

Geological Probing in a Low-carbon Energy Future: *A new frontier for ionizing radiation?*

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ABSTRACT

The paper examines potential roles for ionizing radiation-based subsurface probing techniques, ubiquitous in the petroleum industry, to extract geological information essential for transition to a low-carbon energy future to mitigate climate change. These techniques in general come mainly in two tracks, radioactive source-based techniques used for initial characterization of geological formation and accelerator/generator-based techniques used to monitor fluids contained in it. Tests of generators/accelerator tools to replace radioactive source tools in the formation characterization phase show promise but also face considerable challenges; most petroleum industry practitioners have been reluctant to switch as a result. Could successful tests to support geological probing needed in a low-carbon future alter the dynamic and motivate the transition?

The present paper first briefly notes the basics of subsurface nuclear techniques and the state of generator-based alternatives to radioactive source tools used. It then reviews tested or proposed applications of nuclear techniques in geological probing needed in low-carbon energy transition, with an emphasis on generator-based techniques. Several necessary technological advances suggested for the latter are briefly noted.

INTRODUCTION

Ionizing radiation-based techniques play a major role in geological probing for hydrocarbon exploration and production (Ellis and Singer, 2007). Historically, use of ionizing radiation evolved in two tracks: 1) radioactive source-based tools to initially characterize a formation and 2) D-T generator tools with scintillators, to perform behind-casing spectroscopy and monitor changes in the formation fluid later in the life cycle of a well. Starting in the 1980's, accelerator-based tools have been tested to replace radioactive sources to obtain bulk parameters such as density and neutron porosity. The results have been mixed. On the other hand, D-T generator-based devices with advanced scintillators, developed recently utilizing both inelastic and capture gamma rays and thus providing a more complete mineralogy, are beginning to replace Am-Be source (n-gamma) capture spectroscopy tools that had been utilized since the mid-1990's. A survey of the state of accelerator-based well logging technology can be found in Badruzzaman (2023a).

To mitigate climate change, the Intergovernmental Panel on Climate Change (IPCC), early on, proposed three broad categories of low-carbon energy transition options, namely, 1) fossil fuels with carbon capture and storage (CCS), 2) nuclear power, and 3) renewables (IPCC, 2007). Drilling of wellbores and subsurface probing would be involved in all three options. However, probing of subsurface with nuclear for non-petroleum energy options applications is not new; some go back to the 1970's, for example in the Kilauea volcano in Hawaii (Keller et al., 1974). The reader will find more on ideas tested and novel ones being explored in Badruzzaman (2023b) and references therein.

In this paper we first briefly review nuclear logging basics and the state of source replacement to set the stage for discussing the subsequent discussion of nuclear techniques to probe the subsurface in the low-carbon energy options.

NUCLEAR LOGGING BASICS

In the *reservoir characterization* phase of a geological formation, two sets of logs (parameters vs. depth) are obtained. Bulk parameters, as natural gamma-ray, density, lithology, and neutron porosity are measured using

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ionizing radiation and fluid saturation is measured using electrical resistivity, while acoustic and NMR tools are often used to provide complementary parameters (Ellis and Singer, 2007). Both provide a porosity, but it is not as accurate as the density-based porosity; the lithology from acoustic tools is not as accurate as that from nuclear tools and neither acoustic nor NMR technique can determine mineralogy (Badruzzaman et al., 2015).

The second set of characterization measurements, *spectroscopy*, for mineralogy determination, is more recent. Two (n-gamma) capture spectroscopy tools using Am-Be sources and BGO crystals were developed (Herron and Herron, 1996; Galford et al., 2009) and utilized mainly in unconventional (shale) reservoirs. Neither can identify carbon and both are challenged in determining magnesium and aluminum. Herron and Herron (1996) construct an emulated aluminum using spectral yields of other elements, as a sum of SiO₂, CaCO₃, MgCO₂ and Fe. Obtaining clay from this approach becomes complex and requires different mixing rules that may vary across lithologies and wells. Often complicated, local, empirical, single- or multiple-tool interpretation models were needed.

In *reservoir monitoring* measurements, tools with D-T generators emitting 14-MeV neutrons for a duration of several microseconds and at least two scintillators are used to record two broad sets of data: 1) (n-gamma) spectra during burst and 2) gamma rays from thermal neutron capture vs. time after the source is turned off (Ellis and Singer, 2007). The burst spectra contain inelastic and capture components that are separated using special processing. The temporal variation of gamma rays from thermal neutron capture allows delineation between gas and liquids and, at high salinity, allows quantification of water volume fraction vs. oil volume fraction. Spectral data from carbon and oxygen determination of oil saturation directly. The spectral data can also determine the elemental mineralogy; the concept was the forerunner to the recently reported advanced (n-gamma) spectroscopy tools utilizing both inelastic and capture spectra for characterization, noted later.

RADIOACTIVE SOURCE REPLACEMENT FOR RESERVOIR CHARACTERIZATION

Replacing source-based tools used to generate density and neutron porosity logs has been of interest for over four decades. Both non-nuclear and generator-based alternatives have been tested. While non-nuclear techniques offer complementary data essential to fully characterize a formation, they have not sufficed in replacing nuclear-based parameters such as porosity, lithology, or mineralogy, by providing accuracies or attributes that radioactive source tools provide (Badruzzaman et al., 2015).

Nuclear-based alternatives tested for *bulk parameters* include neutron generators to determine the neutron porosity, and both LINAC-based high energy Bremsstrahlung and low-energy X-Ray sources, for density. The results have been mixed. On the other hand, D-T generator tools with advanced scintillators that can record both inelastic and capture spectra, similar to their monitoring counterparts, are beginning to replace Am-Be source spectroscopy tools that rely only on capture spectra. The reader can find details in Badruzzaman (2023a) and references therein. We briefly discuss these next.

Am-Be Source Replacement in Neutron Porosity Tools: Several generator-based neutron porosity tools have been proposed and several developed and tested. Between 1991 and 2000, two D-T generator-based neutron porosity tools concepts were proposed to replace the Am-Be source, one for wireline logging and the other for logging-while-drilling tools. The latter did well but the wireline tool often led to erroneous results. A D-D generator neutron porosity tool was recently reported for shallow well bores. A recent paper examined, using Monte Carlo simulation, the potential performance of tools with various proposed neutron generators relative to Am-Be source tools (Badruzzaman et al., 2019). Figure 1a and Figure 1b, respectively, (adopted from the cited reference) display, the neutron source spectra from an (α -n) dense plasma focus (DPF) accelerator and from three fusion generators.

Figure 2 displays the Near/Far counts ratio as the proxy of the porosity response. The proximity of neutron spectra to that of the Am-Be would determine the proximity of the neutron response to Am-Be response while the neutron yield of the generator will determine statistical precision and thus the logging speed. So, as seen in the figure, the porosity using the DPF accelerator neutrons would be identical to those using the Am-Be source in view of Figure 1(a). However, designing a DFP-based logging tool to fit the hardware in a logging tool would be complex. D-T neutrons with higher neutron energies exhibit a lower porosity sensitivity. D-D neutrons with lower energies show a

greater porosity sensitivity but this arises from the lower Far detector counts. This would result in lower Far counts and a slower logging speed.

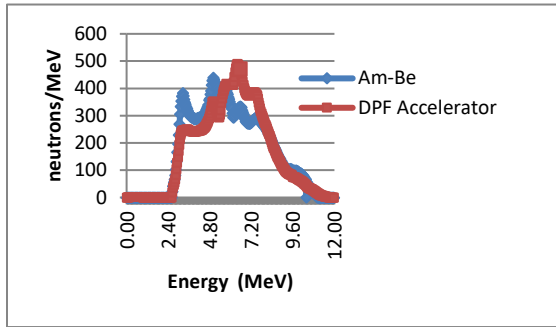


Figure 1a. Am-Be vs. DPF accelerator. Both utilize (α -n) reaction

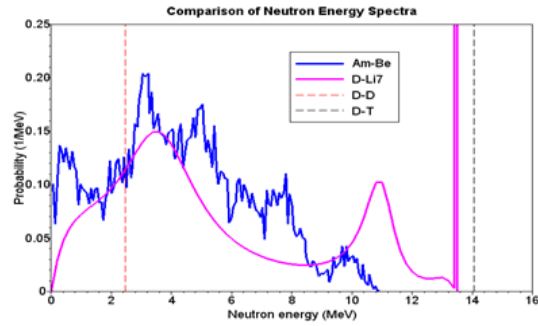


Figure 1b. Neutron spectrum from fusion generators

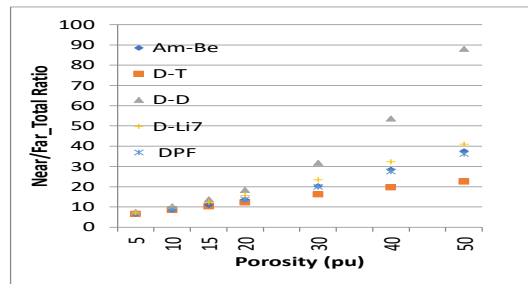
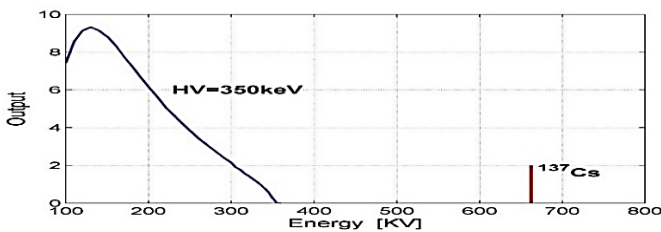


Figure 2. Neutron porosity response using various neutron generators vs. Am-Be source (Badruzzaman et al., 2019).

One unresolved issue is the proximity of the predicted porosity to legacy data. Several approaches have been suggested to normalize the neutron porosity from generators to match the legacy data. They are often complicated as discussed by Badruzzaman et al., (2019).

Cs-137 replacement in density tools: Successful design and field tests of a 3.5 MeV LINAC-based borehole density tool were reported in the mid-1980's (King et al., 1987). However, the tool was not commercialized due to several challenges including a downturn in the industry. The concept of using X-ray sources in general was recently revived with considerable promise (Badruzzaman, 2014; Simon et al., 2018). Badruzzaman (2014) showed that a 3.5 MeV LINAC would clearly provide a density response that closely represents the Cs-137 density as was reported by King et al., (1987) from the field test of their experimental tool.

Simon et al., (2018) used the 350-keV X-ray source shown in **Figure 3** vs. the Cs-137 source in standard density logging tools.



Note the much greater photon flux from the X-ray source.

Figure 3. 350-keV X-ray spectrum vs. 662-keV line from Cs-137 (Simon et al., 2018).

As shown in **Figure 4**, the authors replicated the linear density variation reported in Badruzzaman (2014) using a 3.5 MeV Bremsstrahlung source.

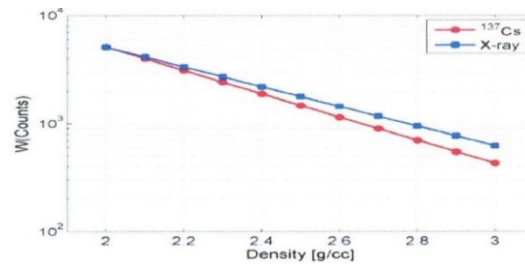


Figure 4. Density response 350-KeV X-ray density tool vs. a Cs-137 source tool (Simon, et., 2018).

However, the low-energy X-ray density tool would exhibit a substantial photoelectric signature, unlike the 3.5 MeV Bremsstrahlung concept. So, a large photoelectric (PE) correction would be needed to obtain the density. Simon et al., (2018) addressed this by acquiring a large experimental database in a variety of formation and mud conditions with differing density and PE value, and utilizing a non-linear interpolation to obtain the density under a given condition. So, in principle, they needed no explicit photoelectric ‘correction.’ Alternatively, Huawei et al., (2022), reported a two-parameter algorithm for a similar (350-keV) X-ray tool to correct for the large photoelectric effect. An unresolved question is: could the photoelectric ‘correction’ needed to determine the density of the formation be used to predict its lithology?

D-T-generator-based advanced mineralogy-Alternative to Am-Be-based capture spectroscopy tools: While generator-based tools have struggled for acceptance in characterization measurements, D-T generator-based spectroscopy tools with advanced scintillators had become ubiquitous in reservoir monitoring (Badruzzaman, 2023a). These were the forerunners of D-T generator based of mineralogy tools for the characterization phase reported by Pemper et al., 2006 and Radtke et al., 2012. The availability of both inelastic and capture data for some elements significantly improves their precision, accuracy, and interpretation consistency, particularly of magnesium, a key element for differentiating calcite from dolomite. Only inelastic data provides carbon, thereby allowing determination of inorganic carbon from carbonate minerals, and obtaining total organic carbon (TOC). TOC is essential for evaluating many unconventional formations such as shales.

Radtke et al., (2012) utilized LaBr₃ scintillator that has a much better energy resolution than other scintillators used for (n-gamma) spectroscopy in well logging. Additionally, this scintillator retains its robust temperature behavior, such as light output, at higher temperatures where other scintillators used in the industry, such as NaI and BGO, would degrade. These tools are beginning to replace Am-Be source-based spectroscopy tools. **Figure 5** displays a field example of testing their D-T generator spectroscopy of tool by Radtke et al., (2012) in a well in North Dakota. Note the particularly good agreement with core achieved for Al, Mg, and TOC that would have been problematic with an Am-Be (n-gamma) capture spectroscopy tool.

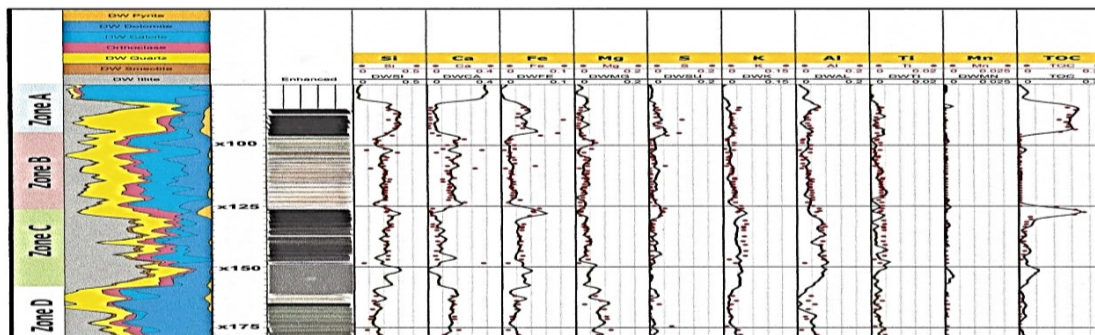


Figure 5. Elemental weight fractions and the total organic carbon (TOC) from gamma ray spectroscopy vs. core samples from a well in North Dakota (Radtke et al., 2012)

GEOLOGICAL PROBING IN LOW-CARBON ENERGY OPTIONS

We previously noted the three IPCC-proposed broad categories of low-carbon energy transition options: 1) fossil fuels with carbon capture and storage (CCS), 2) nuclear power, and 3) renewables (IPCC, 2007). A recent paper assessed these in some detail (Badruzzaman, 2023b). We briefly illustrate the discussion next.

1) Fossil fuels with carbon capture and sequestration (CCS): monitoring sequestered CO₂

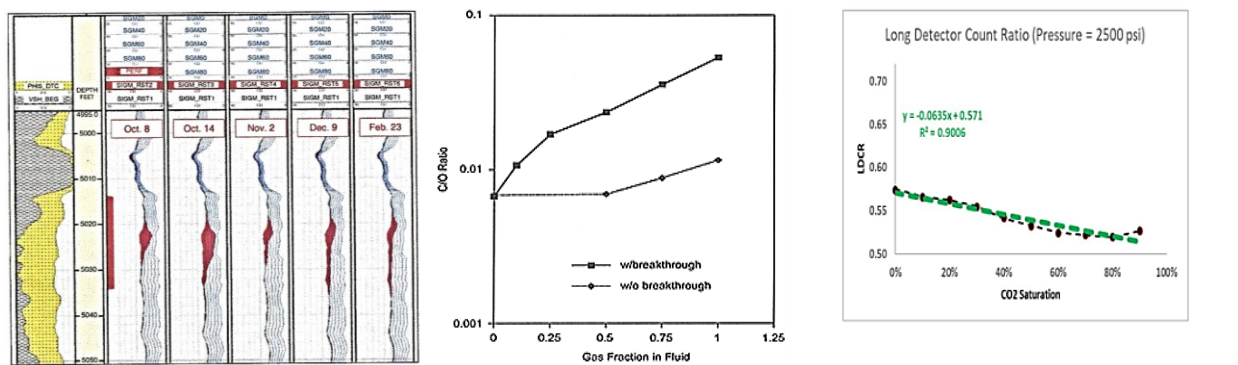
There are several CCS projects underway to mitigate climate change. Both depleted reservoirs and virgin aquifers are under consideration. One of the largest is in Australia initiated by 2019 by a major oil company to inject 100 million metric tons of CO₂ over the life of their CCS system (Gorgon, 2022). The CCS technology is well known to the petroleum industry since CO₂ injection has been utilized to push hydrocarbon liquids out for enhanced oil recovery. However, the volumes of CO₂ that need to be injected to mitigate global warming are orders of magnitude greater than what the industry is familiar with. For example, the world's CO₂ emission will go from 34.3 billion metric tons in 2020 to 42.8 billion metric tons in 2050 (EIA, 2021). CCS projects would arise across the world and require a careful assessment of the geology using seismic and petrophysical techniques including nuclear, to understand properties such as geological sealing, etc., to ensure that the CO₂ remains sequestered.

However, the potential for CO₂ leakage exists and thus the movement of injected CO₂ needs to be monitored. D-T generator based techniques used to monitor fluid movement in hydrocarbon reservoirs were assessed for their ability to monitor injected CO₂ (Sakurai et al., 2006; Badruzzaman et al., 2002; Quintero et al., 2022). **Figures 6 (a), (b), and (c)** collectively summarize the results from the three references. We briefly discuss the conclusions of each.

Sigma technique: Sakurai et al., 2006 conducted a pilot experiment in an aquifer in Frio Brio formation in Texas with support from the US DOE using a dual-detector D-T generator tool. **Figure 6(a)** clearly shows that the evolution of Sigma, the pulsed neutron capture decay coefficient, is related to CO₂ injection. They had also tested the C/O ratio technique, but the data was too noisy for a definitive conclusion.

C/O ratio technique: Badruzzaman et al., 2002 postulated that if there are other gases present, the Sigma technique may not be unambiguous and thus performed a Monte Carlo simulation of the response of a similar dual-detector D-T tool inserted in an aquifer to construct the spectral carbon/oxygen (C/O) ratio. Their results in **Figure 6 (b)** showed that with no breakthrough, the C/O ratio would be an indicator of injected CO₂ only at very high CO₂ saturation. However, with breakthrough, CO₂ can be delineated at much lower saturations.

Inelastic/capture counts ratio: If CO₂ is injected in depleted gas reservoirs, it would be displacing residual methane. and the C/O technique would be unlikely to differentiate between methane (CH₄) and CO₂. Quintero et al. (2022) as seen in **Figure 6 (c)** showed that the ratio inelastic counts/late time capture counts at the farthest detector could be used, instead. The authors noted that response would be sharper at higher pressures.



(a) PNC Sigma

(b) Spectral C/O ratio

(c) Inelastic/late capture counts ratio

Figure 6. CO₂ monitoring potential using D-T generator-based techniques.

(ii) Nuclear Power:

Burial site characterization: Nuclear power will continue to supply a significant fraction (10%) of electricity (EIA, 2021) as the world attempts to transition away from fossil fuels and seeks to adopt solar and wind that are intermittent. Approximately, 390,000 metric tons of spent fuel were generated between 1954 to the end of 2016 from nuclear-based electricity production (IAEA, 2018). This volume is much smaller than the amount of coal ash generated in the US alone, *per year* (approximately 100 million tons). However, disposal of high-level waste (HLW), is of much interest with geological burial being the primary option. While host rocks vary across national programs, almost all national waste disposal programs are heading towards using bentonite clay for buffer. In general, geological burial would involve drilling and formation characterization. Two recent papers, one on the Swiss HLW project and the other from the German HLW project, have demonstrated the usefulness of conventional petrophysical techniques such as acoustic, electrical, NMR, and source-based density/PE and neutron methods (Garrard and Desorches, 2022; Meier and Strobel, 2022). Use of radioactive source tools could be dicey in that a source stuck downhole would be problematic. Thus, a generator-based neutron porosity tool would be preferred. It would be instructive to test the X-ray tool noted previously to obtain the density. Additionally, could its large photoelectric effect be a lithology indicator?

Swiss researchers also tested the Am-Be based (n-gamma) capture spectroscopy technique to compute elemental yields. However, a D-T generator based mineralogy tool would be preferable since, as discussed previously, it can determine presence of carbon, magnesium, aluminum, etc. Aluminum is a key element of most clays and magnesium is a key component of some bentonites (Allen and Wood, 1988). These elements together with sodium, potassium, manganese, etc., are needed to reconstruct the mineralogy in detail.

Monitoring buried radioisotope: While radioactive waste disposal, especially, of HLW, into geological formations is mostly in the concept phase, it may be timely to consider the need to monitor potential leakage of buried radioisotopes and examine if nuclear logging devices would be useful. Several authors cited in Badruzzaman (2023b) have tested nuclear logging techniques to locate radioisotopes in contaminated radioactive sites. Two papers were on tests performed at the Hanford site in Washington State to locate human-made radioactivity leaking from the buried drums. One relied on excess radioactivity measured to assess leakage. The other, by Ellis et al., (1995), utilized an advanced spectral gamma-ray tool calibrated specifically to delineate gamma rays from Co-60 and Cs-137 vs. naturally occurring gamma ray. They used a D-T generator based neutron tool to measure the hydrogen index and Sigma. These authors reported results from three monitor wells at Hanford, located within a few miles of one another. They noted the following.

The total gamma ray counts indicated the gross contamination at each well relative to the natural gamma-ray (GR) background. The spectroscopy data identified the specific isotope that was the cause of the contamination and its location vs. depth. In one well, it was entirely Cs-137, in the second well it was primarily Co-60, while in the third well, it was a mixture of Co-60 and Cs-137. Additionally, as displayed in **Figure 7**, the Cs-137 in the third well was at a shallower depth than Co-60.

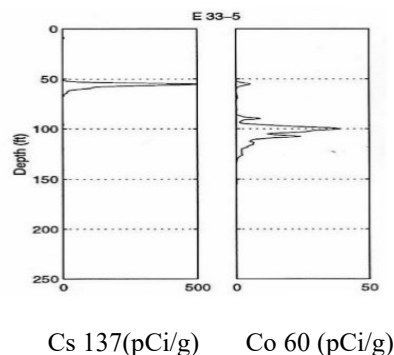


Figure 7 Contamination in Well E-33-5 at Hanford measured by Ellis, et al., 1995.

Note that the upper zone, just below 50 ft., is highly contaminated with about 500 pCi/g of Cs-137 at the maximum. A nearly 50-ft zone centered around 100 ft. is contaminated with Co-60 at a level not exceeding 40 pCi/g.

Moisture content: Ellis et al., (1995) utilized a D-T generator neutron porosity tool, instead of the conventional Am-Be source tool, to quantify the moisture content. A moisture content map would be indicative of presence of water in the subsurface and would allow monitoring of migration of water to predict pathways for contaminant migration. **Figure 8** displays an example of simultaneous moisture content and contamination location in Well W15-7 (Ellis et al., 1995). The observations are noted in the box

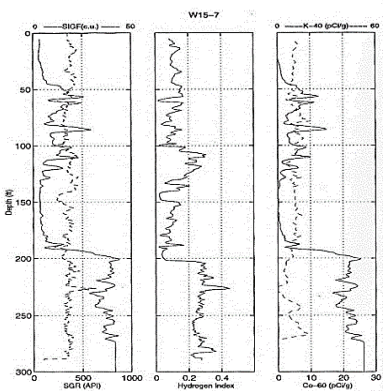


Figure 8. Simultaneous moisture and contamination measurement.

- Left panel: Total GR (API units; the Sigma (capture units). Note expanded GR scale due to high gamma ray counts in the formation.
- Middle panel: Hydrogen index
- Right panel: K-40 (from natural radioactivity) and Co-60 concentration in picocuries per gram (pCi/g).
- Left panel: Layers of contamination between 50 ft to 100 ft with a jump at about 200 ft; coincides with the water table seen from the middle panel.
- Right panel: Co-60 (vs. the K-40) source of high GR in the water table and the region below it.
- Left panel: SIGF (from thermal neutron decay in generator tool) in water located had a low salinity. Am-Be tool cannot supply this information.

(iii) Locating Strategic Minerals for “clean” Energy Options

It has been reported that clean (CO₂-free) energy options require much larger amounts of minerals than fossil fuel energy systems (IEA, 2021; Dominish et al., 2019). As shown in **Figure 9** adopted from IEA (2021), an electric vehicle (EV) requires six times the mineral inputs of a conventional internal combustion engine vehicle and an onshore wind plant would require nine times more mineral resources than a natural gas-fired power plant.

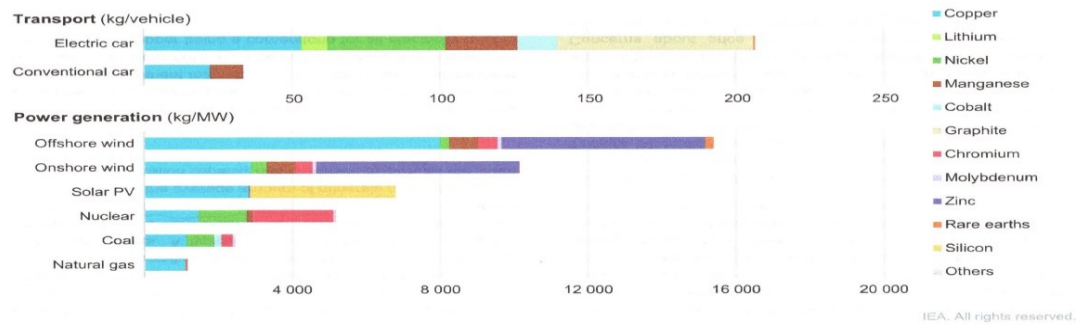


Figure 9. Total demand for selected minerals vs. energy source options (adopted from IEA, 2021)

Dominish et al., (2019) had shown that requirements such as those in Figure 9 would lead to a several-fold growth, by 2050, in the demand for many of these minerals as energy transition proceeds, and this growth in demand is unlikely to be met by recycling. Thus, the current practice of extracting these minerals from near-surface sources using open-pit mining would not suffice² and one may have to extract these from further depths. However, indiscriminate drilling would be environmentally damaging and unsustainable. Thus, it suggested that mining industries explore the (n-gamma) mineralogy techniques discussed previously in this paper to surgically locate these

² In addition to being environmentally damaging, such practices have led to child labor abuses (Amnesty International report, Index: AFR 62/3183/2016(2016)).

minerals in the subsurface. Table 1 displays several earth elements that various gamma spectroscopy methods can identify.

Table 1. Some earth elements determinable by gamma-ray spectroscopy techniques

Element	Technique		
	n-gamma capture	n-gamma inelastic	Natural gamma ray
Al	yes	yes	
Ba	yes	yes	
C		yes	
Ca	yes	yes	
Cl	yes		
Cu	yes		
Fe	yes	yes	
Gd	yes		
H	yes		
K	yes		yes
Li		yes	
Mg	yes	yes	
Mn	yes		
Na	yes		
Ni	yes		
O		yes	
S	yes	yes	
Si	yes	yes	
Ti	yes	yes	
Th			yes
U			yes

Note that spectral techniques will be able to locate several of the elements noted in Figure 9. However, to be quantifiable, their concentrations should be ‘sufficient;’ the author is assessing this aspect.

(iv) Other Low-carbon Energy Applications.

Geothermal: As noted previously, radioactive source logging tools have been tested to assess geothermal reservoirs. Clearly, from the above discussions we can conclude that generator-based tools can be used here, instead. Consequently, use of risky source-based tools in these fragile formations must be reconsidered.

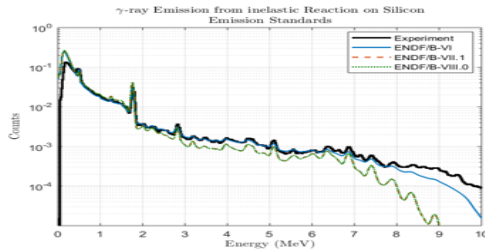
Delineating natural hydrogen vs. methane: As an analysis in Badruzzaman (2023b) notes, generator-based neutron porosity tools should be able to delineate hydrogen from methane when both occur in the same formation.

TECHNOLOGY ADVANCE NEEDS

Fazio et al., Eds., 2020 had recommended a number of innovations in the use of accelerators including in subsurface applications. Badruzzaman (2023a) elaborated specifically on the latter. Briefly, some of the desired major advances in these applications were as follows.

1. Multi-purpose, higher yield, compact neutron generators: The desired higher neutron yields would be 10^7 n/s for D-D and 10^9 n/s for D-T vs. their current nominal yields of 10^6 n/s and 10^8 n/s, respectively. D-D tools with higher output may be able to compete with Am-Be tools for neutron porosity without losing depth-of-investigation sensitivity. Higher yield D-T generators would allow a better resolution of elemental concentration in low-concentration mineralogy. Higher neutron yields will allow faster logging with both.
2. Compact X-ray or mono- or nearly mono-energetic gamma sources: These would allow a direct replacement of Cs-137 for density and better density imaging. Density imaging of geological strata using a Cs-137 density is currently rudimentary.
3. Advanced fast scintillators, radioactivity-free, with multimodal imaging capability: Faster scintillators allowing detections in tens of nanoseconds could permit delineation of inelastic spectra with almost no capture correction. Multimodal imaging will allow density and neutron imaging to visualize rock strata, fluid flow imaging as a permeability indicator, and alpha particle imaging for directional information. Absence of radioactivity would allow recording of natural gamma-rays with advanced scintillators; internal radioactivity in LaBr₃ precludes that.
4. Diagnostic & PHM capability: These would predict, a priori, potential generator failures that can be catastrophic in expensive drilling regimes such as offshore, or hard to access areas.
5. Advanced simulation & visualization software: These will augment the benefits already derived from using Monte Carlo radiation transport techniques. These will allow exploration, a priori of novel measurement concepts, and speed-up design, validation, and field-test of new devices.

6. Improvements in nuclear cross-section libraries: In nuclear logging, Monte Carlo simulations are essential in designing, calibrating, and complementing measurements. However, (n-gamma) cross sections of several of the light elements needed for the simulation may be inadequate. **Figure 10** displays this for silicon (n-gamma) inelastic cross section as assessed by Mauborgne et al., 2017. Similar assessment is needed for elements present in strategic minerals.



Calcium, another element of interest in geological probing, was also poor. Based on their tool response, the authors of the figure decided against use of the later evaluation of silicon inelastic.

Figure 10. (n-gamma) inelastic scattering cross section of silicon at various ENDF evaluations (figure courtesy of Marie-Laure Mauborgne of Schlumberger)

SUMMARY

Nuclear techniques would be extremely valuable in geological probing to support low-carbon energy options. While radioactive source-based tools would still be useful in the characterization phase, generator/accelerator-based tools would offer a significant advantage by supplying additional information and would not have the safety/security risks associated with radioactive sources. However, several hardware and software advances are needed to make the machine source-based tools as sturdy as radioactive source-based tools while providing additional information.

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