Applied Radiation and Isotopes 66 (2008) 1781–1787

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

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

Calculation of attainable superheats and predicted embryonic flux rates in commercial water isotope targets

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ARTICLE INFO

Article history:
Received 22 January 2008
Received in revised form
25 April 2008
Accepted 25 April 2008

Keywords:
Fluorine-18
Water targets
Bulk and spinodal nucleation
Thermalhydraulics
Statistical mechanics

ABSTRACT

Phase change in a fluid subject to a large spectrum of radiations is examined. The statistical variation in the fluid energy is considered in concert with the radiation field attributes to establish conditions for spinodal decomposition and bulk nucleation. This approach is developed in general and carried forward for the specific case of ^{18}O-enriched water used in commercial targets for the production of ^{18}F. Sensitivities of the outcome to specific attributes of the fluid state model are examined. The possibility for very high bulk nucleation site densities is exposed and may explain the observed thermal-fluid behavior in commercial water targets. These targets operate at elevated pressure and temperatures at and in excess of the saturation temperature. They are also subjected to a large spectrum of radiations, with differing levels of energy deposition. The conditions for spinodal decomposition and bulk nucleation (with and without radiation) in these targets are evaluated. It is likely that some bulk nucleation is occurring, and causing the density reduction locally in the target. Suitable experiments to evaluate this potential are more fully possible.

1. Introduction and background

Historical evaluations of nucleation due to radiative energy first considered the energy required for individual nucleation events, and examined the radiation interaction with the fluid that could provide the energy to cause the nucleation. These theories have been confirmed with experiments. In the more general case, the statistical distribution of energy in a fluid interacts with the statistical deposition of radiative energy, resulting in some probability density (e.g., flux rate) for nucleation events. These approaches have also been confirmed with experiments, but only for limiting situations where a few initial nucleation events occur. This evaluation extends consideration to conditions where the nucleation site density is large, and conventional phase change (boiling) models may fail to represent the fluid phase change behavior. The development is general, but centers on the performance of commercial ^{18}O-enriched water targets used to produce ^{18}F. Other applications may include astrophysics, materials processing, and neutron scattering research.

Positron emission tomography (PET) is a nuclear medicine imaging technology, first developed by Phelps et al. (1975). Human imaging using these techniques was first published in 1976 (Phelps et al., 1976; Hoffman et al., 1976). Positron-emitting molecular probes that illuminate biologically significant processes were also developed. In 1978 Fowler, Wolf and coworkers at Brookhaven National Laboratory developed a glucose analog labeled with ^{18}F (2-deoxy-2-fluoro-D- glucose or [^{18}F]FDG (Ido et al., 1978). The first images of this tracer in the human brain were obtained the next year (Phelps et al., 1979). However ^{18}F is not easily produced or distributed. It has a half-life of 109.8 min, necessitating production of the isotope in the city or region of the patient, and on the day of injection. In order to produce the [^{18}F]FDG required for clinical imaging, compact commercial particle accelerators and compact targets for the production of ^{18}F are required. The most common precursor form of the isotope for synthesis is aqueous [^{18}F]fluoride ion, produced on small accelerators via proton bombardment of ^{18}O-enriched water.

In 1983 Bruce Wieland and Al Wolf demonstrated that small volume enriched water targets could be used to produce aqueous ^{18}F (Wieland and Wolf, 1983). By the mid-1990s the state of the art was ~1 Ci/h/target (Solin et al., 1988; Tewson, 1989; Tewson et al., 1988; Guillaume et al., 1991; Roberts et al., 1995; Van Brocklin et al., 1995; Lepera and Dembowski, 1997; Berridge and Kjellstrom, 1999). Overwhelmingly these designs were natural convection/phase change targets. Steinbach et al. (1990) have shown in an elegantly simple paper that phase change even at modest beam powers is nearly unavoidable, and contributes to the

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0969-8043/$ - see front matter © 2008 Published by Elsevier Ltd.
doi:10.1016/j.apradiso.2008.04.019

heat transport for liquid targets. In 2000, Alvord and Ruggles experimentally confirmed the existence of a stable vapor jet originating at the base of the entrance window (Ruggles and Alvord, 2000). This vapor jet provides, through shear forces, high-velocity upflow of the liquid volume the protons stop in. Without this upflow the liquid water in this region would reach temperatures above the critical point, and would certainly nucleate in the bulk. This model assumes most of the liquid volume is at saturation conditions.

Ionization heating at the level of 660 W is deposited on an 8-mm-diameter circular area. Moreover, the proton beam intensity is not uniform, but is a roughly Gaussian profile in both horizontal and vertical dimensions. This means that the central 4 mm diameter spot is subjected to power densities of $\sim 3 \times 10^{10}\,\mathrm{W/m^3}$. The water in upflow due to vapor shear will have attained some superheat (on the order of 5 K) before entering this center hot spot. The vertical water velocity in the system is calculated to be about 0.5 m/s, and the transport time through this heated zone is about 8 ms. The heating rate for water in this small volume is about 7700 K/s, and the superheat just in this region is then 61 K. Added to the 5 K superheat entering the region, and the exiting flow can be expected to be 66 K above saturation, or at 318 °C (for 600 psia and saturation temperature of 252 °C).

It is then important to understand the tolerable superheat of water at the pressures encountered in a fluoride production target. In particular, the reduction in this tolerable superheat temperature due to the particular spectrum of radiations in the target should be known for the model to be complete. It is probable that the vapor jet model as published in 2000 represents an upper limit to performance (lower limit in steam volume at a given power). Understanding the superheat limits of water under irradiation should refine the vapor jet model.

The widespread commercial utilization of the new high-power water targets not withstanding, there remain challenges and opportunities for the designer of commercial water targets. The cost of enriched water is still between $40 and $100/g, and the targets utilize 2–3 g per bombardment. The designs in place still utilize a larger inventory of water than the minimum thickness one would calculate using the vapor-jet model. There appear to be more subtle phase change or density reduction phenomena in the targets that have yet to be fully understood. The theory of radiation assisted nucleation has not been applied to superheated water and to the particular radiations of interest here, nor have correlating measurements at elevated pressure and temperature and in the specific radiation field been carried out. This work applies nucleation theory in the presence of a radiation field to water, with particular attention to conditions of current target design, and seeks to establish whether available nucleation rate measurement apparatus would be useful in determining the likelihood of further density reduction of operating water targets.

2. Energy loss

Building on the Plane Wave Born Approximation, Bethe (1930) developed a quantum mechanical treatment of ionization cross sections that remains a sufficiently powerful and simple tool for calculation of the energy loss per unit length (called linear energy transfer or LET) for any charged particle passing through matter. In his derivation, which took into account interaction not only with the potential of the atom but also all the wave functions of the individual electrons, he showed that the minimum energy transfer can be replaced by the average ionization potential, which is an average of the binding energies of all the atomic electrons. The Bethe–Bloch equation for energy loss takes the form

$$-\frac{\mathrm{d}E}{\mathrm{d}x} = \frac{4\pi e^4 z^2}{mv^2} NZ \ln\left(\frac{2mv^2}{I}\right) \tag{1}$$

where e is elementary charge, z is the charge of the incident particle, m is the electron mass, v is the incident particle velocity, N is the density of the target particles, Z is the charge of the target nucleus, and I is the average ionization potential of the atom (using cgs electrostatic units). Using this, and given a maximum energy transfer, one can quickly and roughly estimate average LET for the particles considered herein.

In order to bound the energies, and therefore the LETs considered, it is important to estimate the maximum energy transfer for a number of free particle collisions that could happen in the target. The particular inelastic collision of most interest in these targets (^{18}O(p,n)^{18}F) is endoenergetic. As such that collision will always result in recoil energies that are less than the elastic collisions. The kinematics of nonrelativistic elastic collisions are worked out in many texts (see for example Evans, 1955)

$$T_{\max} = T4\frac{M_1 M_2}{(M_1 + M_2)^2} \tag{2}$$

Using this equation a table of estimated maximum energies can be generated (Fig. 1). For the maximum energy cases, one can also use the Bethe range-energy equation to estimate the maximum LET that will be seen in the resulting recoil particle track.

The LET calculations in Fig. 1 are calculated using SRIM 2003 (Ziegler et al., 1985), which essentially employs versions of Eq. (1) plus elastic scattering to calculate stopping and range, and the energy imparted to recoils. The notable reactions from the perspective of generating high-LET particles are elastic scattering of protons on ^{18}O atoms, and elastic scattering of neutrons on ^{18}O atoms. The highest LET of a recoiling ^{18}O atom in water is at the highest energy of 2.2 MeV. Similarly the highest LET noted for ^{18}O recoiling from a neutron event is at the maximum energy as well.

In addition to the clearly significant proton–oxygen and neutron–oxygen scattering, there are four other reactions to note. Proton–proton (elastic scattering of protons by hydrogen nuclei), and neutron–proton (elastic scattering of neutrons by hydrogen nuclei) are second-order processes, resulting in low-energy proton tracks at a rate $\ll 1$ per incident proton. They will not be addressed further in this analysis. In the same sense, electron–electron reactions do not contribute significantly to microstructure of the type that would result in significant thermal spikes, and so only

			Target Particle	Proton	Electron	^{18}O
Incident Particle	Max energy (MeV)	Rest mass (MeV)	Rest mass (MeV)	938	0.511	16,854
Proton	11.0	938	% transfer	100%	0.22%	20.0%
			max resulting energy (MeV)	11.0	0.024	2.20
			max LET at or below max energy (MeV/cm)	**900**	**800**	**10,300**
Electron	0.024	0.511	% transfer	0.22%	100%	0.01%
			max resulting energy (MeV)	0.000052	0.024	0.000003
			max LET at or below max energy (MeV/cm)	N/A	**800**	N/A
Neutron	7.4	935	% transfer	100%	N/A	19.9%
			max resulting energy (MeV)	7.4	N/A	1.47
			max LET at or below max energy (MeV/cm)	**900**	N/A	**8,000**

Fig. 1. Comparison of various LETs.