CArbon-14 Source Term

CAST

Advisory Group Review of WP 7 CAST Final Overview Report (D1.17)

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CAST – Project Overview

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

The CAST consortium brings together 33 partners with a range of skills and competencies in the management of radioactive wastes containing carbon-14, geological disposal research, safety case development and experimental work on gas generation. The consortium consists of national waste management organisations, research institutes, universities and commercial organisations.

The objectives of the CAST project are to gain new scientific understanding of the rate of release of carbon-14 from the corrosion of irradiated steels and Zircaloys and from the leaching of ion-exchange resins and irradiated graphites under geological disposal conditions, its speciation and how these relate to carbon-14 inventory and aqueous conditions. These results will be evaluated in the context of national safety assessments and disseminated to interested stakeholders. The new understanding should be of relevance to national safety assessment stakeholders and will also provide an opportunity for training for early career researchers.

For more information, please visit the CAST website at:
http://www.projectcast.eu

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Executive Summary

One of the tasks of the CAST Advisory Group is to review the final synthesis reports from the different Work Packages. This report represents the review of the final synthesis report from WP 7 on the overall CAST project [NEEFT 2018].

In addition to summarising the outcomes of the CAST project, D7.23 provides additional information and insights from the author. The release of C-14 from the four different waste forms is also put into a safety context by comparing the rates of release expected based on the results from CAST with the release rate from soils of natural C-14 from cosmogenic sources.

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1 Introduction

During the CAST project, the focus of Work Package 7 was the dissemination of the results from the project to various stakeholders. This dissemination included various activities, such as workshops for regulators and other stakeholders, a periodic newsletter, training courses, the CAST website, and working with specific CAST partners.

Deliverable D7.23, an overview of the CAST project, is not focussed on dissemination activities but instead gives a summary of the technical activities and achievements from the project [NEEFT 2018]. The report is more than a summary, however, and includes additional information and insights from the author. It concludes with an interesting comparison between the likely consequences of C-14 released from the four waste forms considered in CAST (irradiated steels and zirconium alloys, spent ion-exchange resins (SIERs), and irradiated graphite) with that from natural C-14 (as CO_2) released from soils.

2 General Comments

Deliverable D7.23 [NEEFT 2018] is written in the form of a commentary on the CAST project. It is part summary, part source of additional insights from the author, and part a vehicle for putting the outcome of the CAST project and the significance of C-14 from the four waste forms into a safety assessment context.

NEEFT [2018] appropriately highlights some of the issues arising from the CAST project. Due to the limited release of C-14 from irradiated steels and Zr alloys under simulated disposal conditions, it was not possible to determine the speciation from these sources. There is some emphasis in the report on the successful speciation of stable carbon, possible in large part because of the much higher concentration. However, these results should be treated with some caution, especially in the case of steels where much of the stable carbon will be present as carbides, since the dissolution behaviour of carbides (which are known to be electrochemically active) may be quite different from that of C-14 present in solid solution. Importantly, however, NEEFT [2018] also highlights some of the conservatisms that were, and may still be, inherent in the treatment of C-14 in the safety assessment. For example, it is clearly conservative to assume that organic C-14 is released in uncharged, molecular form that is not retarded by precipitation or sorption. It is also now apparent based on the CAST results, that the release of C-14 from SIERs may be vastly over-stated, both because there is no measurable release (over laboratory timescales) from cemented resins and because the inventory may be over-estimated if it is based on measurements on wet resins (which may release >99.9% of the inventory upon drying).

There is a certain degree of mechanistic discussion and inference in the report. Certainly in the case of irradiated steels and Zr alloys, it was a challenge for the CAST partners to simply obtain active samples and to set up and conduct exposure tests. The main focus of efforts was to try to obtain measurable C-14 release which, given the small inventories and the extremely slow release rates, proved to be a challenge. Whilst there were indications from the experiments of radiolysis effects and interactions between released C-14 and H_2 produced by corrosion, the available experimental evidence was too limited to draw definitive conclusions.

NEEFT [2018] contributes new information and insights to the CAST project. For example, there is a discussion of recently updated neutron-capture cross sections which have implications for the significance of the different sources of C-14 (namely, neutron activation of N-14, O-17, or C-13). There is also discussion of the implications of differences in the type of cement (ordinary Portland cement (OPC) versus that produced using blast furnace slag).

3 CAST Summary

Sections 4, 5, and 6 of NEEFT [2018] provide a summary of the CAST results in terms of the respective sources, inventories, and speciation of released C-14 for the four different waste forms.

In terms of the source of C-14 for each of the waste forms, the activation of N-14 is the major pathway for irradiated steels and Zr alloys and for a portion of the C-14 in i-graphite (that associated with pores and edge sites). The C-14 in the i-graphite matrix and at interstitial sites is produced by activation of C-13. Clearly, the source(s) of C-14 in SIERs depends on the nature of the nuclear power plant, the mode of operation, the type of resin, and the history of treatment and storage of the resin.

NEEFT [2018] provided a high-level analysis of the C-14 inventory for each of the four waste forms. On a mass basis, the inventories reported in the CAST project were of the order of 10^5 Bq/g for irradiated steels, 10^4 Bq/g for Zr alloys, 10^3 Bq/g for SIERs (although with variations depending on the source and whether the measurements were made on wet or dried resins), and a range from 10^0 Bq/g to 10^6 Bq/g for i-graphite. In this latter case, the wide range is a result of the different origins and irradiation histories of the different samples of i-graphite considered in CAST.

In terms of the speciation of released C-14, there were challenges in the case of irradiated steels and Zr alloys because the amount released to either solution or in the gas phase was too small to analyse. For unconditioned SIERs, the speciation simply reflected the speciation of the original ionic species. In the case of i-graphite, NEEFT [2018] cautioned that crushing graphite (in order to increase the effective surface area in an attempt to increase the release rate) could preferentially release C-14 from pores and edge sites which, if not captured, could compromise the conclusions of the speciation from the matrix and from interstitial sites.

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In order to place the C-14 release into context, NEEFT [2018] presented an interesting comparison between the rate of cosmogenic C-14 release from soils and the likely release rate from the various waste forms. Of the four waste forms, only irradiated steels were considered to release C-14 at a rate higher than the cosmogenic rate, but this rate would be attenuated by transport, retardation, and dilution between the repository and the surface. In summary, the expected behaviour for the four waste forms was summarised as follows:

- Irradiated steels – although the C-14 release rate at the source could be a factor of 100 times higher than the cosmogenic $^{14}CO_2$ release rate from soil, it was expected that attenuation through the engineered barriers, geological barrier, and biosphere would result in a dose consequence several orders of magnitude lower than that associated with the cosmogenic C-14.
- Irradiated Zr alloys – due to the lower inventory (factor of x10) and the lower corrosion rate (and reduced oxide inventory), the C-14 release rate at the source was estimated to be similar to that for the surface release of natural C-14 from soil. Once the attenuation through the various engineered and natural barriers is factored in, the dose consequence of C-14 from irradiated Zr alloys can be expected to be less than that from natural C-14.
- SIERs – no equivalent source term was defined for SIERs in D7.23 as no release was observed from cemented resins in the CAST project. Thus, it is concluded that spent resins will be an insignificant contributor to the dose.
- i-graphite – very little release was observed from i-graphite in the CAST project and it is apparent that much of the inventory is not releasable. Therefore, it was concluded that the source term would be less than that due to natural C-14, significantly so once attenuation by the engineered and natural barriers is taken into account.

This was an interesting way to put the relative significance of these C-14 sources into contet and nicely summarised the entire intent of the CAST project. Based on this analysis, it is apparent that, despite remaining uncertainties, C-14 release from the four waste forms considered in CAST should be no greater, and likely significantly less, than that from natural C-14 from cosmogenic sources.

Reference

NEEFT, E.A.C. 2018. Final overview of CAST (D7.23). CAST (Carbon-14 Source Term) Report, issued 27/06/2018.