Applied Radiation and Isotopes 110 (2016) 224–229

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Radioisotopes produced by neutron irradiation of food

S. Albright *, R. Seviour

West Building (W3/01), The University of Huddersfield, Huddersfield, West Yorkshire, England HD1 3DH, United Kingdom

HIGHLIGHTS

- We show that neutron interrogation of food can produce many radioisotopes.
- We show a strong dependance between food and certain radioisotopes.
- Some isotopes are shown to have an energy dependence.
- Previous claims that 24Na is the main threat is shown to only apply in special cases.

ARTICLE INFO

Article history:
Received 11 January 2015
Received in revised form
29 November 2015
Accepted 8 December 2015
Available online 31 December 2015

Keywords:
Ingestion dose
Cargo interrogation
Food activation
Induced activity
Neutron activation

ABSTRACT

The use of neutrons for cargo interrogation has the potential to drastically improve threat detection. Previous research has focussed on the production of ^{24}Na, based on the isotopes produced in pharmaceuticals and medical devices. For both the total activity and the ingestion dose we show that a variety of isotopes contribute and that ^{24}Na is only dominant under certain conditions. The composition of the foods has a strong influence on the resulting activity and ingestion dose suggesting that the pharmaceuticals and medical devices considered initially are not a viable analogue for foodstuffs.

There is an energy dependence to the isotopes produced due to the cross-sections of different reactions varying with neutron energy. We show that this results in different isotopes dominating the ingestion dose at different energies, which has not been considered in the previous literature.

1. Introduction

Millions of containers pass through national borders every year, the port of Felixstowe alone sees approximately 3 million containers measuring approximately $6.1\,\text{m} \times 2.44\,\text{m} \times 2.59\,\text{m}$ annually. Inspecting the contents of these containers for content validation is vital for combating smuggling and for international terrorism prevention.

Current X-ray based security methods struggle to determine the presence organic material when shielded by high density, or disguised by other low density, materials. The potential for using neutrons in place of X-rays is gaining interest, with the potential for greatly increased threat detection (Liu et al., 2008). A variety of neutron techniques are available, the fast neutron technique Pulsed Fast Neutron Analysis (Brown and Gozani, 1995) has a lot of interest however there are techniques using all energies available. A detailed discussion of neutron scanning techniques is available in Runkle et al. (2009), Buffler (2004) and the references within.

When considering the use of neutron interrogation the efficacy for contraband identification is not the only consideration. The activation of irradiated goods is an unavoidable side effect of neutron irradiation and some research has been performed to identify the level of threat this may pose (Nelson, 2006; Tenforde, 2002; Giroletti et al., 2012).

For cargo interrogation 14 MeV $T(d, n)$ sealed neutron generators are typically used, e.g. in Nelson (2006), Giroletti et al. (2012) however 8.5 MeV $D(d, n)$ reactions are also considered, as in Tenforde (2002). ^{252}Cf fission sources can also be used for techniques requiring white or thermal spectra.

The authors of Nelson (2006) considered the effect of irradiating a broad range of materials, from jars of Ragu to metal sheets, with a 14 MeV neutron beam. The time required for the irradiated material to return to background was measured and used as the figure of merit. The time required for induced activity to decay away showed that the longest times were less than typical storage times at port. The results were limited however as only a single thickness of material, e.g. 1 jar of Ragu or 1 thin sheet of metal, was used leading to minimal moderation. As the beam

* Corresponding author.
E-mail addresses: simon.albright@hud.ac.uk (S. Albright), r.seviour@hud.ac.uk (R. Seviour).

http://dx.doi.org/10.1016/j.apradiso.2015.12.032

Table 1
The elemental composition of the foods simulated. The relative mass (g/100 g) of each element (3 s.f.) in the simulated foods is given.

Element	Almonds	Banana	Brie	Cocoa	Corn	Potato
H	6.47×10^{0}	9.85×10^{0}	8.59×10^{0}	6.26×10^{0}	6.72×10^{0}	1.01×10^{1}
C	4.34×10^{1}	1.11×10^{1}	2.38×10^{1}	4.34×10^{1}	4.03×10^{1}	9.61×10^{0}
N	3.67×10^{0}	1.90×10^{-1}	3.59×10^{0}	2.93×10^{0}	1.63×10^{0}	1.30×10^{0}
O	4.44×10^{1}	7.85×10^{1}	6.16×10^{1}	4.42×10^{1}	5.05×10^{1}	7.83×10^{1}
F	0	2.20×10^{-6}	0	0	0	0
Na	1.00×10^{-3}	1.00×10^{-3}	6.30×10^{-1}	2.10×10^{-2}	3.50×10^{-2}	6.00×10^{-3}
Mg	2.70×10^{-1}	2.70×10^{-2}	2.00×10^{-2}	5.00×10^{-1}	1.30×10^{-1}	2.30×10^{-2}
P	4.80×10^{-1}	2.20×10^{-2}	1.90×10^{-1}	7.30×10^{-1}	2.10×10^{-1}	5.70×10^{-2}
S	3.00×10^{-1}	1.60×10^{-2}	3.00×10^{-1}	2.40×10^{-1}	1.30×10^{-1}	1.10×10^{-1}
Cl	1.50×10^{-3}	1.50×10^{-3}	9.70×10^{-1}	3.20×10^{-2}	5.40×10^{-2}	9.20×10^{-3}
K	7.10×10^{-1}	3.60×10^{-1}	1.50×10^{-1}	1.52×10^{0}	2.90×10^{-1}	4.20×10^{-1}
Ca	2.60×10^{-1}	5.00×10^{-3}	1.80×10^{-1}	1.30×10^{-1}	7.00×10^{-3}	1.20×10^{-2}
Mn	2.30×10^{-3}	3.00×10^{-4}	0	3.80×10^{-3}	5.00×10^{-4}	2.00×10^{-4}
Fe	3.70×10^{-3}	3.00×10^{-4}	5.00×10^{-4}	1.40×10^{-2}	2.70×10^{-3}	8.00×10^{-4}
Cu	1.00×10^{-3}	1.00×10^{-4}	0	3.80×10^{-3}	3.00×10^{-4}	1.00×10^{-4}
Zn	3.10×10^{-3}	2.00×10^{-4}	2.40×10^{-3}	6.80×10^{-3}	2.00×10^{-3}	3.00×10^{-4}
Se	2.50×10^{-6}	1.00×10^{-5}	1.45×10^{-4}	1.43×10^{-4}	1.55×10^{-4}	3.00×10^{-7}

was almost entirely unmoderated there would have been very few neutrons in the thermal region where many cross-section resonances lie.

The authors of Tenforde (2002) showed that under 8.5 MeV neutron irradiation the dominant threat isotope in pharmaceuticals and medical devices was ^{24}Na. The method used in Tenforde (2002) was to calculate the induced activity based on a spectrum with fast and thermal components rather than a full Monte-Carlo approach.

The results of Tenforde (2002) were extended in Tenforde (2003) to include ^{24}Na production in food. As with Tenforde (2002) the conclusion of Tenforde (2003) was that no unacceptable level of activation would be seen. To determine if the induced activity would pose a problem the authors of Tenforde (2002), Tenforde (2003) calculated the ingestion dose for irradiated goods. The acceptable dose received by the public as a result of irradiation was set at 1 mSv/year, the greatest dose calculated was 1 μSv/year.

The threat isotopes considered by Tenforde (2003) may not be readily applied to all foodstuffs as the compositions of foods and pharmaceuticals are not necessarily equivalent. Additionally the target elements considered by Tenforde (2002) have omissions which may not matter for pharmaceuticals but may be significant for some foods. Activation reactions based on Ca(n,X) were not included in Tenforde (2002), but Ca is found in significant quantities in a variety of foods, including dairy and tofu.

The results of Tenforde (2003) were further extended by the authors of Giroletti et al. (2012) who considered ^{24}Na production by 14 MeV neutron irradiation. As with Tenforde (2003) only ^{24}Na production was considered, however the increased neutron energy may enable additional reactions and the applicability of pharmaceuticals as an analogue of food is still to be verified.

In this paper we show that the induced activity and ingestion dose are caused by a variety of isotopes. Furthermore we show that the conclusion that ^{24}Na is the dominant threat isotope is only valid under certain conditions. Finally we show that there is an energy dependence for induced activity and ingestion dose, which may justify consideration of the interrogation energy.

2. Simulations

By performing numerical simulations we are able to produce any energy neutron beam desired. In this paper we consider the radioisotopes produced by neutron beams with energies from 1 MeV to 20 MeV incident upon a variety of foods, we then show how the dominant cause of ingestion dose and activity varies as a function of time, energy and on the food type irradiated.

We used two numerical techniques to identify the radioisotopes produced, MCNPX (Forster et al., 2004) and Fispact-II (Sublet et al., 2012). MCNPX is a Monte-Carlo radiation transport code, which uses point wise data sets and numerical models to track particles through materials. Fispact-II is a nuclear inventory code, which provides time dependent production, and decay, of radionuclides under arbitrary irradiation conditions.

Neutron transport in MCNPX was used to generate the neutron spectrum, the spectrum was then passed to Fispact-II to simulate the isotope production. The MCNPX simulations used a pencil beam source of perfectly monochromatic neutrons. The neutrons were directed at a cubic volume, 1 m on each side, of material and the spectrum was recorded 10 cm before the far face of the volume. The Fispact-II simulations were run with a fluence of 10^{9} neutrons and irradiation time of 60 s to provide the activation products.

The results obtained from these simulations are specific to the flux and fluence used. Changing the flux and/or fluence will influence the levels of activation after irradiation. For example at higher flux the interrogation time will be reduced meaning that short lived isotopes will be at higher levels immediately after irradiation.

In both MCNPX and Fispact-II the foods were simulated with the same elemental ratios and natural isotopic composition. The trace elements, protein and water content per 100 g provided the elemental composition, with the remaining mass simulated as cellulose. The protein composition was calculated using the generic formula $C_nH_{1.85n}N_{0.28n}O_{0.3n}S_{0.01n}$ (Torabizadeh, 2011) and the trace elements considered were Ca, Fe, Mg, P, K, Na, Cl, Zn, Cu, Mn, Se and F. The masses of each element used in g/100 g are shown in Table 1.

The foods used were Almond, Banana, Brie, Cocoa, Corn, Potato and Rice, they were chosen as they cover a broad variety of compositions and are commonly containerised for import/export. The elemental ratios used in these simulations was chosen to approximate the average composition of distributed foods. The elemental composition will vary with country of origin and cellulose is not the only organic component however these approximations are sufficient to highlight any significant effects.