

^3H speciation in bioshield concrete

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Background to this study

- **PhD study supervised by Ian Croudace & Phil Warwick**
- **Funded by KOREAN Government**
- **PhD Title “*Optimisation of tritium determinations for nuclear decommissioning materials*”**
- **Effect of sampling & sample storage on ^3H data from diverse materials (e.g. concretes, metals, graphite, plastics)**

Contents

- **Introduction-** Tritium in bioshield concrete
- **Methodology-** Combustion & LSC
- **Results-** ^3H distribution & speciation in bioshield core
- **Discussions-** ^3H in structural & bioshield concrete
- **Conclusions-** Analytical implications

Significance of concrete

- **Concrete mainly consist of a mixture of**
 - Portland cement, sand, water, rock aggregates
- **Constitutes a large volume of waste (NIREX, 2002)**
 - Concrete, cement & rubble consist of
21% of LLW
12% of ILW
- **Various usages in the nuclear industry due to (R.C. Hochel & E.A. Clark)**
 - Strong
 - Durable
 - Inexpensive
 - Shield radiation well

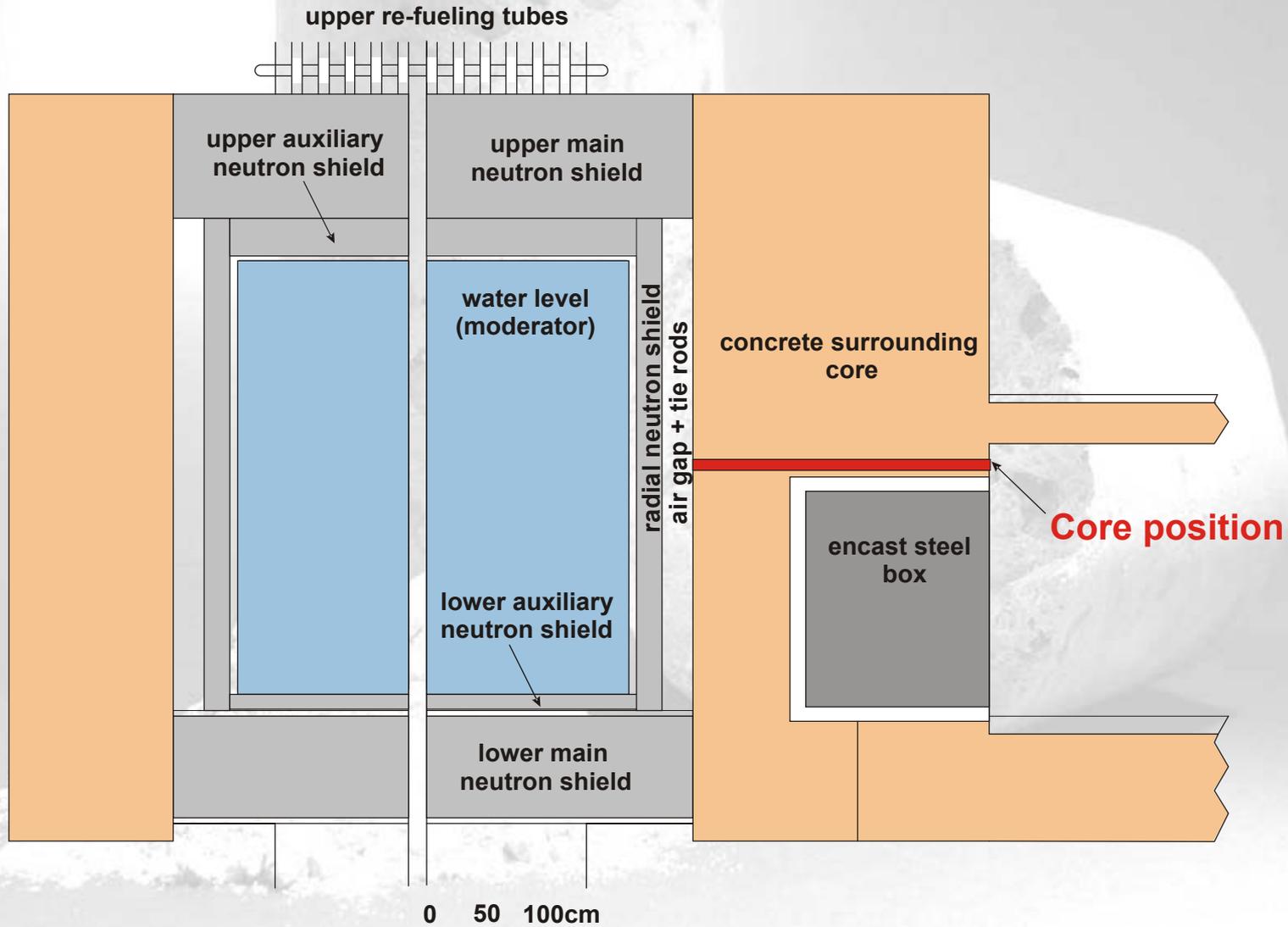
(e.g. Building structure material & reactor shielding material)

Glossary

- **What is bioshield concrete?**
 - **Reactor shielding concrete**
- **What is structural concrete?**
 - **Typical building material except for neutron shielding in reactor**

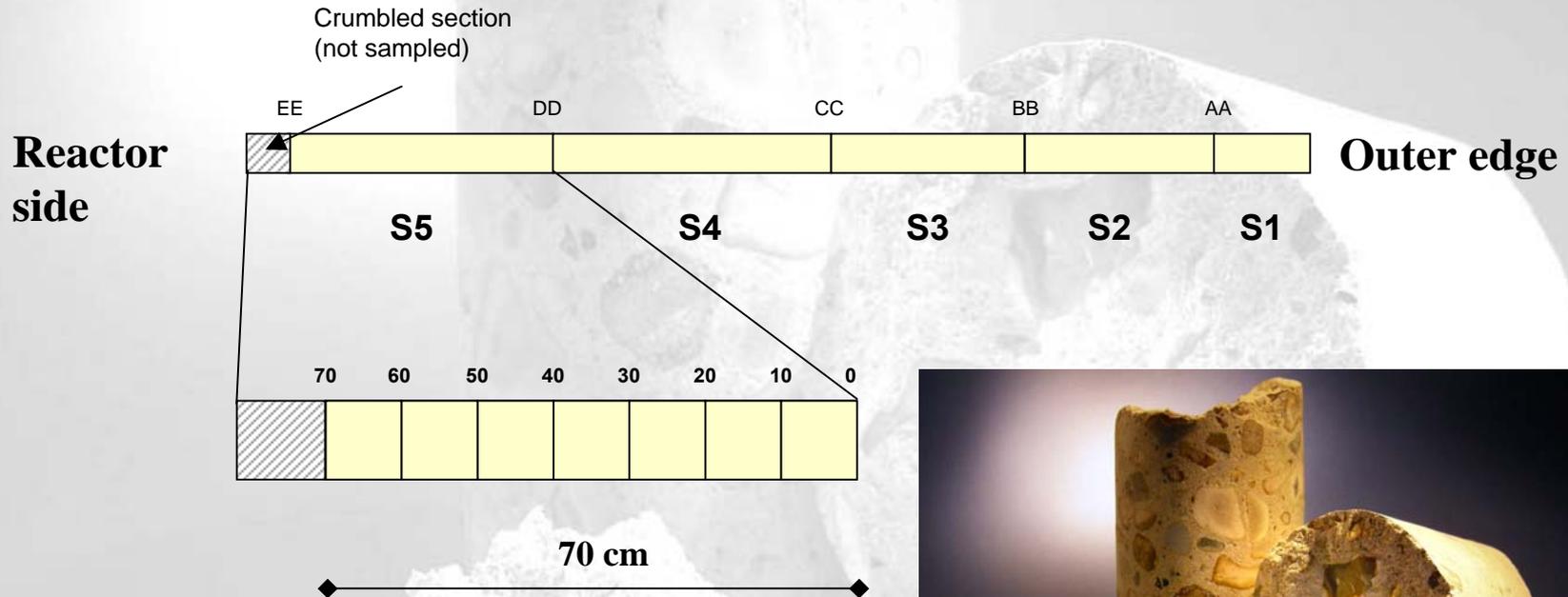
Introduction

Location of the bioshield core



Introduction

Sectioning schematic for concrete core



Production of tritium in reactor

- Neutron capture by deuterium
- Neutron capture by lithium-6 (n, α)
- Ternary fission

^3H extraction & Measurement



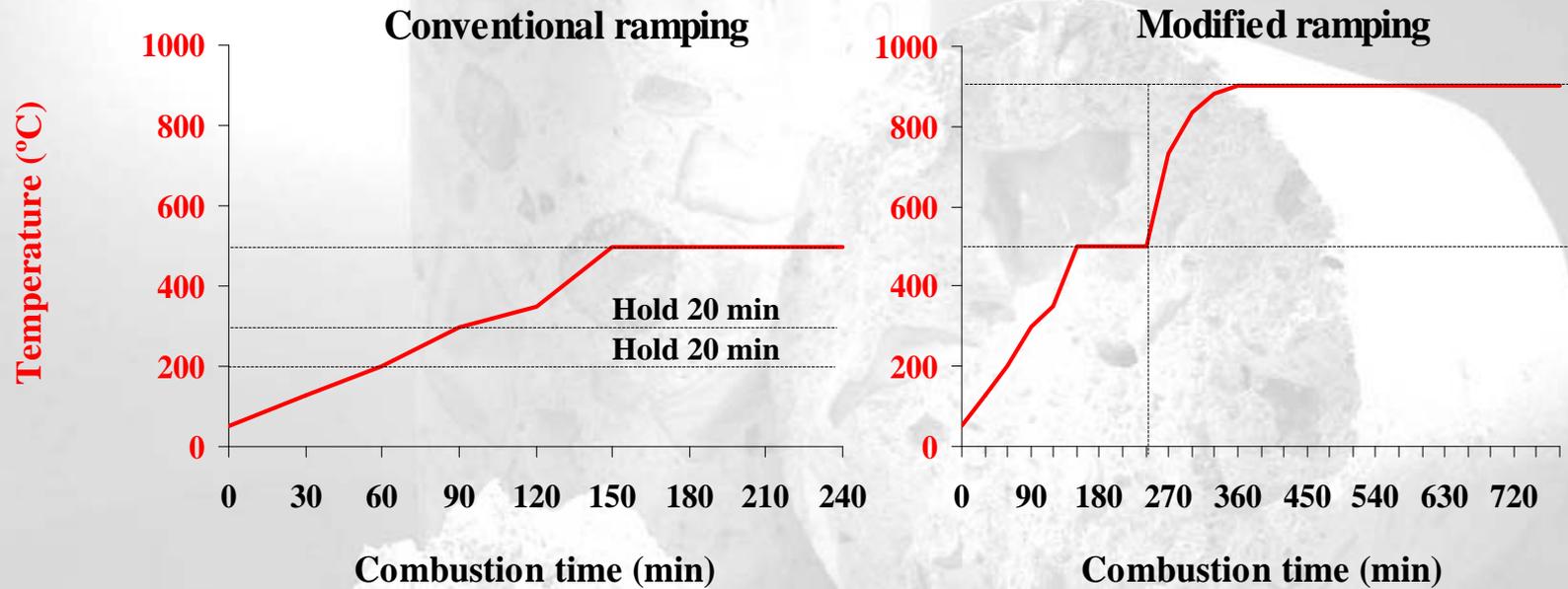
Raddec Pyrolyser TM System



Quantulus Liquid
Scintillation counters

Methodology

Ramping cycle

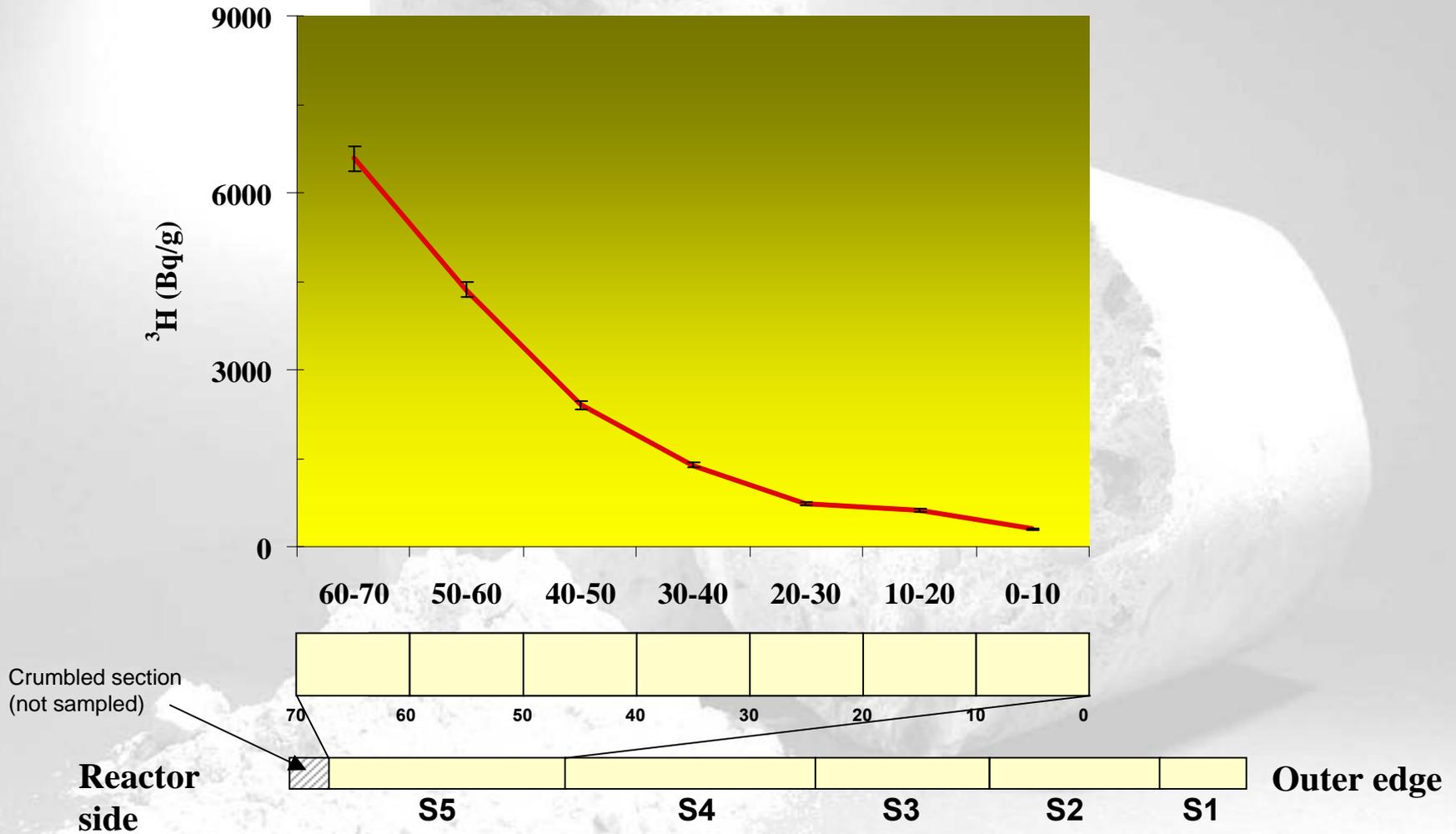


Tritium evolution profile

- **Providing**
 - Information on the form of ^3H
 - Implications to sample preservation and analysis
- **Factors affecting the evolution profile**
 - Origin of the ^3H
 - Form of ^3H in the sample
 - Sample composition

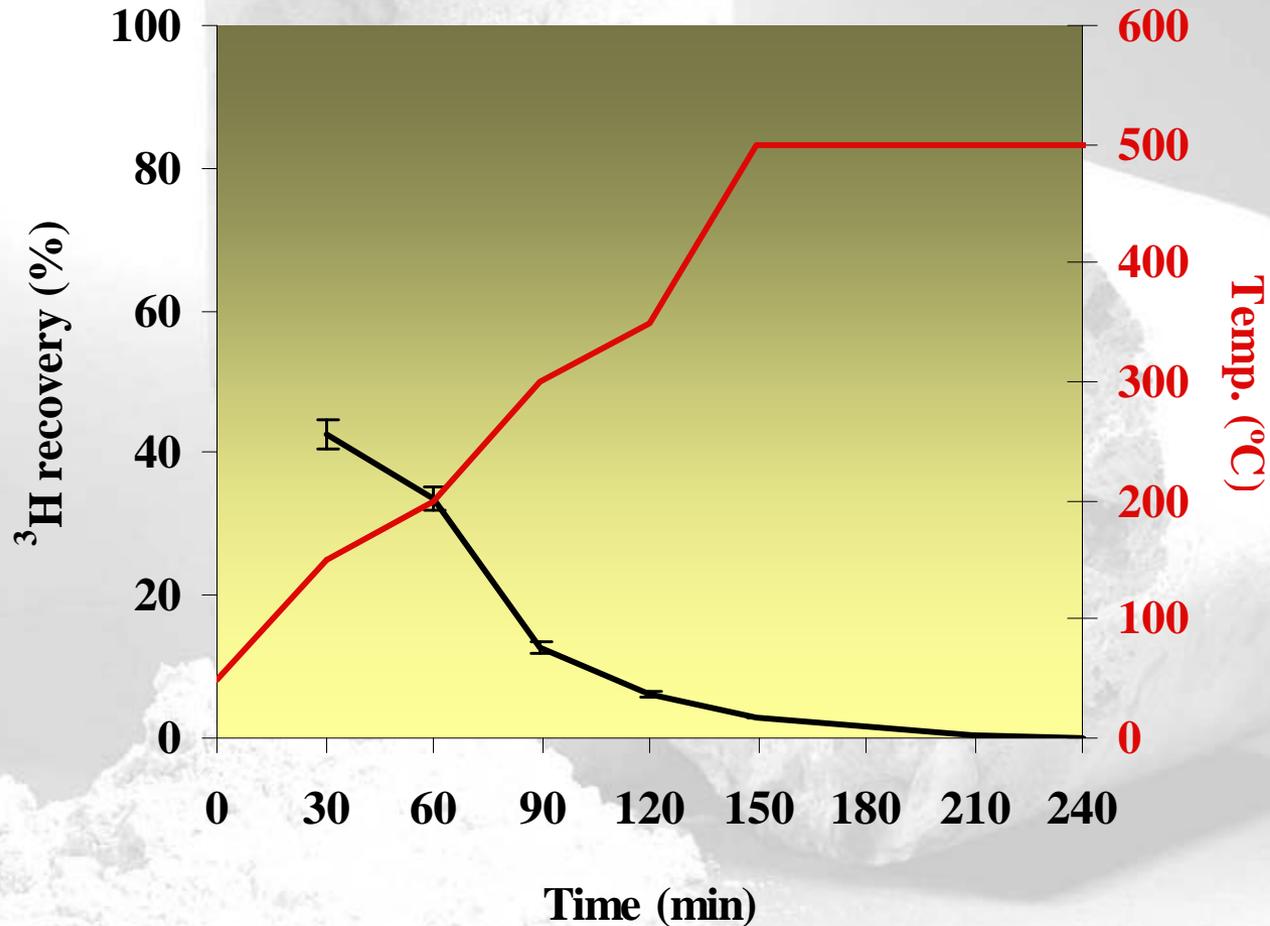
Results

^3H distribution of bioshield core



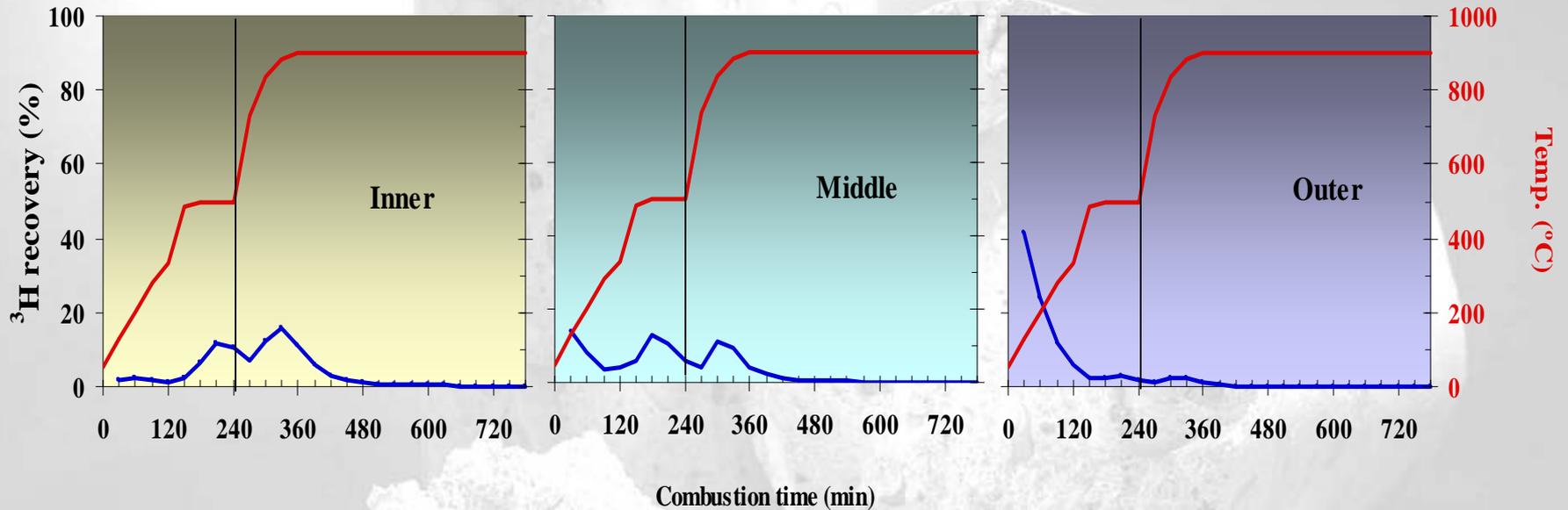
Results

Rapid ^3H evolution from structural concrete



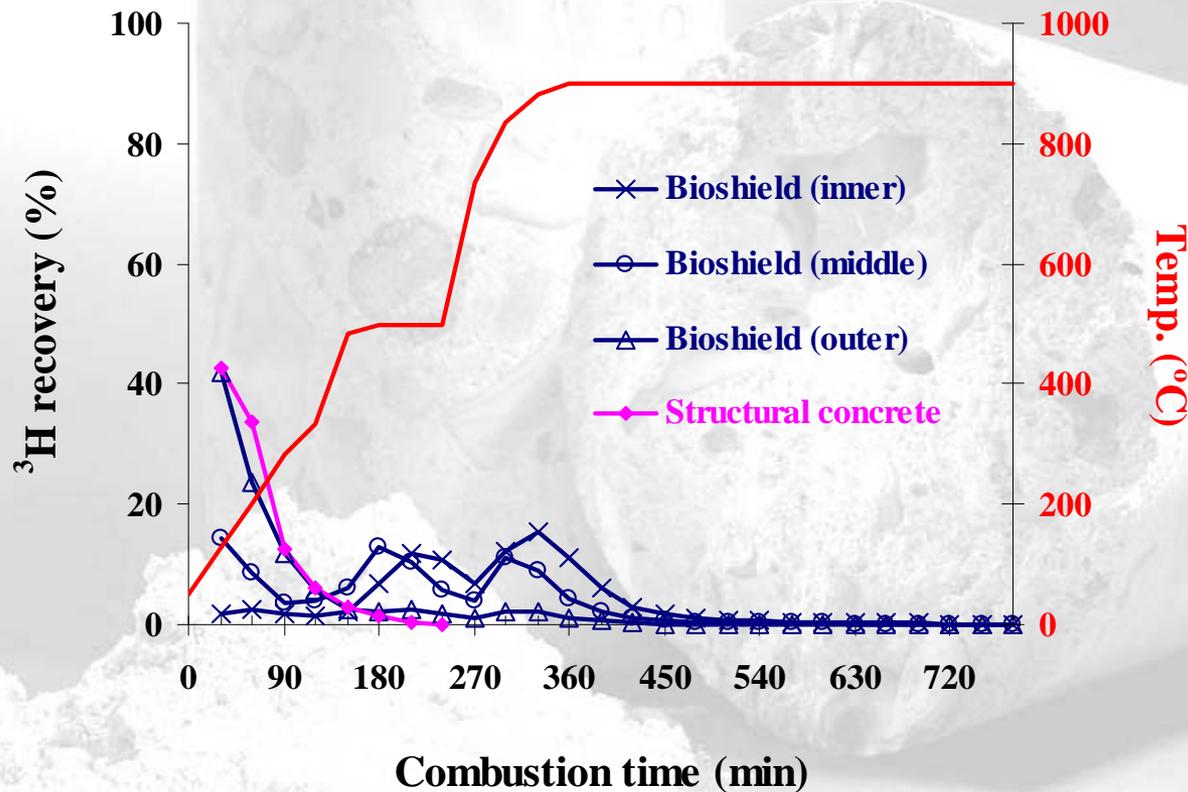
Results

Evolution profile with depth (Bioshield concrete core)



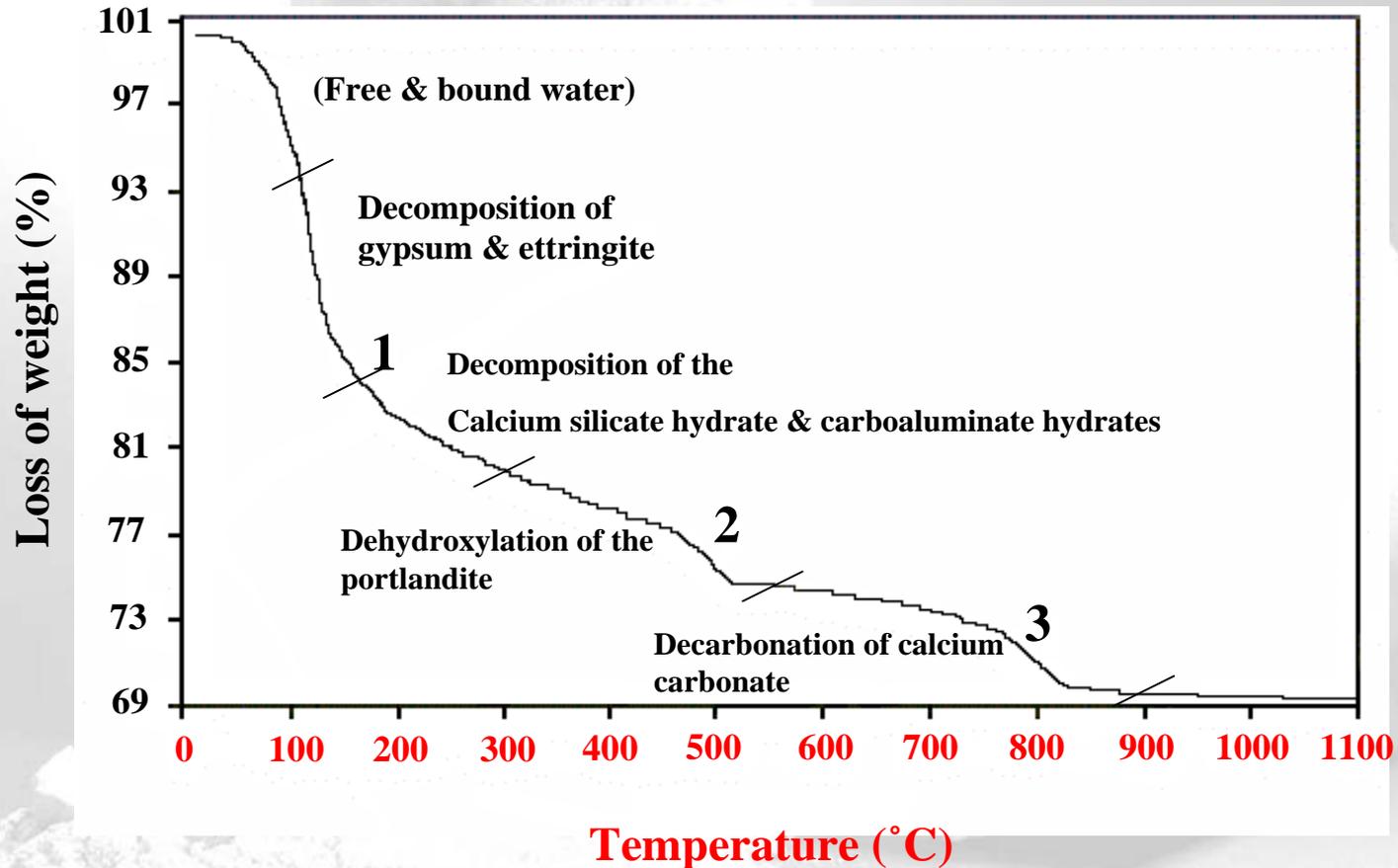
Results

Tritium in a structural & bioshield concrete



Discussions

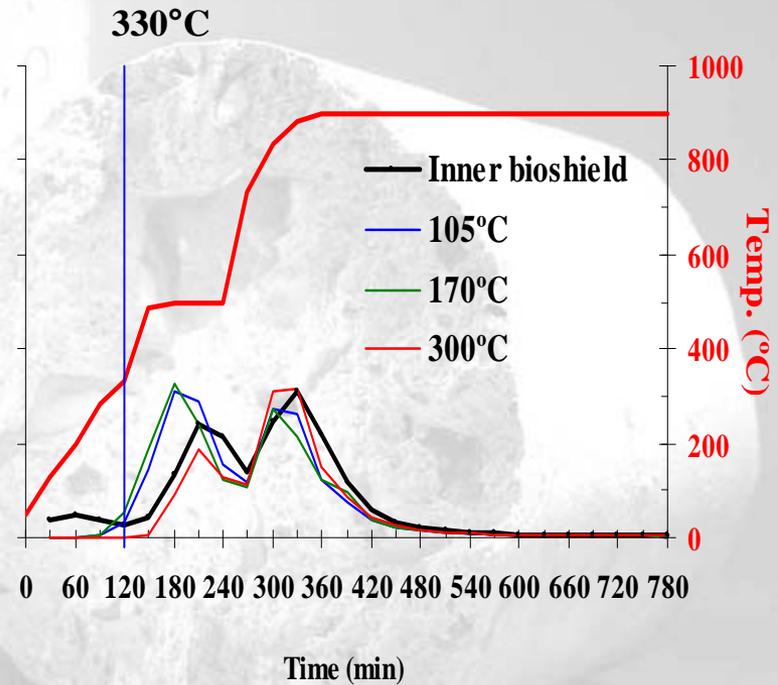
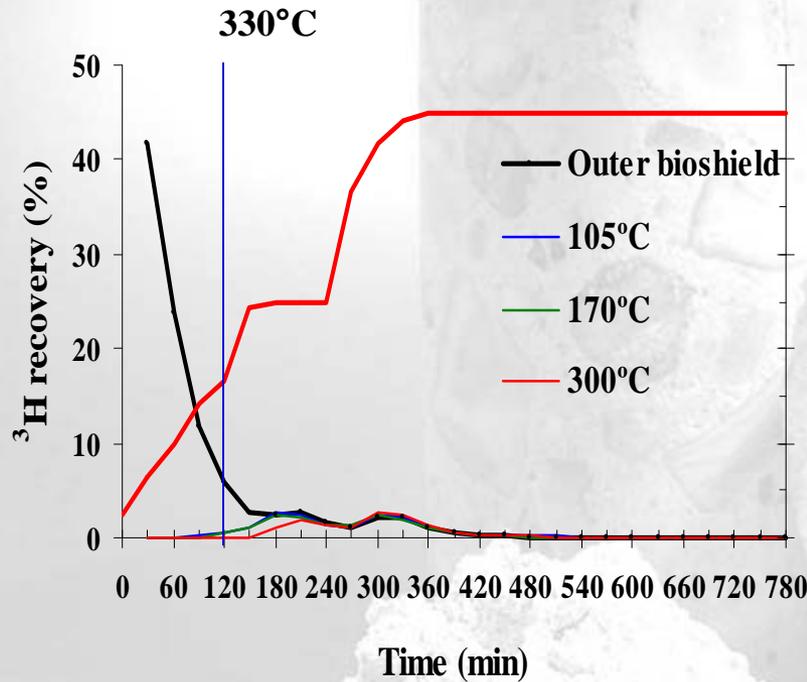
The reaction of concrete and cement paste with an increase of temperature (Alarcon-Ruiz *et al.*, 2005)



Thermogravimetric analysis curves

Discussions

How much loosely bound & strongly bound tritium?



Discussions

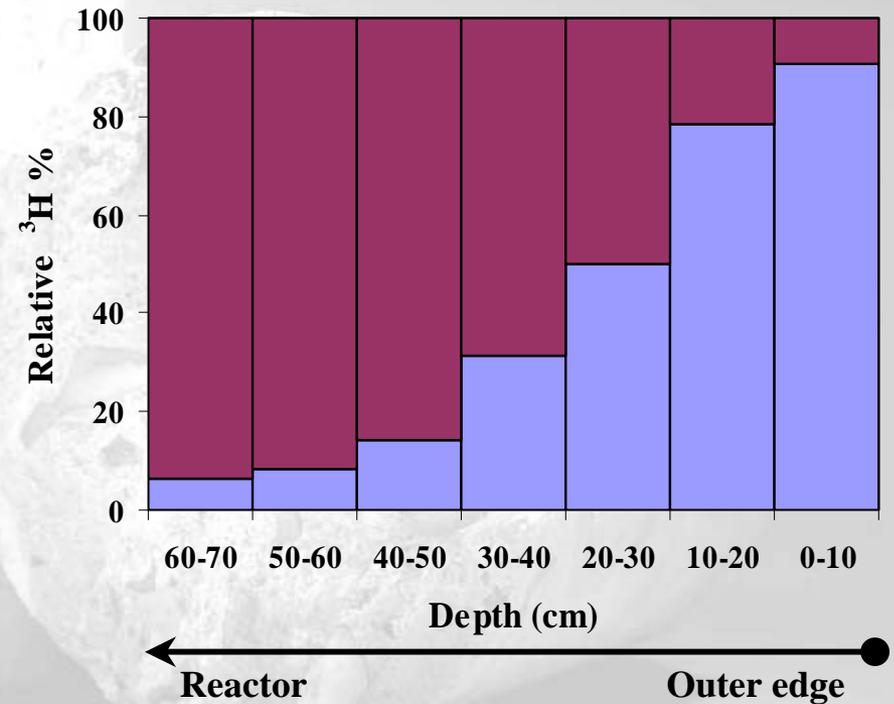
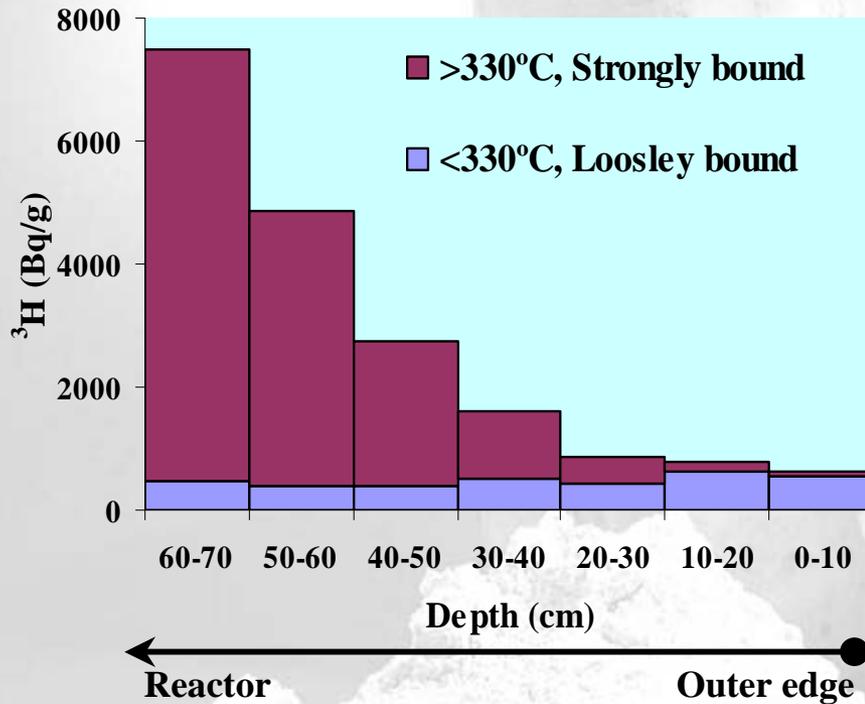
Comparison of ^3H and water in concrete and cement paste

Water group	Numata <i>et al.</i> , (1990)*			Present study		
	Temperature (°C)	% of ^3H in cement material	% of water	% of ^3H of Outer (0-10cm)	% of ^3H of Inner (60-70cm)	Temperature (°C)
Liquid water (Free & capillary water)	<200	77	65	66	4	<200
Water of crystallization	200-450	15	20	20	5	200-485
Water constituent (Calcium hydroxide)	450-550	7	12	7	29	485-500
Water constituent (Calcium silicate hydrate)	600-850	1	3	8	61	500-900

* ^3H data obtained from cement paste contaminated by tritiated water vapour

Discussions

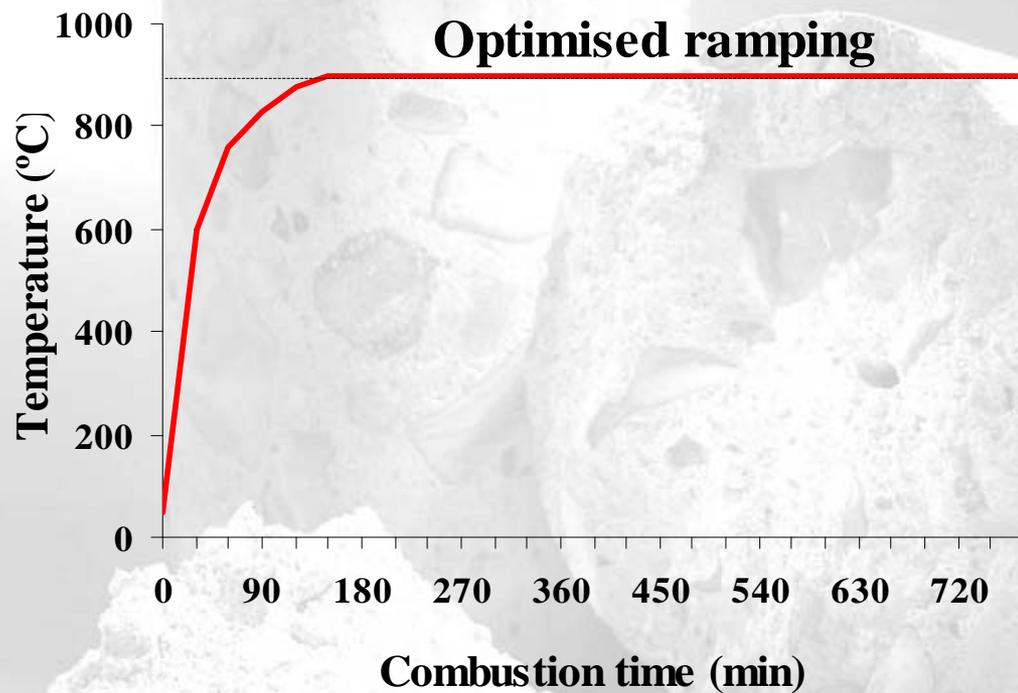
Form of tritium in bioshield concrete core



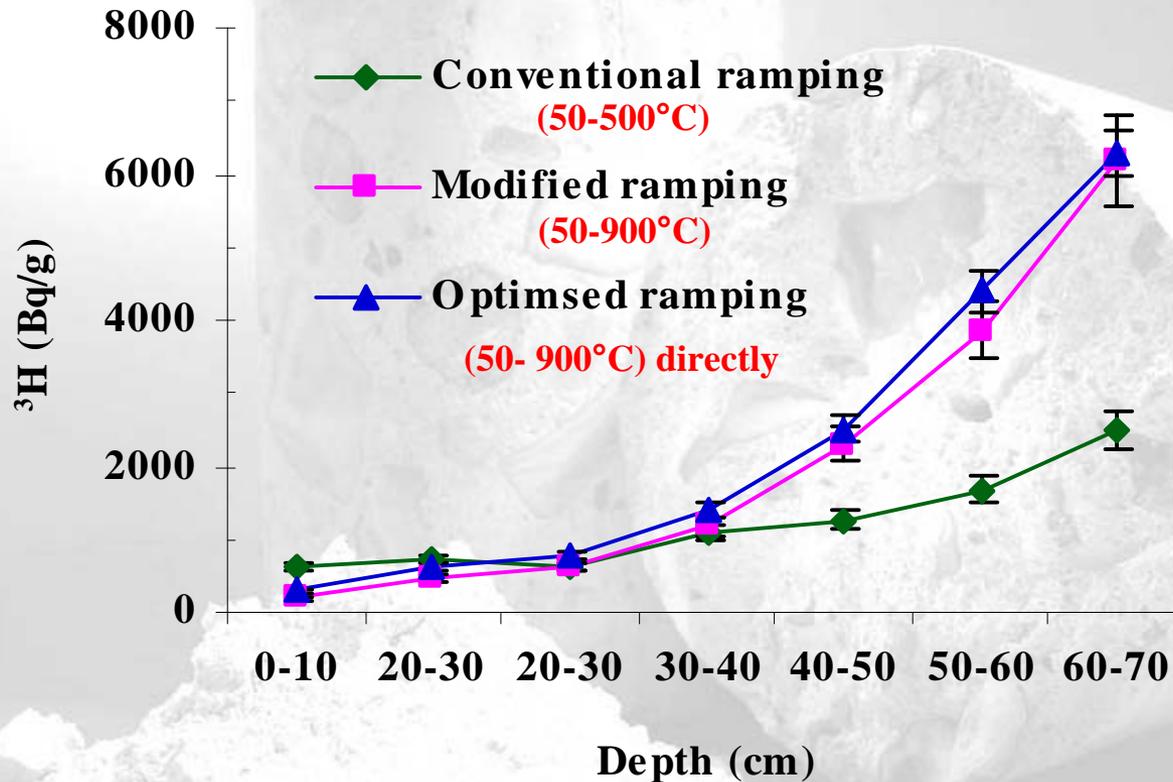
The origin of tritium in reactor concrete

- **Near the reactor side**
 - **Strongly bound ^3H**
 - **Mainly from neutron activation of lithium-6**
- **Outer edge of reactor side**
 - **Loosely bound ^3H mainly from HTO**

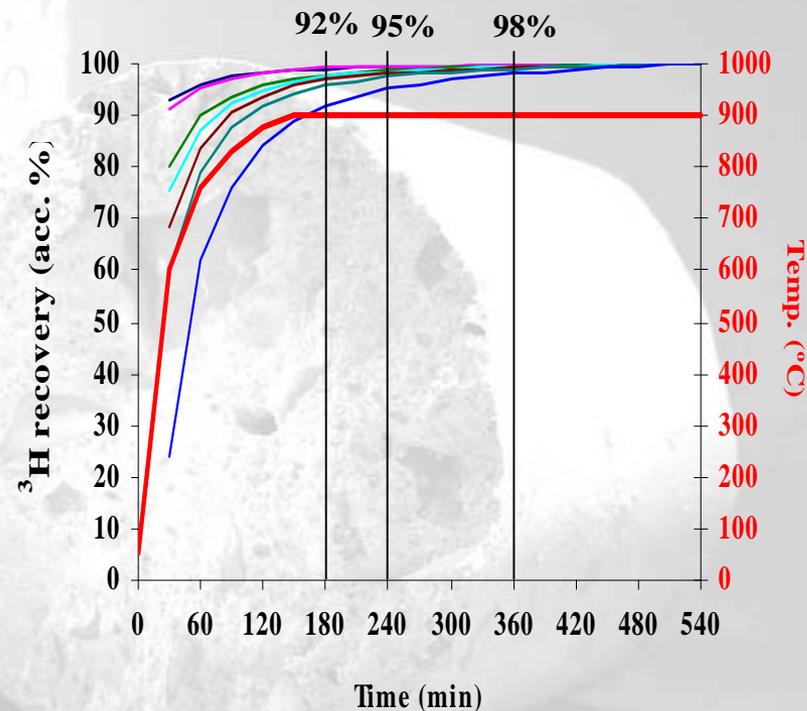
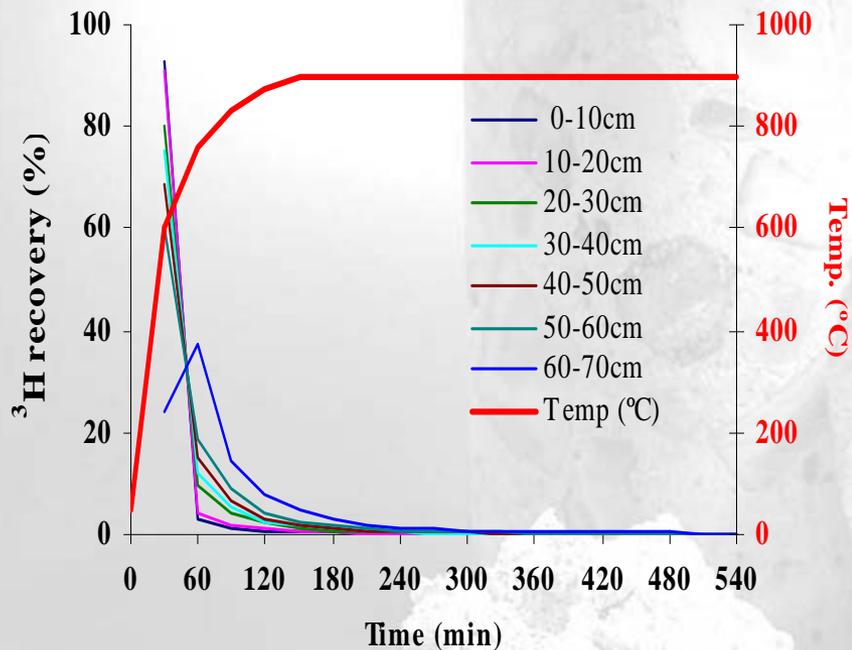
Recommended ramping



Comparison of total tritium measured

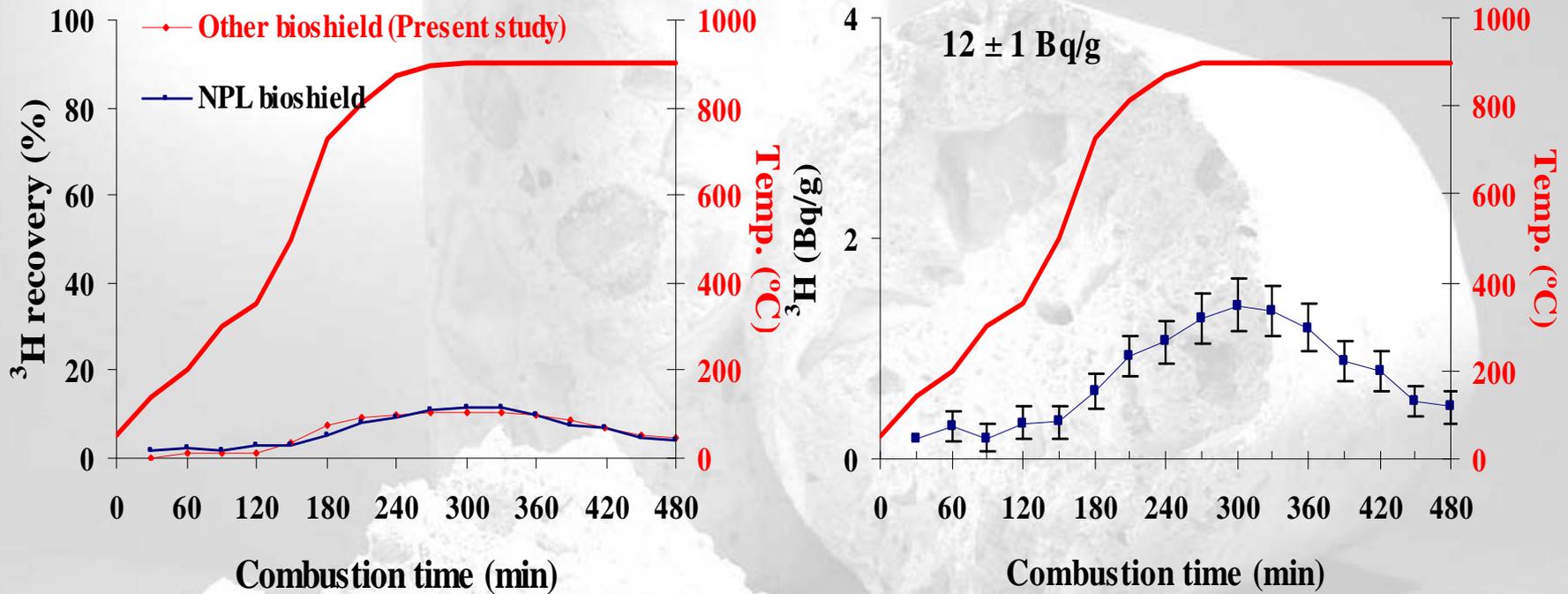


Evolution profile of optimized ramping



Accumulated %

NPL intercomparison exercise 2007



Conclusions: General

- **Demonstrates the importance of determining evolution profiles prior to commencing routine analysis**
- **Evolution profiles provide valuable information on -**
 - the form and origin of ^3H in the sample
 - its implication to sample storage and analysis.
- **Evolution profile of specific materials depends on -**
 - the form of ^3H in the sample
 - the origin of ^3H
 - the sample composition

Conclusions: General

- **Tritium may be produced mainly by neutron capture of ^2H or ^6Li in reactors**
- **Bioshield concrete mainly contaminated by ^3H produced by neutron activation of trace ^6Li ($\sigma_{\text{th}} \sim 940$ barns)**
- **The location of concrete determines the origin of the ^3H**
 - **Closer to reactor:** Strongly bound tritium, produced via neutron activation, dominantly present which can be liberated at high temperature ($>860^\circ\text{C}$)
 - **Far from reactor:** Loosely bound ^3H (from HTO) present which can be liberated rapidly at relatively low temperature ($<120^\circ\text{C}$)

Conclusions: Analysis

- **High temperature (>860°C) and time** are required to liberate ^3H from bioshield concrete (and graphite)
- **Rapid combustion up to 900 °C** without a holding stage will reduce total combustion time

The End

