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An analytical approach of thermodynamic behavior in a gas target system on a medical cyclotron

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HIGHLIGHTS

- We developed an analytical model describing gas targets on a medical cyclotron.
- Heat transfer coefficients are calculated.
- Excellent agreement is observed compared to experimental data

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ABSTRACT

An analytical model has been developed to study the thermo-mechanical behavior of gas targets used to produce medical isotopes, assuming that the system reaches steady-state. It is based on an integral analysis of the mass and energy balance of the gas-target system, the ideal gas law, and the deformation of the foil. The heat transfer coefficients for different target bodies and gases have been calculated. Excellent agreement is observed between experiments performed at TRIUMF's 13 MeV cyclotron and the model.

1. Introduction

Isotopes for molecular imaging, especially for Positron-Emission Tomography (PET) are widely produced on low energy (7–30 MeV) cyclotrons using targets comprised of gas, liquid or solid materials. The application of these isotopes is wide spread, ranging from cancer detection and diagnosis, to neurology in the study of Parkinson's Disease or Alzheimer's (Ruth, 2009), to applications in the Life Sciences (Unkles et al., 2011; Heath et al., 2007; Semeniuk et al., 2009). With the recent issues encountered by a number of isotope production reactors around the world (Van Noorden, 2013), attention has rapidly shifted to alternative production sources for a growing number of isotopes that span imaging, therapeutic and industrial applications (Kraeber-Bodr et al., 2015). One alternative to reactor-based isotope production includes cyclotrons. With nearly 1000 machines installed and operating around the globe (Schaffer et al., 2015), including a growing number of higher power machines ($>$ 20 MeV, $>$300 μA), there has come an increase in demand for advanced target hardware with enhanced power dissipation capabilities in order to maximize production quantities. The thermodynamic processes involved in a solid target irradiation include the interaction of a proton beam with the target material, as well as the heat removal (Helus, 1983). Additional complexity is found in gas targets where convective currents are formed during irradiation (Heselius and Solin, 1986). In addition, phase transitions in liquid targets are observed (Peeples et al., 2011; Peeples, 2006, 2008; Alvord, 2008; Hong et al., 2013). Understanding the thermodynamics inside a gaseous or liquid target system during irradiation at different operating conditions can potentially contribute to an optimal target design with the promise of higher yields and improved operational reliability. Herein, we study the global behavior of gas targets as a system without phase change.

In the past, several groups have systematically studied the behavior of gas targets and derived equations based on observations and simple models. For example, Robertson et al. (1961),

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McDaniels et al. (1972), and Oselka et al. (1977) and were among the first to describe the gas density reduction in a gaseous target caused by the impinging proton beam. Koble et al. (1989) studied the effect of natural convection on regular gas targets and developed a model to estimate the density reduction inside the proton beam profile, concluding that free convection is the dominant mechanism of heat transfer, especially at higher gas densities. Wojciechowski et al. (1988) have proposed an energy balance to estimate the range of the proton beam in the target gas, estimated the foil temperature during irradiation, established an approximation for the maximum target pressure before foil failure, and developed an empirical model correlating the proton beam current with the pressure rise during irradiation. Using this correlation and assuming the ideal gas law, they calculated the bulk gas temperature and the heat transfer coefficient between the target gas and the cooling system with a beam current of $0-45\ \mu A$ at 26.5 MeV beam energy. A comprehensive study about the influence of an intense ion beam on high pressure gas targets has been carried out by Heselius et al. (1982). The penetration of high energy α-particles (2–18 MeV) in helium, neon and argon gas targets has been visualized using interferometry. In addition, they studied both horizontal and vertical target orientation. In the horizontal target, they discovered an asymmetric gas density reduction in the beam volume due to an asymmetric beam penetration in the vertical direction (perpendicular to the target length). However, the gas density reduction in the vertical target was discovered to be symmetric in the beam volume. They reported that the asymmetric behavior in the density reduction takes place due to the upward buoyancy-driven transport of the heated gas in the enclosure due to gravity. Moreover, Heselius et al. (1982) estimated the temperature distribution in the target cross-section using interferometry results and the ideal gas law. Finally, Heselius and Solin (1986) developed mathematical models to both model the stopping location of the proton beam and temperature distribution in the vertical direction in high pressure gas targets. Wieland et al. (1990) developed a novel method to measure the beam penetration in gas targets with respect to different initial pressure and beam currents. They used an electro-meter at the back of the target to detect the current of charged particles when the beam collides with the back wall which is used as an indicator for the beam penetration. Based on their results, increasing the beam current requires increasing the initial pressure in the chamber to stop the beam inside the gaseous domain.

Ache and Wolf (1988) studied the effect of radiation on the reaction of recoil carbon-11 in a nitrogen–oxygen system. They studied the oxidation of $[^{11}C]CN$ and $[^{11}C]CO$ as a function of energy dissipated by the proton beam in the system and found that $[^{11}C]CN$ was mostly oxidized to $[^{11}C]CO_2$. Moreover, they reported the linear behavior of the oxidation of $[^{11}C]CO$ to $[^{11}C]CO_2$ with radiation dose.

Experimental studies have been carried out on the effect of the gas target body material on the final production yield (Roberts et al., 1995; Bishop et al., 1996). Buckley et al. (2000, 2004) studied the dependence of different target materials on the yield of $[^{11}C]CH_4$ produced in the $^{14}N(p,\alpha)$ reaction, and found a dependence of the yield as a function of irradiation duration that deviates from the simple production equation. The authors suggest that the produced ^{11}C adheres to the wall before it can form $[^{11}C]CH_4$ and is therefore lost in the production. Gillings et al. (2014) studied the effect of target temperature on recoverable yield of $[^{11}C]CH_4$. Based on their experimental observations, the target yield increases by increasing the temperature of the system and the irradiation time.

Understanding the transient thermodynamics of turbulent flow coupled with non-equilibrium beam–fluid thermodynamics and fluid–structure interaction in gas targets is extremely difficult. An insight into this highly coupled mechanism can be achieved by simplifying the analysis and studying the gas target system at steady-state. Here, the goal is to understand the thermodynamics of a regular gas target at the steady-state and develop a mathematical model. In this work, our hypothesis is that a steady-state exists during irradiation. We apply the developed model in an attempt to understand the range of validity of the steady-state hypothesis, analyze the pressure rise from the energy balance equation to compute the values of the overall heat transfer coefficients, and find a trend for the overall heat transfer coefficients at various operating conditions using experimental results.

2. Material and methods

2.1. Experiments

All irradiations were performed at the TR13 cyclotron at TRIUMF, a self-shielded 13 MeV cyclotron with external ion source, accelerating negative hydrogen ions. Details of the cyclotron are described by Laxdal et al. (1994) and Buckley et al. (2000). Several target bodies and target materials were used, see Table 1. All targets were used in the same configuration: the target was mounted on the target selector with a vacuum seal. An aluminum foil, 25 μm thick, separated the target from the cyclotron vacuum. As the proton beam deposits 0.2 MeV into the foil (SRIM, 2013), a helium cooling window assembly directs a helium jet onto the back of the aluminum foil with a flow of around 65 *l*/min at 70 kPa (10 psig). On the other end of the helium cooling window, a 38 μm thick HAVAR® foil separates the target chamber from the helium flow. The proton beam deposits 0.8 MeV into this second foil which is consequently cooled by a second helium jet with the same flow in the same window. The target chamber holds the target gas. Three different target bodies were used, all 12 cm long. The aluminum target body consists of a conical shape with an internal minimum diameter close to the HAVAR® foil of 1 cm and a maximum diameter at the distal end of the target of 2 cm. The niobium target body has a cylindrical shape with an internal diameter of 1.5 cm. The tantalum target body likewise has a cylindrical shape and an internal diameter of 1.25 cm. All target bodies were surrounded by a sleeve with water cooling in between (flow of 6 *l*/min). Argon gas, nitrogen gas, nitrogen gas with 0.5% oxygen, and nitrogen gas with 10% hydrogen were used as target gases. Initial gas gauge pressures ranged from 2100 kPa to 2400 kPa (300–350 psig) and beam currents ranged from 0 to 20 μA. All targets are loaded and unloaded through the same high-pressure valve, mounted about 300 cm from the target and connected with a stainless steel line with an inner diameter of 0.159 cm (1/16 in.).

Table 1
Description of TR13 gas-targets under various operating conditions.

ID	Target body	Target gas	Initial pressure P_o (kPa)	Internal surface area (cm^2)
A11	Conical Al	Ar	2100 (300 psig)	570
A21	Conical Al	N_2/O_2	2100 (300 psig)	570
A22			2250 (325 psig)	
A23			2400 (350 psig)	
N11	Cylindrical Nb	N_2/O_2	2250 (325 psig)	570
N12			2400 (350 psig)	
N21	Cylindrical Nb	N_2/H_2	2250 (325 psig)	570
T11	Cylindrical Ta	N_2	2100 (300 psig)	470
T21	Cylindrical Ta	N_2/O_2	2100 (300 psig)	470
T31	Cylindrical Ta	N_2/H_2	2100 (300 psig)	470