PROPOSAL

Measurement of the $^{35}\text{Cl}(n,\gamma)$ cross section at n_TOF EAR1


$^1$Universidad de Granada, Spain
$^2$University of Manchester, UK
$^3$European Organization for Nuclear Research, Switzerland
$^4$University of Edinburgh, UK
Plan of the talk

- Motivations
- Status of data
- Setup proposed and counting rate estimations
- Request of protons
Motivations

Dual interest: Health and Environment

• 1. Boron Neutron Capture Therapy
  Cl present in brain and in skin at 0.3%.
  Important for dose calculations in healthy tissue

• 2. Nuclear power plants
  Cl present in the materials of the reactor
  Important for criticality calculations and waste repositories

75% of Cl is $^{35}$Cl, its capture cross section is so large (44 b. at thermal) that even at low concentrations its activation is important.

I. Porras, Universidad de Granada, Spain
1. Boron Neutron Capture Therapy (BNCT)
BNCT: an effective one-day treatment

I.v. infusion of boron compound 4 hours before irradiation (BPA: 400 mg/kg)

Boron accumulates in malignant cells 4 times more than in healthy tissue

Irradiation with epithermal neutron beam according to Monte Carlo calculations (Evaluated data)

\[ ^{10}\text{B}[\text{n},\alpha]^{7}\text{Li} \]

Reaction

\( ^{10}\text{B} \) capture neutrons and produces heavy ions which has a range (5-9 um) of the order of the cell size. The biological effectiveness is much greater than conventional RT.

Thanks to L. Kankaanranta & M. Kortesniemi
BNCT: unique RT option for infiltrative tumors
BNCT: promising therapy for brain tumors

Outcome of BNCT Osaka clinical trial for 21 brain tumor patients, combined with X-Ray Conventional Radiotherapy, and compared to standard treatments

BNCT: promising therapy for brain tumors

Outcome of BNCT Osaka clinical trial for 21 brain tumor patients, combined with X-Ray Conventional Radiotherapy, and compared to standard treatments


Can we optimize the treatment further?
Outcome of BNCT Finnish trial for 22 brain tumor patients (Glioblastoma) depending on dose delivered on Planning Tumor Volume

Adjusting dose may improve outcome

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<th>Dose PTV (Gy)</th>
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But ...

PTV dose 35 Gy (W)

PTV dose 31 Gy (W)
Adjusting dose may improve outcome

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large safe margin, because of uncertainties
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A more precise dose calculation (includes improving all cross sections) could lead to optimize treatment time

⇒ Better therapeutic outcome and possibly survival

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large safe margin, because of uncertainties
Most important processes contributing to dose in brain (healthy tissue)
Energy range of interest: thermal to 100 keV

A spectrum of the main neutron beams for BNCT is shown in the graphs. The graphs compare the neutron flux per unit lethargy across different energies. The energy range of interest is thermal to 100 keV.

I. Auterinen, T. Serén, K. Anttila et al.,
2. Nuclear fission reactors
Chlorine in fission reactors

- $^{35}\text{Cl}$ is present in:
  - Polyvinyl chloride pipes (is 57% Cl by weight)
  - Chlorides are impurities in fuel cladding.
    IAEA-TECDOC-927: Influence of water chemistry on fuel cladding behaviour
  - Cl in nuclear grade graphites (purification processes).
    LLWR Environmental Safety Case: Cl-36 Mobility in Reactor Circuits and its Potential Significance for the ESC 2011 Inventory, LLWRP 1106: SERCO/E003756/008

- $^{36}\text{Cl}$ is produced in fission reactors by $^{35}\text{Cl}(n,\gamma)$ capture
Motivations

Detection of $^{36}$Cl in nuclear reactor waste

When most people think of radioactive waste, elements such as uranium, plutonium, or even americium come to mind. Very few would immediately name chlorine as a problematic element for radioactive waste disposal. But one of chlorine’s radioactive isotopes, $^{36}$Cl, has a particularly long half-life (hundreds of thousands of years) and is extremely water-soluble, making it a potential long-term threat to the environment if it is not disposed of properly. For $^{36}$Cl content to be determined accurately, it must first be separated from its matrix and from other radionuclides, which can be a difficult task if the $^{36}$Cl is embedded in the components of a nuclear reactor. In the April 15 issue of Analytical Chemistry (pp 3126–3134), Xiaolin Hou and colleagues at the Technical University of Denmark present a new set of methods could help authorities safely dispose of the construction materials.

When this nuclear reactor is decommissioned, Hou and colleagues’ methods could help authorities safely dispose of the construction materials.

Cl is a major concern in radioactive waste due to:

- **Half-life of 301000 y.**
- **High mobility (extremely water soluble)**
Motivations

E. Leclerc, G. Smith and P. Lloyd
Review of Cl-36 behaviour in the biosphere and implications for long-term dose assessment ................................................................. 21

- CI-36 inventories in low and intermediate level waste stores are of interest to ANDRA because CI-36 contributes 77% (5.7E-3 mSv/y) of the peak dose at Centre de l’Aube.

- The JGC Corporation in Japan are concerned with Cl-36 issues in Low and Intermediate Level Waste because CI-36 is assessed to be the major contributor to dose following disposal of this waste, assumed to arise due to use of river water for irrigation.

In order to accurately predict the amount of $^{36}$Cl in irradiated fuel, the $^{35}$Cl$(n,\gamma)$ cross section must be known to a high level of accuracy
Energy range of interest

thermal to ~ 1 MeV
Status of data
Uncertainties of 10% between evaluations: too much for precise estimation of $^{36}$Cl production inside the reactor core.
Only one capture measurement in resonances:

Discrepances in the measurement at thermal point
All measurements with nat Cl (75\% $^{35}$Cl, 25\% $^{37}$Cl)

We propose the first measurement with pure $^{35}$Cl
Low E behaviour: bound state

ENDF: Use bound state to fit the thermal value of Mughabghab. -75 eV to -180 eV reported from different authors, different possibilities for low energy behaviour.
Low E behaviour: bound state

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Has this any impact on BNCT dose delivered to brain?

MC Simulations for Snyder head model with different cross section.
Low E behaviour: bound state

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Has this any impact on BNCT dose delivered to brain?

MC Simulations for Snyder head model with different cross section

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<tr>
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<th>Dose 35Cl (n,γ)</th>
<th>%Dose 35Cl(n,γ)</th>
<th>Total Dose</th>
</tr>
</thead>
<tbody>
<tr>
<td>Upper bound</td>
<td>0.516</td>
<td>12.446</td>
<td>4.1830</td>
</tr>
<tr>
<td>ENDF/B-VII</td>
<td>0.514</td>
<td>12.410</td>
<td>4.1439</td>
</tr>
<tr>
<td>Estimation 1</td>
<td>0.551</td>
<td>13.196</td>
<td>4.1421</td>
</tr>
<tr>
<td>Estimation 2</td>
<td>0.325</td>
<td>8.167</td>
<td>3.9803</td>
</tr>
<tr>
<td>Lower bound</td>
<td>0.081</td>
<td>2.162</td>
<td>3.7660</td>
</tr>
</tbody>
</table>
Low E behaviour: bound state

ENDF: Use bound state to fit the thermal value of Mughabghab. -75 eV to -180 eV reported from different authors, different possibilities for low energy behaviour.

Has this any impact on BNCT dose delivered to brain?

MC Simulations for Snyder head model with different cross section

ICRU: Doses must be known with less than 5% uncertainty
Set up and counting rate
Capture Setup in EAR-1

- 4 x C6D6 detectors, (neutron insensitive)

Commonly used in capture measurements at n_TOF
Samples of Na\textsuperscript{35}Cl or K\textsuperscript{35}Cl

Already produced:

- 500 mg 3.2e-3 at/b
- 250 mg 1.6e-3 at/b
Counting rate estimation

3000 bpd  9.8e-3 at/b

Counts / 2e18 protons

Neutron Energy (eV)
Protons requested

$1.5 \times 10^{18}$ protons for $^{35}$Cl measurement
$0.5 \times 10^{18}$ protons for normalization and background

Total $2 \times 10^{18}$ protons
2 PhD students involved

- **F. Ogállar** (University of Granada, Spain)  
Low energy values and application to dose calculations (together with $^{35}$Cl(n,p) recently performed) for BNCT of brain tumors

- **S. Bennett** (University of Manchester, UK)  
Resonance analysis and application to calculations of $^{36}$Cl activity

Close coordination between groups for set-up and analysis
Conclusions

- Low E data (missing data) are important for BNCT
- Data in the resonance region (uncertain, only one measurement with nat Cl) important for radioactive waste calculation
- Feasible experiment: samples and detector available and count rate above background guaranteed
- Two PhD students from different universities benefiting from the analysis and applications
THANK YOU FOR YOUR ATTENTION
Sample of Na$^{35}$Cl or K$^{35}$Cl

Already produced:

500 mg 2 mm

250 mg 1 mm
BNCT: promising therapy for brain tumors

Outcome of BNCT Osaka clinical trial for 21 brain tumor patients, combined with X-Ray Conventional Radiotherapy, and compared to standard treatments

BNCT Treatment Planning – Example of protocol

- Finnish clinical trial – mean values
- Dose not accurately known - BNCT is planned with a safe margin

<table>
<thead>
<tr>
<th></th>
<th>Brain tumor (PTV)</th>
<th>Healthy tissue (limiting factor)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Fractionated Dose (Gy)</strong></td>
<td>60</td>
<td>60</td>
</tr>
<tr>
<td></td>
<td>(30 sessions of 2 Gy)</td>
<td>(30 sessions of 2 Gy)</td>
</tr>
<tr>
<td><strong>Current dose of the BNCT treatment (Gy-Eq) – single session</strong></td>
<td>35</td>
<td>12.1</td>
</tr>
<tr>
<td><strong>Current photon equivalent dose of the BNCT treatment (Gy-Eq) – fractionated</strong></td>
<td>50.5</td>
<td>41.5</td>
</tr>
<tr>
<td></td>
<td>(25 sessions of 2 Gy)</td>
<td>(21 sessions of 2 Gy)</td>
</tr>
<tr>
<td><strong>Target photon equivalent Dose BNCT treatment (Gy-Eq)</strong></td>
<td>71.0</td>
<td>60</td>
</tr>
<tr>
<td></td>
<td>(35 sessions of 2 Gy)</td>
<td>(30 sessions of 2 Gy)</td>
</tr>
<tr>
<td><strong>Target BNCT dose (Gy-Eq)</strong></td>
<td>46.0</td>
<td>16.0</td>
</tr>
</tbody>
</table>
Adjusting dose may improve outcome

A more precise dose calculation (includes improving all cross sections) could lead to optimize treatment time ⇒ Better therapeutic outcome and possibly survival
Energy range of interest: thermal to 100 keV

Spectrum of the main neutron beams for BNCT compared to the accelerator one of Ibaraki based on $^9\text{Be}(p,n)$ reaction

I. Auterinen, T. Serén, K. Anttila et al.,
For resonances above 500 keV, the capture widths were set to the average capture width of the resonances observed in the capture measurements. Average $^{35}\text{Cl}$ capture widths were 606 MeV for $s$ waves and 860 MeV for $p$ waves.

Only one capture measurement in resonances:

PHYSICAL REVIEW C 65, 058801 (2002)

*New Maxwellian averaged neutron capture cross sections for $^{35,37}\text{Cl}$*


**Capture measurements performed only for $^{\text{nat}}\text{Cl}$**

$(75\% \ ^{35}\text{Cl}, \ 25\% \ ^{37}\text{Cl})$
Thermal value

BNCT for head and neck tumors
Detector response:

88Y source: γ’s of 898.0 (93.7%) and 1836 keV (99.2%)
Plot of the calibration run, counts vs channels:
Run#200437, B6D62@900V FS 0.5V, Offset 200mV, Threshold -228
Detector response:

Convolution with a gaussian resolution
Detector response:

Linear transformation channel-energy of the experimental spectrum
\[ E\ (\text{keV}) = a\ n\ (\text{ch}) + b \quad \text{with} \quad a = 133.25\ \text{keV/ch} \quad b = 10\ \text{keV} \]
Detector response: calibration L6D6

\[ E \text{ (keV)} = a \, n \text{ (ch)} + b \]

\[ a = 68.21 \text{ keV/ch} \quad b = 142 \text{ keV} \]

L6D6C@1650V, \(^{88}\text{Y}\) source (calibrated amplitude spectrum)

Simulation w/ 10% resolution
Run #200669
Using area instead of amplitude
## Calibration results:

<table>
<thead>
<tr>
<th>Detector</th>
<th>a (keV/ch)</th>
<th>b (keV)</th>
<th>Run #</th>
</tr>
</thead>
<tbody>
<tr>
<td>B6D61@800V</td>
<td>49.</td>
<td>82.</td>
<td>200440</td>
</tr>
<tr>
<td>B6D63@800V</td>
<td>71.</td>
<td>59.</td>
<td>100745</td>
</tr>
<tr>
<td>B6D63@900V</td>
<td>24.</td>
<td>62.</td>
<td>100744</td>
</tr>
<tr>
<td>L6D6B@1500V</td>
<td>120.</td>
<td>0.</td>
<td>100741</td>
</tr>
<tr>
<td>L6D6B@1600V</td>
<td>85.</td>
<td>0.</td>
<td>100742</td>
</tr>
<tr>
<td>L6D6C@1600V</td>
<td>85.</td>
<td>70.</td>
<td>200440</td>
</tr>
</tbody>
</table>
All measurements: Full beam in vacuum

Configuration **September** 2014:

Detectors: B6D62@900V, L6D6C@1600V

Positions of detectors:
- **Top** #200465, 467-471  FS: 0.5V, Offset 200mV, Thr -228
- **Middle** #200446-454  FS: 0.5V, Offset 200mV, Thr -228
- **Low** #200442-444  FS: 0.5V, Offset 200mV, Thr -228

Configuration **October** 2014 (after shielding):

Detectors: B6D61@800V, B6D62@970V, L6D6C@1650V

Positions of detectors:
- **Top** #200630-631  FS: 0.5V, Offset 200mV, Thr -240
- **Middle** #200696-708  FS: 1.0V, Offset -50mV, Thr -240
- **Low** #200626-627  FS: 0.5V, Offset 200mV, Thr -240
Tests EAR-2 (thanks to Frank Gunsing)
Gold for normalization
Gamma-flash recovery

Counts/bin/protons (no cuts)

- B6D61
- L6D6C

Time - Tγ (ns)

Counts/bin

10^{-17} - 10^{-16} - 10^{-15} - 10^{-14}
Detectors

- The response detector fits well the simulations assuming an energy resolution $\Delta E = \text{const} \ E$
- $\text{B6D6: } \Delta E/E = 5\%$, $\text{L6D6: } \Delta E/E = 10\%$
- Efficiency and recovery from $\gamma$-flash of $\text{L6D6}$ are superior
## BNCT Treatment Planning

<table>
<thead>
<tr>
<th>Tissue</th>
<th>Dose prescribed with photons (Gy)</th>
<th>Photon Isoeffective Dose of the BNCT treatment (Gy-Eq)</th>
<th>Target photon Isoeffective Dose BNCT treatment (Gy-Eq)</th>
</tr>
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<tbody>
<tr>
<td>Healthy tissue</td>
<td>Fractionated: 60 (30 sessions of 2 Gy)</td>
<td>41.5 (21 sessions of 2 Gy)</td>
<td>60 (30 sessions of 2 Gy)</td>
</tr>
<tr>
<td></td>
<td>Single-fraction: 16</td>
<td>12.1</td>
<td>16.0</td>
</tr>
<tr>
<td>Brain tumor</td>
<td>Fractionated: 60 (30 sessions of 2 Gy)</td>
<td>50.5 (25 sessions of 2 Gy)</td>
<td>71.0 (35 sessions of 2 Gy)</td>
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<tr>
<td></td>
<td>Single-fraction: 41.5</td>
<td>46.7</td>
<td>61.6 (PTVdose of 50 Gy)</td>
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- BNCT delivered with safe margin (12.1 Gy to healthy tissue, could receive up to 16 Gy)
- A more precise dose calculation (improving all cross sections) could lead to optimize treatment time ⇒ **Better therapeutic outcome and possibly survival**