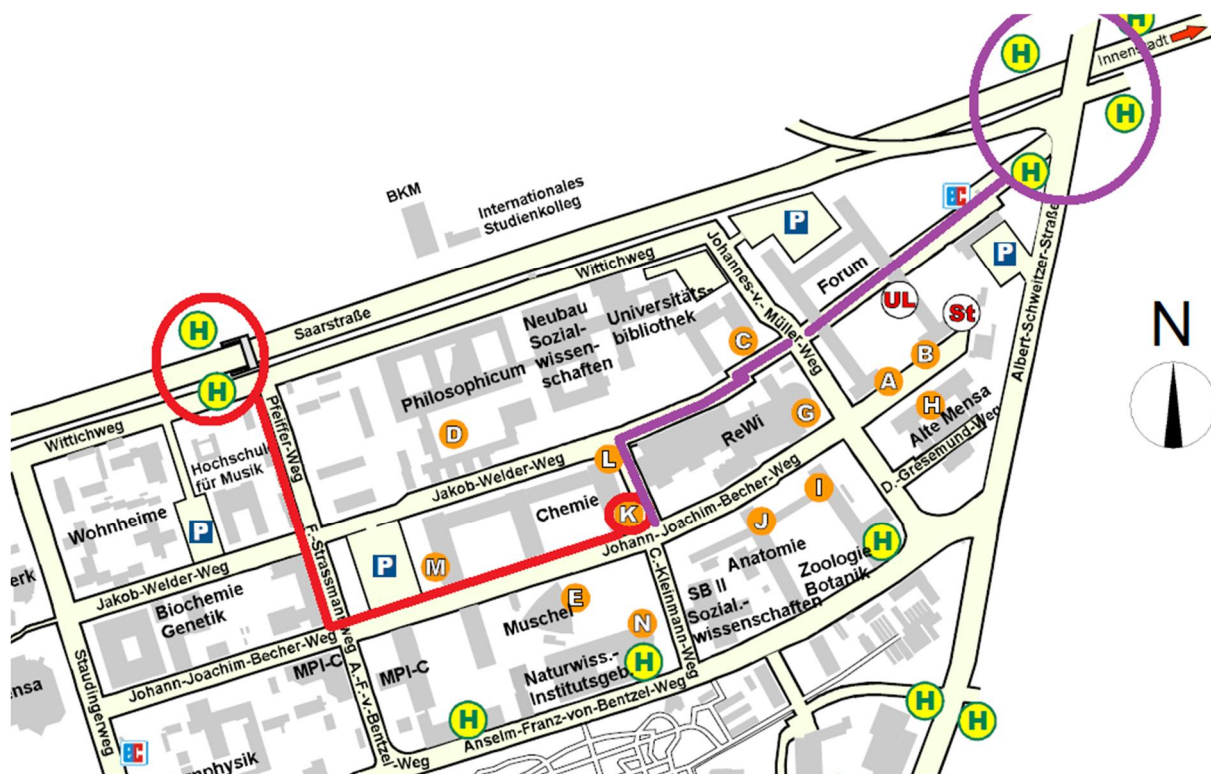


To get from the Intercity Hotel or the mainstation to the workshop site (K on the map) it is possible to use the busstop “Hauptbahnhof West”, which is in front of the Hotel and at the rear exit of the station.

With busses 54, 55, 58, 68 it is two stops to “Friedrich von Pfeiffer Weg”, marked red in the map.

Alternatively it is possible to use busses 6, 6A, 56, 57, 69 one stop to “Universität”, marked purple in the map .

Please note that it is only possible to use the entrance on Johann-Joachim-Becher Weg. The workshop room is on the second floor.





## Programme

### Thu 28 June

- 10:30 – 10:45 Welcome address
- 10:45 – 12:30 Session: EPR Detectors
1. Tobias Schmitz  
*"The Alanine Detector in Neutron Fields"*
  2. Hugo Palmans  
*"NPL's Alanine Dosimeter applied to Particle Beams"*
  3. Eirik Malinen  
*"EPR dosimetry using lithium formate"*
- 12:30 – 13:30 Lunch
- 13:30 – 15:00 Guided Tour: The TRIGA Mark II research reactor of the University of Mainz
- 15:00 – 16:30 Session: Monte Carlo Modelling
1. Matthias Blaickner / Markus Ziegner  
*"The impact of different source definition techniques on the simulated irradiation field of the TRIGA Mainz"*
  2. Rochus Herrmann  
*"Prediction of the Response Behaviour of One-Hit Detectors in Particle Beams"*
- 16:30 – 17:30 Session: Radiochromic Detectors
1. Peter Sandegaard Skyt  
*"Exploring the dose response of a radiochromic hydrogel dosimeter"*
- 19:00 Dinner at Weingut Menk in Ingelheim (Transport will be organised from the Hotel)

### Fri 29 June

- 09:00 – 10:30 Session: Chemistry
1. Carl Schiesser  
*"Free Radical Chemistry and Biotechnology: Fundamental Principles and Applications"*
  2. Steve Bottle  
*"Using Fluorescence and Nitroxides to Detect Free Radicals: Applications from Materials Science to Medicine"*
  3. Linda Feketeová  
*"Gas-phase chemistry using mass spectrometry"*
- 10:30 – 12:00 Discussion  
*"The perfect dosimeter for mixed radiation fields"*
- 12:00 Closure of the workshop and lunch

## Abstracts

Thu 28 June – Session: EPR Detectors

### The Response of Alanine Detectors in Thermal Neutron Fields

T. Schmitz<sup>1</sup>, N. Bassler<sup>2</sup>, P. Sharpe<sup>3</sup>, H. Palmans<sup>3</sup>, M. Blaickner<sup>4</sup>, M. Ziegner<sup>4,5</sup>, P. Langguth<sup>6</sup>,  
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Irradiation fields in BNCT are mixed neutron and gamma fields. In most facilities dosimetry is done by neutron flux monitoring and ion chambers [1]. At the TRIGA Mark II research reactor at the University of Mainz, Germany, the possibilities of alanine ESR detectors have been investigated. Ionizing particles generate radicals in the detector, which can be detected using Electron Spin Resonance (ESR) spectroscopy.

All alanine pellets used, have been manufactured and read out at the National Physical Laboratory (NPL), United Kingdom [3]. Alanine has been irradiated in four different experimental conditions at the TRIGA Mark II research reactor Mainz [2]. In the used irradiation position a predominantly thermal neutron field with a strong gamma component is delivered. In the experiments a Polymethylmethacrylate (PMMA) phantom and a Teflon phantom have been used. Further pellets have been irradiated with a lead/bismuth gamma shield and a boric acid neutron shield. To predict the dose and its components for each pellet, the Hansen & Olsen Model [4] together with the Monte Carlo Code FLUKA [5] has been used.

The measured dose response of all pellets will be shown and compared to earlier measurements in liver tissue [6]. Also it will be shown that the dose response could be reproduced by the calculations.

For all experiments three dose components have been separated in the calculations. A proton dose is generated in the  $^{14}\text{N}(n,p)^{14}\text{C}$  reaction and secondary gammas are generated by various  $(n,\gamma)$  reactions. The primary gamma dose is deposited by gammas from the reactor core. Using the Hansen & Olsen Model the absorbed dose for the pellets could be determined by means of the measured dose response. Therefore relative efficiencies have been calculated for all primary and secondary particles which account for different radical yields for different particle types and energies.

Alanine dosimeters are suitable for measurements in mixed neutron and gamma fields and the alanine response can be fully understood by the used interpretation model. In further experiments in other facilities the dosimeters will be exposed to higher neutron energies which are more common for BNCT treatments.

## References:

[1] Rogus RD et al, Medical Physics 21, 1611-1626, 1994. [2] Hampel G et al, Applied Radiation and Isotopes 67, 238-241, 2009. [3] Sharpe P and Sephtan J, Applied Radiation and Isotopes 52, 1185-1188, 2000. [4] Hansen JW and Olsen KJ, Radiation Research 104, 15-27, 1985. [5] Battistoni G et al, Proceedings of the Hadronic Shower Simulation Workshop 2006; AIP Conference Proceedings 896, 31-49, 2007.



## LET-signatures in lithium formate EPR dosimeters following ion irradiation

Eirik Malinen<sup>1,2</sup>, Einar Waldeland<sup>2</sup>, André Krivokapic<sup>1</sup>, Eli Olaug Hole<sup>1</sup> and Einar Sagstuen<sup>1</sup>

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<sup>2</sup>*Department of Medical Physics, Oslo University Hospital, Oslo, Norway*

Polycrystalline lithium formate monohydrate is a material suitable for EPR (Electron Paramagnetic Resonance) dosimetry at dose levels ranging from about 0.1 to at least 1000 Gy. Irradiation with densely ionizing ions causes subtle changes in the EPR spectrum of lithium formate. We have shown that the EPR line width increases with increasing LET, where irradiation with e.g. 500 MeV nitrogen ions results in a 6 % increase in line width compared to irradiation with photons. This is caused by increased dipolar spin-spin interactions due to elevated local radical density following high-LET irradiation. This phenomenon is also expressed as less pronounced microwave power saturation for samples exposed to high-LET radiation. Non-linear model analysis of such power saturation properties has shown that the spin-spin relaxation time is reduced for such ion-irradiated samples. The talk will give an introduction to EPR and methods for assessing LET-signatures in EPR spectra. Li-6 enrichment of lithium formate dosimeters for more sensitive neutron dosimetry will be discussed. Also, relevant results from the recent ACQUARATE experiment for lithium formate and alanine will be included.

Thu 28 June – Session: Monte Carlo Modelling

## The impact of different source definition techniques on the simulated irradiation field of the TRIGA Mainz

M. Blaickner<sup>1</sup>, M. Ziegner<sup>1,2</sup>, R. Khan<sup>2</sup>, T. Schmitz<sup>3</sup>, G. Hampel<sup>3</sup>, H. Böck<sup>2</sup>

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### Introduction

In order to build up a reliable dose monitoring system for Boron Neutron Capture Therapy (BNCT) applications at the TRIGA reactor in Mainz two different source definition techniques have been compared. A source plane situated in the thermal column and determined with the ATTILA software some years ago [1] was evaluated against a reactor criticality calculation with MCNP. The techniques were compared with regard to their neutron and photon spectra in the thermal column as well as to their accordance with flux and dose measurements.

### Materials and Methods

The ORIGEN code was used to incorporate the complete burnup history of the TRIGA Mainz from 1967 to 2011. The resulting core configuration was implemented into the Monte Carlo code MCNP5 and a criticality calculation was performed, expanding the neutron and photon transport from the reactor core to the thermal column. This way the spectra and source strengths for both particles were calculated at the exact same location as the already existing source plane determined by ATTILA.



Furthermore an experimental irradiation setup was implemented into the same MCNP simulation, namely a polymethyl-methacrylate (PMMA) phantom in a distance of 92 cm from the core, partially shielded by bismuth and containing gold foils for flux determination and alanine dosimeters for dose measurements [2]. In order to correlate the absorbed dose within the alanine pellets with the measured dose response a relative effectiveness (RE) factor of 0.55 was used for the dose component caused by neutrons and 1 for photons respectively.

The same irradiation setup was implemented into another, much more simplified MCNP model, containing only part of the thermal column and using the already existing ATTILA source plane as input. Both simulations were compared with the experimentally measured flux and the dose response of the alanine dosimeters.

## Results

The expanded MCNP criticality calculation yielded a neutron spectrum and a source strength which is in very good agreement with the data previously determined by ATTILA. However with regard to photons the source strengths determined in the criticality calculation turns out to be lower by a factor no less than 4.4 and the spectrum shows big deviations for energies below 100 keV.

Both simulation models show a very good agreement with the neutron flux measured by gold foil activation. The mean deviation is only 1.6% for the simplified model and 4.6% for the criticality calculation.

The alanine response predicted by the simplified model exceeds the values of the measurements on an average by ~27%, however this deviation hardly changes with regard to the individual alanine pellets which are situated at different positions. The criticality calculation by contrast shows a much better accordance, decreasing the mean overestimation to ~10%, reaching as low as 2% for some individual pellet. The observed differences are completely owed to the deviations of the photon contribution since the neutron component for both approaches is virtually the same.

## Discussion

The very good agreement between both simulation models and measured neutron flux underlines the solidness of the neutron transport in both approaches.

The considerable decrease of the photon flux for the expanded criticality calculation is confirmed by the results of the alanine response where the same observation causes the much better agreement with the measured data. Consequently the expanded criticality calculation should form the basis for the definition of a new source plane and therewith future dosimetry calculations.

## References

- [1] B. Wortmann, "Auslegung und Optimierung einer Bestrahlungsanlage für die Bor-Neutronen-Einfangtherapie an austransplantierten Organen.," Technische Universität Dresden, Germany, 2008.
- [2] T. Schmitz et al., "Dose determination using alanine detectors in a mixed neutron and gamma field for boron neutron capture therapy of liver malignancies.," Acta oncologica (Stockholm, Sweden), vol. 50, no. 6, pp. 817-22, Aug. 2011.

Thu 28 June – Session: Radiochromic Detectors

### Exploring the dose response of a radiochromic hydrogel dosimeter

Peter Sandegaard Skyt<sup>1,2</sup>, Isak Wahlstedt<sup>2</sup>, Ludvig Paul Muren<sup>1</sup>,  
Jørgen Breede Baltzer Petersen<sup>1</sup> and Peter Balling<sup>2</sup>

<sup>1</sup>*Dept of Medical Physics, Aarhus University Hospital, Denmark*

<sup>2</sup>*Dept of Physics and Astronomy, Aarhus University, Denmark*

#### Purpose:

Radiochromic dosimeters together with optical dose read-out methods are new and promising methods within three-dimensional dosimetry. The most common radiochromic dosimeters are based on the uncolored leuco malachite green dye, LMG, that react to the colored malachite green when irradiated. A promising dosimeter is the commercially available Presage dosimeter, however, the post-irradiation stability considerably depend on both temperature and dose level. A similar but non-commercial dosimeter has been suggested (Vandecasteele et al, Phys Med Biol 2011) introducing the possibilities of improving the dosimeter by changing the chemical composition. The dose response of this dosimeter, however, is low and in our study we have therefore investigated a range of chemical compositions in order to improve the dose response.

#### Methods:

The radiochromic dosimeter is based on LMG dissolved in chloroform ( $\text{CHCl}_3$ ) with trichloroacetic acid ( $\text{CCl}_3\text{COOH}$ ) as initiator. To obtain a volume dosimeter these components were mixed in a gel consisting of gelatin and water. The non-polar LMG was dissolved in the water solution by adding sodium dodecyl sulfate (SDS) which encapsulates LMG in micelles. To improve the dose response, a range of compositions have been investigated by changing the chemical concentration of the original dosimeter, changing the pH value, adding NaCl to vary the micelle size and changing the initiator.

#### Results:

The average dose response of the original dosimeter was measured to an optical density of  $(3.8 \pm 0.7) \cdot 10^{-3} \text{ cm}^{-1} \text{ Gy}^{-1}$ , but with a high inter-batch variation. Changing the concentration of the components of the original dosimeter did not improve the dose response or resulted in a high auto oxidation rate or non-transparent dosimeters. Since the dosimeters are read-out with optical methods the latter is not preferable. At low pH values and when adding NaCl, non-transparent dosimeters were produced. Changing the initiator gave low or similar dose response as the original dosimeter.

#### Conclusion:

The dosimeter showed a low dose response with a high inter-batch variation. Varying the chemical composition of the dosimeter did not improve the dose response or produced non-transparent dosimeters.



Fri 29 June – Session: Chemistry



**Free Radical Chemistry and Biotechnology:**  
Fundamental Principles and Applications



**Carl H. Schiesser**

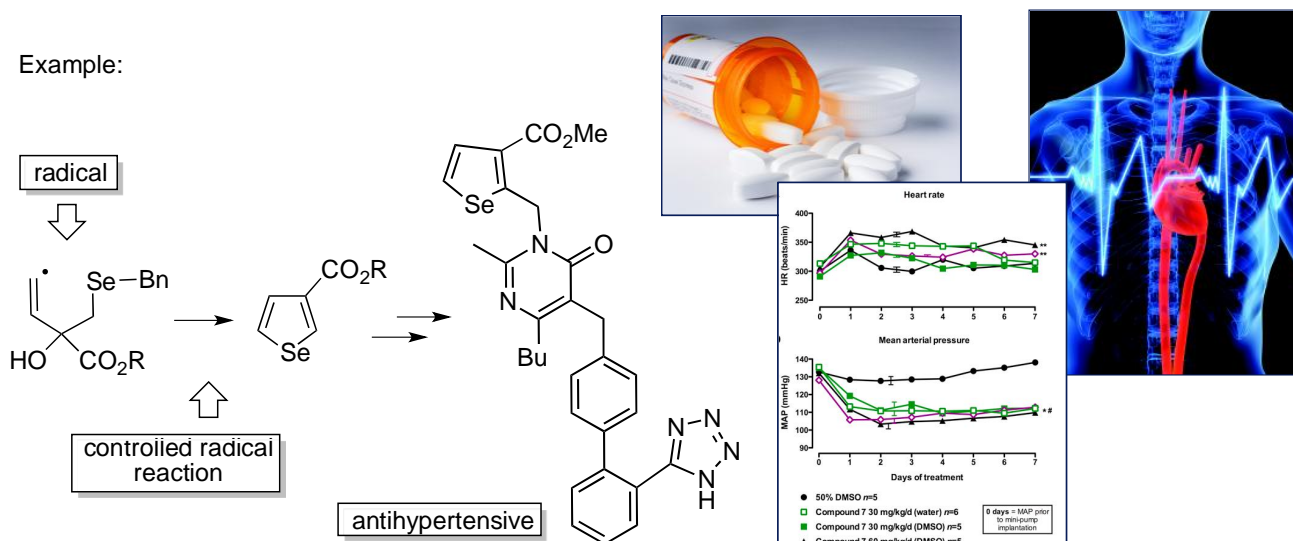
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There was a time when free radicals were scorned by organic chemists and when “practically every organic text book written” contained a statement that free radicals were “incapable of an independent existence”.<sup>1</sup> Except for polymer chemistry, these reactive species were mostly regarded as poorly-understood curiosities, often scape-goats for unwanted outcomes during synthesis, or when the practitioner required that elusive explanation for his or her unwanted observation. Those were the *Dark Ages* of free radical chemistry, the lengthy period between the “discovery” of organic free radicals by Gomberg in 1900 and their resurgence some seventy or so years later.<sup>2,3</sup>

The *Dark Ages*, of course, were interspersed with significant contributions by key research groups, but free radicals remained largely inaccessible to synthetic chemists until their *Renaissance* during the period 1970 – 1990 in which the factors that control the reactivity, regiochemistry and stereochemistry of radical reactions began to be teased out. The dramatic rise in the understanding of free radical chemistry during this period led to their general acceptance in the wider chemistry community.<sup>4-6</sup> Free radicals can now be harnessed for the preparation of important materials that range from pharmaceutical products through to polymers and an understanding of their mechanism of action has led to better methods for the protection of biomolecules and other materials from oxidative-stress related damage that are often associated with free radicals.

This presentation will cover some of the fundamental principles that govern the reactivity and stability of organic free radicals and will culminate with some examples of their utility.

Example:



## References:

1. Rice, F. O.; Rice, K. K., *The Aliphatic Free Radicals*, Johns Hopkins Press, Baltimore, **1935**, and refs. cited therein.
2. Gomberg, M., *J. Am. Chem. Soc.* **1900**, 22, 757.
3. See foreword in: Renaud, P.; Sibi, M. P., *Radicals in Organic Synthesis Vols. 1 and 2*, Wiley-VCH, Weinheim, 2001.
4. Beckwith, A. L. J.; Schiesser, C. H., *Org. Biomol. Chem.* **2011**, 9, 1736.
5. Beckwith, A. L. J.; Schiesser, C. H. *Tetrahedron*, **1985**, 41, 3925.
6. Spellmeyer, D. C.; Houk, K. N. *J. Org. Chem.*, **1987**, 52, 959.

## Using Fluorescence and Nitroxides to Detect Free Radicals:

Applications from Materials Science to Medicine



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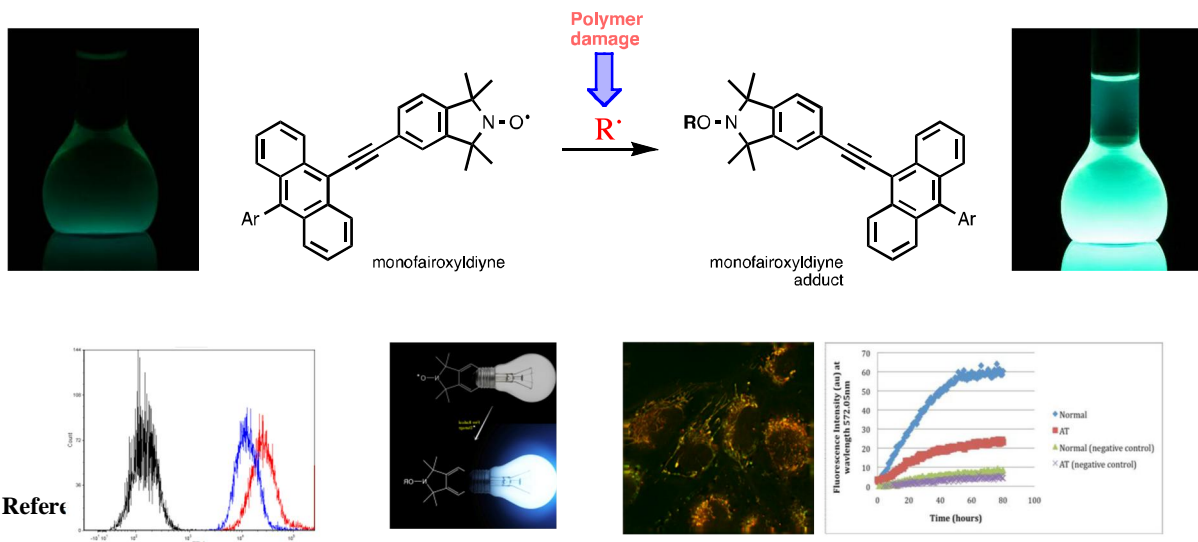
Steven E. Bottle



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Nitroxides are powerful antioxidants and scavengers of free radicals that have been widely used since the 70's as probes for reactive species and as stabilizers for polymers. Most nitroxide research and applications have involved the six-membered TEMPO class, largely because of commercial availability. We have focused our efforts on fused aryl nitroxide analogues of the isoindoline class, as these can possess some advantages over other nitroxides<sup>1</sup>. Significantly when the nitroxide group is proximate to an extended aryl fluorophore, excited state quenching and energy transfer short-circuits the normal fluorescence process. We have synthesized a range of novel nitroxides, including water soluble analogues that have found applications probing the free radical basis of oxidative diseases<sup>2</sup>, as sensors for oxidative damage in materials<sup>3</sup> and even for monitoring pollution from ultra-fine particles<sup>4</sup>. Our novel analytical approach employs nitroxides that contain potent, yet masked, fluorophores that are built in to the carbon skeleton of the molecule. These systems allow the monitoring of radical reactions according to the fluorescence detected upon their conversion to the diamagnetic scavenging products<sup>5</sup>. When the nitroxide captures a free radical to form the stable alkoxyamine, the natural fluorescent nature of these compounds is no longer suppressed and over a 100-fold increase in fluorescence can be detected compared to the nitroxide parent. We have described these nitroxide-fluorophore systems as *profluorescent* nitroxides (PFN) and they represent a powerful new methodology for monitoring free radical reactions<sup>6</sup>. This talk will describe some of the synthetic routes used to develop these systems and outline some of the current applications of these interesting compounds.



1. H-Y Ahn, K.E. Fairfull-Smith, B.J. Morrow, V Lussini, B. Kim, M.V. Bondar, S.E. Bottle, K.D., Belfield, *Journal of the American Chemical Society* (2012), 134(10), 4721-4730.
2. B.J. Morrow, D.J. Keddie, N. Gueven, M.F. Lavin, S.E. Bottle, *Free Radical Biology & Medicine* (2010), 49(1), 67-76.
3. J.M. Colwell, J.R. Walker, J.P. Blinco, A.S. Micallef, G.A. George, S.E. Bottle, *Polym. Deg. Stab.* (2010), 95(10), 2101-2109
4. B. Miljevic, M.F. Heringa, A. Keller, S.E. Bottle et al. *Environmental Science & Technology* (2010), 44(17), 6601-6607
5. K.E. Fairfull-Smith, J.P. Blinco, D.J. Keddie, G.A. George and S.E. Bottle, *Macromolecules*, (2008) 41(5), 1577-1580.
6. J.B. Blinco, K.E. Fairfull-Smith, B.J. Morrow, S.E. Bottle, *Australian Journal of Chemistry* (2011), 64(4), 373-389.

## Gas-phase chemistry in mass spectrometry

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Mass spectrometry has expanded in recent years into many different areas of research. It became a toolbox for uncovering the intrinsic properties of molecules. The mass spectrometer we use at the University of Melbourne has an electrospray ionization source to form ions and a linear ion trap as a mass analyzer, where ions can be trapped for up to 10 seconds and their fragmentations, through vibrational excitation, as well as chemical reactions can be investigated. A Fourier transform ion cyclotron resonance cell is coupled to the ion-trap for high-mass resolution and to study the fragmentation of ions by free electrons. The basic principles of mass spectrometry will be presented, together with an example of the formation of positive and negative ions of the amino acid tryptophan and their fragmentation through vibrational and electronic excitation.

Acknowledgements: George Khairallah, Richard O'Hair

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## Thursday evening: Weingut Menk – Ingelheim

The first day will be concluded with a nice vine tasting and dinner at Weingut Menk in Ingelheim, a small city close to Mainz.

Surrounded by hiking trails Weingut Menk is located at one of the highest points of Ingelheim. The family enterprise provides a beautiful look on the castles of the Rhein valley and the cities Bingen and Mainz.



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## A little tourist guide for the city centre of Mainz

Located directly on the 50th parallel north, Mainz and his 2000 year history has been many things to many people:

### *St. Martin's Cathedral*

"This cathedral above the Rhine Valley with all of its might and glory would have remained in my memory even if I had never seen it again", wrote author Anna Seghers. This tremendous cathedral is a key feature of the cityscape even today, thousand years after it was built.



### *St. Stephan - Chagall Window*

A total of 200,000 visitors a year prove that St. Stephan is an attraction! Tourists from around the world make the hike up to the Stephansberg to see Marc Chagall's sparkling blue windows.

### *The Old Town*

Attractive squares, beautifully restored half-timbered houses and magnificent Baroque churches give the Old Town its charming character. Behind Rococo facades and in fine Baroque houses you will find elegant boutiques, cafés and wine taverns.



### *Gutenberg Museum*

Today you can experience four thousand years of the history of writing from around the world in the Gutenberg Museum. Johannes Gutenberg of Mainz played one of the key roles in the process when he invented printing with moveable type and the printing press.

### *The Mainz of Roman Times*

Mogontiacum, the Mainz of Roman times, was an important place in antiquity and developed from a military into a civil centre of the region.



## Coordinators

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