NEWSLETTER 2

The nature and effects of the processes and events used to assess the safety of a geological disposal facility must be considered for timescales of hundreds of thousands of years. One of the main issues is the ability to assess these events and processes with sufficient confidence over these long timescales. Confidence depends on the quality and presentation of the research executed. This newsletter is one of a series of newsletters which are intended to inform stakeholders with a general interest in the CAST project.

CAST (Carbon-14 Source Term) is a research project that aims to develop understanding of the potential release of carbon-14 from radioactive waste materials under conditions relevant to waste packaging and disposal to underground geological disposal facilities. The project focusses on the release of carbon-14 as dissolved and gaseous species from irradiated metals (steels, Zircaloys), irradiated graphite and spent ion-exchange resins.

The CAST consortium brings together 33 organisations from 14 countries in the EU, Switzerland, Ukraine and Japan. The involvement of waste management organisations ensures that the project is aligned to European geological disposal programmes and that end results are of use in the safety assessments.

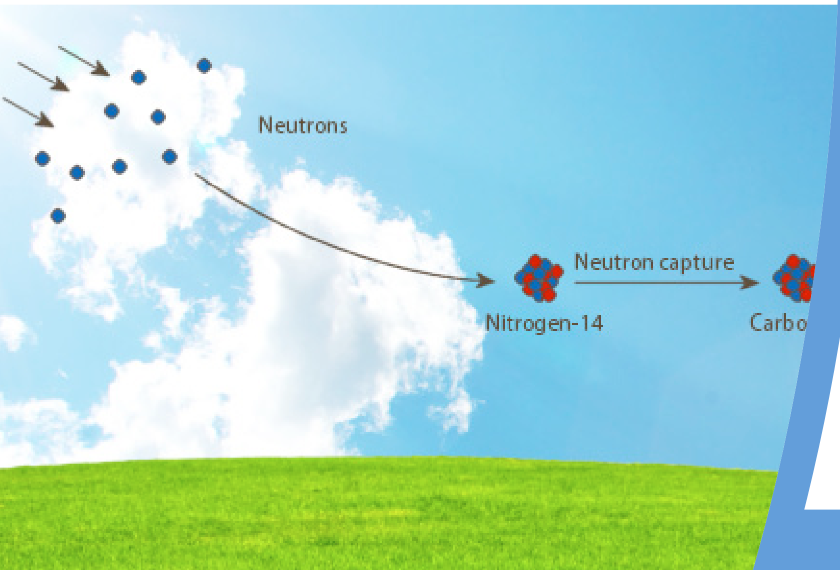
The progress of CAST is visualised in a digital growing tree and continuously updated on the website of CAST www.projectcast.eu

Carbon-14

Outside the Earth's atmosphere, high energetic particles e.g. protons are generated. A cascade of secondary cosmic rays is made by collisions with these high energetic protons (primary cosmic rays). The Earth's magnetic field and atmosphere provide protection against cosmic radiation. A part of these secondary rays are neutrons. The neutron flux at altitudes typical for intercontinental flights is about 10 neutrons $cm^{-2}s^{-1}$. There is more shielding against cosmic radiation at the Earth's surface and consequently the environmental neutron flux is smaller i.e. about 10^{-3} cm^{-2} s^{-1}.

Carbon-14 is a natural radioactive substance (radionuclide) that is continuously generated in our atmospshere by reactions between particles present in the atmosphere and neutrons. The precursors of carbon-14 are oxygen-17, nitrogen-14 and carbon-13. The natural abundances of nitrogen-14, carbon-13 and oxygen-17 are 99.64%, 1.07% and 0.038% respectively. In this Newsletter it is explained why - apart from its natural abundance - primarily nitrogen-14 is responsible for the generation of carbon-14 in radioactive waste.

In a nuclear reactor, the (thermal) neutron flux can be several order in magnitude larger than the environmental flux e.g. 10^{14} cm^{-2} s^{-1} in the core. The neutron flux decreases with increasing distance from the core. In this Newsletter, it is explained that the quantity in distribution of this flux can help to understand the potential locations of carbon-14 containing waste.

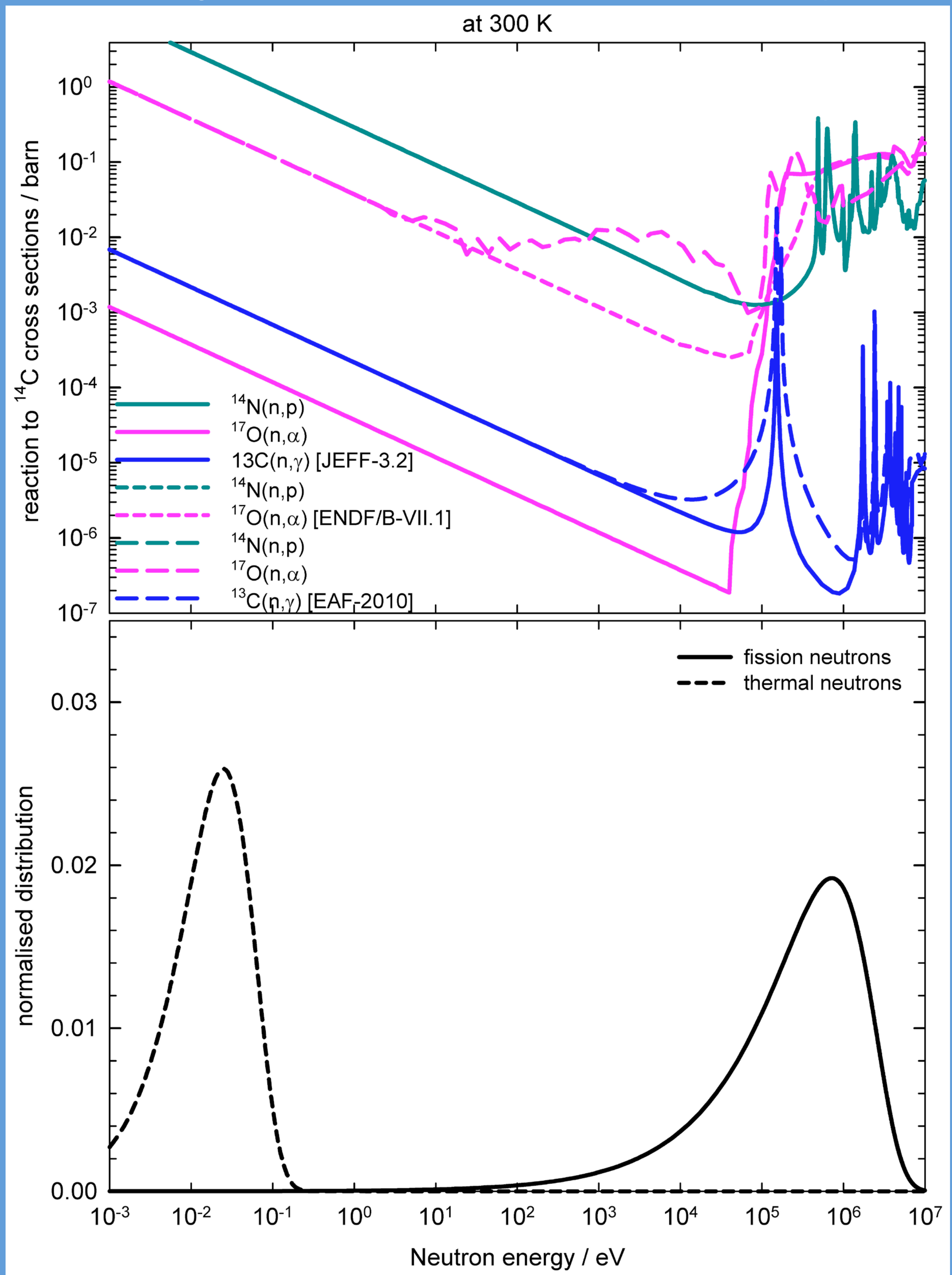


Why are calculations of the content of carbon-14 made?

No gamma rays are emitted during carbon-14 decay, only beta. Its measurement in radiactive wastes cannot be determined non-invasively by gammaspectrometry. As explained in Newsletter 3, carbon-14 in wastes needs to be separated from other radionuclides, if the total carbon-14 content is to be determined. These separations may include conversion of the carbon-14 to a gaseous form for subsequent trapping and analysis. Such analysis require a number of samples to be taken and may be difficult and time consuming. However, it is also possible to calculate the carbon-14 content in many wastes from the operations and decomissioning of a nuclear reactor, limiting the number of samples that may need to be taken for a reliable carbon-14 inventory.

What input is necessary for these calculations?

For a radiological characterisation of the carbon-14 content in unprocessed waste, the cross sections of the precursors of carbon-14, their chemical content in materials, the temperature and energy dependent neutron fluence are needed as input. In CAST, neutron irradiated steel and irradiated graphite are investigated. This waste is often dismantling waste from which the irradiation location in a reactor can be known and consequently the distribution of energy of neutrons and fluence. For processed waste, additional information is necessary e.g. carbon-14 can be redistributed by the used chemicals, temperature and mixing of waste. Irradiated Zircaloy and spent ion exchangers can be processed during the operational period of a nuclear reactor. Workers in nuclear industry - those that operate the nuclear plant or process the waste can contribute to the assumptions made for the calculations e.g. location(s) of the neutron irradiated material in a nuclear plant. The substantiation of the presence of carbon-14 in waste can therefore be best defined upon generation and processing of the waste.



The reaction cross sections shown left have been taken from different libraries, available for free, in the databank from the Nuclear Energy Agency (a specialised agency within the Organisation for Economic Co-operation and Development):

-Joint Evaluated Fission and Fusion File from 2014 [JEFF-3.2];

-the United States library [ENDF/B-VII.1];

-European Activation File [EAF-2010].

The oxygen-17 reaction cross section for thermal neutrons has been reduced by a thousand in the latest evaluation of data. Please note that the y-axis is on a logarithmic scale. For example for thermal neutrons, the reaction cross section for nitrogen is four and three orders in magnitude larger than oxygen-17 and carbon-13, respectively. Consequently, the chemical content of carbon-13 and oxygen17 needs to be three and four orders in magnitude larger than the nitrogen-14 content in order to contribute to the same carbon-14 content. Including the natural abundances as well, the carbon and oxygen content need to be five and seven orders in magnitude larger than nitrogen to contribute to the same carbon-14 content.

The normalised neutron distribution shown left is typical for a light water reactor. Neutrons resulting from the fission of an actinide e.g. uranium-235 are slowed down by collisions with atoms in a moderator. There are water and graphite moderated reactors. The number of collisions that are necessary to have a fission neutron thermalized depends on the density and mass of atoms in a moderator. A mass equal to a neutron is most effective; the slowing down power for light water (H_2O) is small (6-7 mm), for graphite it is larger (about 17 cm). The time necessary to slow down ranges between 10^{-5} till 10^{-4} seconds. The main lifetime of a neutron in a reactor is determined by the period for thermal diffusion that ranges between 10^{-4} till 10^{-3} seconds. At 300 K, the thermal diffusion length for light water and graphite are 2.9 and 59 cm , respectively. Consequently, at same neutron irradiation temperature, carbon-14 can be generated further away from the core in graphite moderated reactors than in light water moderated reactors.

Discrimination between conventional and radioactive waste is made in the planning of dismantling of nuclear plants. Not all carbon-14 contents in materials are considered hazardous. In the latest Council Directive 2013/59/EURATOM laying down basic safety standards for protection against dangers arising from exposure to ionising radiation, the activity concentration for clearance have been set to 1 Bq per gram solid matter. That is a chemical concentration of carbon-14 in e.g. iron of 0.000024 ppm. The chemical concentration of precursors of carbon-14 can be more than a million times larger.

Irradiated steels

Irradiated stainless steel and carbon steel can be present in wastes. Stainless steels have a higher corrosion resistance than carbon steels and are used for many components within the core of a water-cooled reactor (e.g. grid supports, pipework) as well as for the internal cladding of the ferritic steel reactor vessel. Frequently in CAST, the nitrogen content of (irradiated) steel used for experiments was not known and needed to be measured. From a Spanish nuclear reactor and steel representative to that used in a Finnish reactor the maximum in chemical content of nitrogen and carbon is similar to commercial stainless steel: a maximum of 0.10 wt%. The carbon-14 content of an internal part (guide tube assembly) of the Spanish reactor appeared to be 18 Bq per gram solid matter.

CAST report D2.2: Annual Progress Report on WP2 - year 1
CAST report D2.5: Annual Progess Report on WP2 - year 2

Irradiated Graphite

CARBOWASTE was a EU funded research programme that focussed on irradiated graphite. In CAST, the CARBOWASTE results are evaluated in the context of national safety assessments. The knowledge available for (determination of) the carbon-14 inventory expected in Germany, France, Spain, United Kingdom (UK), Romania, Lithuania, Italy and Ukraine has been collected. A nitrogen content of 40 and 70 ppm was used in the Romanian and Lithuanian calculations. Also in irradiated graphite, nitrogen can be the main contributor to the carbon-14 inventory. The activity concentration as a function of the position in the nuclear reactor was calculated and ranged from 10^0-10^6 Bq per gram solid matter. In the UK, for AGR, a nitrogen content of 15 ppm nitrogen is assumed and the contribution to carbon-14 from carbon-13 is calculated to be 40%.

CAST report D5.5: Review of understanding of inventory and release of C14 from irradiated graphite

Past training course

The training courses have been envisaged for Master and PhD students with a background in (nuclear) engineering, physics, geology, chemistry and mathematics. The specific interest of this group is to learn and gain knowledge and skills to address waste management issues associated with carbon-14 containing waste. In Germany, the first training course has been held in July 2016. The course was organised by Karlsruher Institute of Technology (KIT). German and Croatian students attended the training course. Professionals were allowed to attend the training course as well.

A group of 11 nuclear interested persons were taught in how carbon-14 is generated in a nuclear reactor, what types of waste contain carbon-14, how the carbon-14 in waste can be measured and the disposal concepts for carbon-14 containing waste. The group visited hot cell laboratories and waste processing and storage facilities. On the last day, they experienced wearing gas masks and working in a glove box. The presentations used in the training course are published on www.projectcast.eu.

Upcoming training course

The first course was re-scheduled in February 2016 because of a lack of registrations. Carbon-14 in waste may be a too specialised field for Master and PhD students. There also seems to be other organised courses / events that compete for the attention of early career researchers. Therefore, attempts are made to organise the second training course in conjuction with another international event.

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CAST
Co-ordinator: Simon Norris| simon.norris@nda.gov.uk
Website: www.projectcast.eu

Newsletter
Editor: Erika Neeft | erika.neeft@covra.nl