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Photonuclear production of yttrium-88 – A high energy gamma emitter for hydrocarbon extraction applications

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HIGHLIGHTS

- Production of ^{88}Y via ^{89}Y(γ,n) reaction results in high specific activity product.
- The MCNP simulations of ^{88}Y yield agreed with the yield measurements.
- Activities of yttrium byproducts were also measured and found to be as predicted.

ABSTRACT

The use of fracing has risen over the past decade and revolutionized energy production in the US. However, there is still an impetus for further optimization of the extraction of oil and natural gas from vast shale reservoirs. In this work, we discuss photonuclear production of yttrium-88 as a promising radiotracer for fracing operations. Single neutron knock-out from natural monoisotopic yttrium-89 is an inexpensive process resulting in high activity of ^{88}Y with minimal impurities. MCNPX simulations were performed to estimate the ^{88}Y yield. Irradiations of natural yttrium using a 32 MeV electron linac equipped with a tungsten bremsstrahlung converter were done to benchmark the simulations. Activities of ^{88}Y, ^{87g}Y, and ^{87m}Y were measured and found to be in good agreement with the predictions.

1. Introduction

Radiotracers are currently in widespread use as a diagnostic tool in fracing operations. Briefly, after a formation has been fraced, a proppant is injected into the cracks to ensure hydrocarbons have open pathways to flow. To verify the efficacy of a given fracing operation, proppants are often tagged with a relatively short-lived radiotracer which emits gamma ray radiation that can be detected by a downhole detector, thus determining the location of a crack. Radiotracers currently in common use include ^{192}Ir, ^{110m}Ag, and ^{46}Sc, where typically up to 200 mCi are injected per fracing application (Whitten et al., 2000). New radiotracers, such as ^{88}Y, can greatly benefit fracing operations. ^{88}Y cannot be produced in a reactor, as natural yttrium is a monoisotopic element, ^{89}Y. One of the easiest ways to remove a neutron from ^{89}Y is by using an inexpensive 30–40 MeV electron accelerator to produce a high photon flux and induce photo-neutron knockout. To the authors' knowledge, this method has not been explored in this context, and currently there is no reliable supply of ^{88}Y. In this work we will demonstrate that photonuclear production of ^{88}Y will result in a high specific activity product which could potentially be used for many applications including fracing.

The potential advantages of ^{88}Y as a radiotracer (with a half-life of 107 days and major gamma ray emissions of 898 keV and 1836 keV) arise from the spectrum of its emissions. First, its highest energy emission exceeds that of other commonly used radiotracers, enabling it to be easily distinguished from other tracers which may be left in the formation from previous fracing operations. As such, ^{88}Y can be used in conjunction with current radiotracers to enhance their effectiveness. Second, the higher energy of its gamma rays is subject to less attenuation in the formation, enabling sensitivity to proppants deeper into the formation. Third, the fact that its two major gamma rays are widely spaced in energy provides the potential for obtaining higher fidelity information on the location of the radio-tagged proppant in the formation. This arises from the fact that the absorption of gamma rays in matter is energy dependent, and the differential absorption of the two gamma ray lines is indicative of the depth of the proppant in the formation.

To provide a reliable supply of ^{88}Y, we propose a photonuclear

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production method which has proven to be a robust means of production of many radionuclides. This technique has also appeared to be more attractive over nuclear reactor based methods for many other radioisotopes. Compared to reactor-based production it results in a relatively low volume of radioactive waste, considerably lower ecological hazard, and lower regulatory constraints. Electron accelerators have been proven to have feasibility for production of significant quantities of other valuable radioisotopes, including ^{47}Sc, ^{67}Cu and ^{225}Ac (Dovbnya et al., 2001; Mamtimin et al., 2015; Maslov et al., 2006; Rane et al., 2015; Rotsch, 2016; Starovoitova et al., 2014; Starovoitova et al., 2015). The type of linac suggested here is commercially available and comparable in size and cost to the accelerators commonly used in hospitals and sterilization facilities.

2. Methods

2.1. Photonuclear reactions

A high power electron linear accelerator can create both proton and neutron rich isotopes by producing bremsstrahlung photons that can knock out nucleons from stable atoms. A schematic of the process is shown in Fig. 1. An electron beam impinges upon a metal converter, generating a continuous spectrum of high energy gamma rays with maximum energy approximately equal to that of the incident electrons. If the energy of the photon exceeds the reaction threshold energy, neutrons and/or protons can be emitted, producing the desired isotope.

The bremsstrahlung converter is an essential component of the photonuclear production of the radioisotopes with electron linacs. A properly chosen converter maximizes the fraction of the energy from the electrons which is transferred to photons. Conventional converters are made of solid metals with a high melting point, atomic number and density, and include tungsten, tantalum, platinum, lead, and gold. Liquid metal converters capable of dissipating tens of kWatts of beam power can also be used for high power accelerators (Starovoitova and Segebade, 2016). Bremsstrahlung photons form a forward cone and are incident on the sample which is typically placed right after the converter. The most suitable photon energy for photonuclear production of radioisotopes is 10–40 MeV – the region where the photo-absorption cross-section is the greatest.

The production rate R of the produced isotope depends on a number of parameters, such as the density of target nuclides that are irradiated N_T, the threshold energy of the nuclear reaction E_{th}, the maximum energy of photons E_{max}, the photon flux density $\varphi(E,r)$ (photons/cm^2/sec/MeV), and the cross-section of the photonuclear reaction $\sigma(E)$. If the target is significantly large so that the photon flux is not uniform throughout the target, the production rate R can be calculated as:

$$R = \int_{E_{th}}^{E_{\max}} \int_V N_T(\vec{r})\phi(E, \vec{r})\cdot\sigma(E)\, dE d^3\vec{r} \quad (1)$$

The yield of the radioisotope produced in the sample can be found from the production rate as:

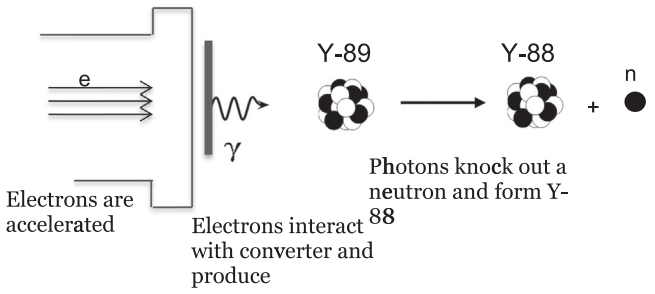

Fig. 1. A schematic of the process whereby an electron accelerator is used to make a high energy bremsstrahlung photon beam for isotope production.

Table 1
Products of photo-activation of yttrium (Chu et al., 1999).

Reaction	Threshold energy, MeV	Product half-life	Energies of emitted gammas, keV	Decay chain
^{89}Y(γ,n)^{88}Y	11.5	107 days	898 (91%), 1836 (99%)	^{88}Y→^{88}Sr (stable)
^{89}Y(γ,p)^{88}Sr	7.1	Stable		
^{89}Y(γ,2n)^{87g}Y	20.8	80 h	495(91%)	^{87g}Y→^{87}Sr (stable)
^{89}Y(γ,2n)^{87m}Y	20.8	13 h	381(77%)	^{87m}Y→^{87g}Y→^{87}Sr (stable)
^{89}Y(γ,np)^{87m}Sr	18.6	2.8 h	388(79%)	^{87m}Sr →^{87}Sr (stable)
^{89}Y(γ,αn)^{84m}Rb	18.9	21 min	215(37%), 248(65%), 464(32%)	^{84m}Rb →^{84}Rb→^{84}Kr (stable)
^{89}Y(γ,γ')^{89m}Y	0.9	16 s	909(99%)	^{89m}Y→^{89}Y (stable)

$$A(t) = R(1 - e^{-\lambda \cdot t_i}) \quad (2)$$

where λ is the decay constant of the produced radioisotope and t_i is the irradiation time.

For medium and heavy nuclei (such as ^{89}Y) the total cross section in the giant dipole resonance region is dominated by emission of a single neutron. Emission of protons and other charged particles is also possible, but in general less likely as these processes are partially suppressed by the Coulomb barrier:

$$\sigma(\gamma, total) = \sigma(\gamma, n) + \sigma(\gamma, p) + \sigma(\gamma, 2n) + \sigma(\gamma, np) + \ldots \quad (3)$$

Also, emission of more than one nucleon is associated with a higher reaction threshold energy. Table 1 lists possible photonuclear reactions on natural yttrium (which is a monoisotopic element composed of ^{89}Y), along with threshold energies, product half-life and their decay chains. All the reactions result in either stable or short-lived isotopes (the longest half-life is about three days) which decay into environmentally benign stable nuclides.

It should also be noted that bremsstrahlung converters inevitably produce considerable neutron flux. The neutron energy spectrum depends on the electron beam energy and converter material, but in general resembles a fission spectrum with a mean neutron energy of about 1–2 MeV. Low energy neutrons may be captured by (n,γ) reactions while neutrons with sufficiently high energy may induce threshold processes such as (n,p) or (n,α). Neutron-induced reactions are listed in Table 2. Buildup of strontium-90 (half-life of 28.9 years) can become an issue, especially for long production cycles. However, if the irradiation is limited to several weeks, ^{90}Sr activity will be insignificant and can be reduced even more by surrounding the yttrium target with low Z material and thus moderating neutrons.

2.2. Monte-Carlo simulations of the Y-88 production rates and yield

To accurately predict the yield, it is necessary to know all the parameters mentioned in Eq. (1). Most of them are straightforward, however the integral of the product of the ^{89}Y(γ,n)^{88}Y cross-section and photon flux is usually challenging to evaluate. We used MCNPX, a

Table 2
Products of neutron activation of yttrium.

Reaction	Product half-life	Energies of emitted gammas, keV	Decay chain
^{89}Y(n,γ)^{90m}Y	3.2 h	202(97%), 480(91%)	^{90m}Y→^{90}Y →^{90}Zr (stable)
^{89}Y(n,p)^{90}Sr	28.9 years	n/a (beta emitter)	^{90}Sr→^{90}Y →^{90}Zr (stable)
^{89}Y(n,α)^{86}Rb	18.6 days	1077(9%)	^{86}Rb→^{86}Sr (stable)
^{89}Y(n,α)^{86m}Rb	1 min	556(98%)	^{86}Rb→^{86}Rb→^{86}Sr (stable)
^{89}Y(n,γ)^{90}Y	2.7 days	1761 (0.003%)	^{90}Y→^{90}Zr (stable)