Technically Enhanced Naturally Occurring Radioactive Materials in Coal Seam Gas Production - A Review

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Abstract

Coal seam gas (CSG) is an unconventional gas production process as global demand for cleaner energy. It is important to understand the inherent radionuclides in CSG and their enhancing mechanisms for better naturally occurring radioactive materials (NORM) management. The enrichment of such radionuclides is a potential risk to health and environmental exposure. This review endeavours to explore the sources of NORM, as well as potential technical enhancement routes for these radionuclides and their progenies in CSG production.

1 Introduction

It is well known the presence of trace amount of naturally occurred radioactive materials (NORM) in coal and overburden. The NORM from coal can be further concentrated during coal mining and its further use. For example, fly ash from coal combustion is a typical route for technically enhanced naturally occurring radioactive materials (TENORM) of coal (Boukhair, et al., 2016). The management strategy and regulation for the coal industry are well established (Government of Western Australia, 2010) (Australian Radiation Protection and Nuclear Safety Agency, 2005). Different to the coal mining process, coal seam gas (CSG) is a non-conventional gas extraction process, in which CSG (mostly methane) and formation water are extracted while coal and overburden largely remain undisturbed in the strata.

The management of conventional oil and gas production was well established in the 1980s (International Association of Oil & Gas Producers, 2008) (UK Department of Energy & Climate Change, 2014). In the United States, it is reported that about 5 million cubic feet (141,000 cubic meters) of accumulated sediment with NORM concern each year (Thompson, et al., 2015). The NORMS, which are inherent in the reservoir and strata rock, are brought to surface with gas, water and liquid hydrocarbon (Garner, 2017) (Bou-Rabee, Al-Zamel, Al-Fares, & Bem, 2009). They are further concentrated in the waste stream, which poses greater environmental and personal health risks (Mously & Cowie) (URS, 2005).

As an evolving unconventional gas process, the risk of the NORM in CSG production is not well understood. The author believes that the experience and lesson learnt in conventional oil and gas industry are invaluable for CSG (Smith, 1992). However, natural radionuclides and their decaying progenies are present and their enhancing mechanisms in CSG need further exploring (Khan & Kordek, 2014), which may lead to potential excessive personal exposure and accumulation of regulated radioactive waste (CSIRO, 2017). It is critical to understand the origin of radionuclides and their enhancing mechanisms for better NORM management.

2 Radionuclides in coal and CSG

It is reasonable to conclude that the NORM in CSG comes from coal and overburden's radionuclides and their progenies. Coal normally contains trace amounts of ^{238}U, ^{235}U, ^{232}Th and ^{40}K. These radionuclides are associated with minerals in the coal, such as sulphides or occur in the minerals making up the coal formation (McOrist & Brown, 2009). The average concentration of U and Th is studied in coal from the Bowen Basin, Queensland Australia(Boyd, 2004). Apparently, the concentration depends on the type of coal, geological formation, etc. The trace elements of Th and U in coal are listed in

Element	World Coal Average	Crustal Average	Bowen No.2 Wtd Ave Conc	vs world coal	vs crustal concentration
Thorium	0.5 - 10	5.8	5.73	Average	1.0
Uranium	0.5 - 10	1.6	1.16	Average	0.7

Table 1 (PARAMI, 2008).

The IAEA report identified ^{238}U, ^{232}Th, ^{210}Pb, ^{210}Po, ^{228}Ra, ^{226}Ra and ^{40}K as major radionuclides in the Australian coal. The concentration range is coincident to the CSIRO report (CSIRO, 2017)

Table 1 Th and U concentration in coal

Element	World Coal Average	Crustal Average	Bowen No.2 Wtd Ave Conc	vs world coal	vs crustal concentration
Thorium	0.5 - 10	5.8	5.73	Average	1.0
Uranium	0.5 - 10	1.6	1.16	Average	0.7

2.1 Uranium

^{238}U is one major NORM source in the coal. Uranium minerals usually have very low solubility in water and hydrocarbon liquid. Therefore, it will only be present in drill cuttings and sometimes found in sludges and sediment. However, uranium is usually the parent of major coal NORM progenies. (Thompson, et al., 2015).

2.2 Thorium

^{232}Th is another NORM source of coal. Similar to uranium, it also has low solubility in both water and liquid hydrocarbon. It will be present in drill cuttings and sludges and sediments. However, the concentration is rather low (Thompson, et al., 2015).

2.3 Radium

^{228}Ra and ^{226}Ra are the decay products of ^{232}Th and ^{238}U individually. Radium mineral has high solubility in formation water and less solubility in liquid hydrocarbon. They are preferentially present in all produced water stream (Leopold, 2007). Radium is a source of external radiation dosage. Radium in water may drop out and co-deposit as scale in equipment (Chambers, 2013).

2.4 Radon

Radon is a gaseous component. It presents in coal and overburden. Radon is a decay progeny of radium and parent of progeny in further reactions (Thompson, et al., 2015). Radon may pass multiple process barrier and covert to ^{210}Pb, ^{210}Bi, and ^{210}Po as particulate matters. Radon is highly soluble in liquid hydrocarbon as well (Leopold, 2007). Since little hydrocarbon condensate is present in CSG, most of the radon is either in gas stream or in the produced water stream.

Radon also has significance in the CSG liquification process. Its boiling point (-61.8^{o}C) lies between propane(-42.1^{o}C) and ethane(-88.6^{o}C). Consequently, Rn is often found enriched in propane stream (Leopold, 2007).

2.5 Lead, Bismuth and Polonium

Radioactive Pb, Bi and Po are progenies of radon and thoron decay. They are often plated out in tubing, piping and downstream equipment. In the natural gas pipeline, it is normally found in pigging trash. These radionuclides are difficult to be detected externally. The only certain operations, such as internal inspection, pipeline pigging, may lead to personal and environmental exposure.

3 Radioactivity in coal and CSG

The radioactive level of coal is the overall results of the radiochemical reactions of these radionuclides and their decay progenies. The Queensland NORM guidance indicated that ^{232}Th (Thorium -232), ^{238}U (Uranium-238) and their decay progenies are the major radioactivity concerns in coal mining. The major decay chain and their progenies of U^{238} and ^{232}Th are illustrated here (McOrist & Brown, 2009). According to the code of practice, the activity concentration of 1 Bq/g is the threshold for NORM containing uranium and thorium (Queensland Government Department of Mines and Energy, 2008). ^{40}K is also managed for personal exposure in the same guidance, although it has less significance.

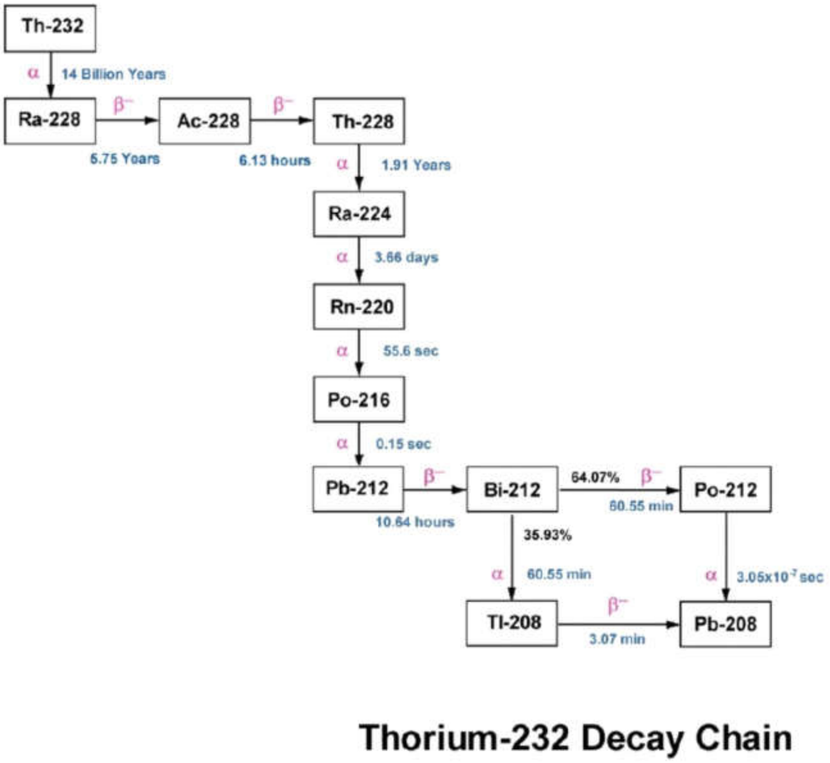

Figure 1 ^{232}Th decay chain, sourced from (McOrist & Brown, 2009).

3.1 Common NORM decay chain in coal

Extensive radioactive level survey for coal has been performed in Australia since the 1980s. It is identified that the levels of radioactivity in Australian and international coals were similar and are considered to be low (DALE, 2006). The average NORM level in Australian coal is 370 to 400 Bq/kg, only equivalent to 1/3 of common garden soil radioactivity. The radioactivity of the main contributing radionuclides is listed in Table 2

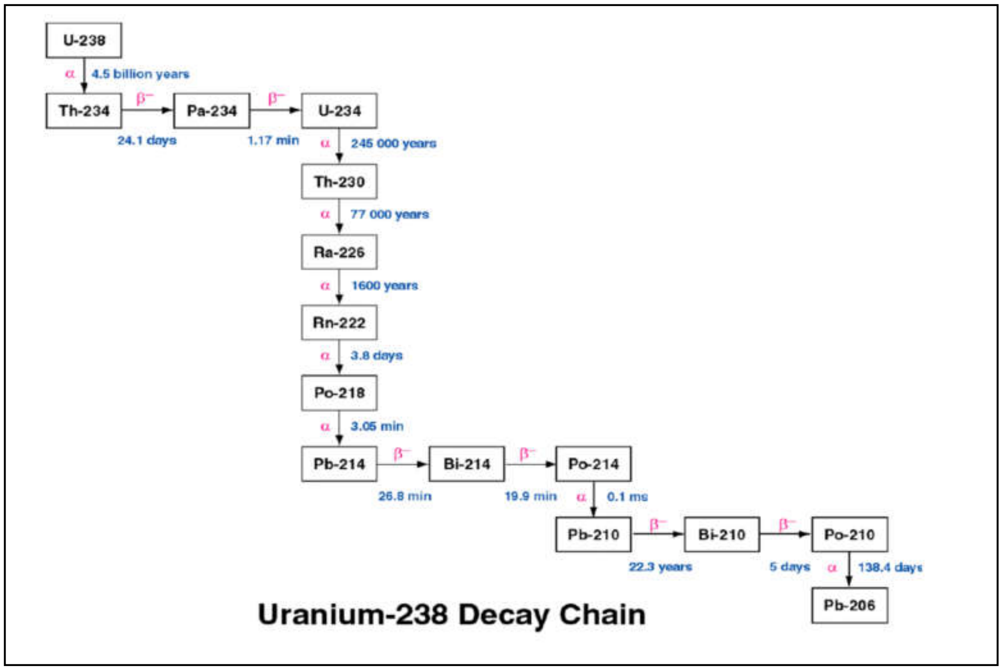

Figure 2 238U decay chain, sourced from (Brown & McOrist, 2009)

Table 2 Radioactivity of radionuclides in 1981 Australian coal survey (McOrist & Brown, 2009).

Activity, Bq/kg	^{238}U	^{226}Ra	^{210}Pb	^{228}Th	^{228}Ra
Port Augusta, SA	60	41	60	85	87
Bunbury, WA	10	12	20	30	32
Kwinana, WA	30	7	30	15	21
Gladstone, QLD	14	9	60	18	18
Callide, QLD	24	14	38	33	33
Collinsville, QLD	27	19	46	21	44
Swanbank, QLD	20	13	21	24	25

It is evident that all reported radionuclides are from ^{238}U and its decay progenies and there is no radon reported. This is understandable that both ^{235}U and ^{232}Th, as well as their progenies have a relatively short half-life and difficult to be captured and analysed in the coal sample. Australian radiation protection safety guide even states that ^{235}U decay chain and thoron (^{220}Rn) from Thorium (^{232}Th) contribute little significance in the NORM level of oil and gas, although saturated Radium(^{226}Ra, ^{228} Ra and ^{224}Ra) and Radon (^{220}Rn) dissolved in the formation water are the primary sources of NORMS (Australian Radiation Protection and Nuclear Safety Agency, 2008).

Table 3 IEAE radioactivity of each radionuclide in Australian coal

Bq/kg	^{238}U	^{226}Ra	^{210}Pb	^{210}Po	^{228}Th	^{228}Ra	^{40}K	Total activity
Australian Coal	8.5~47	11~64	20~ 33	16~28	11~169	19~24	23~140	43-1025

Published IEAE data showed a similar range of radioactivity for each radionuclide in

Australian coal. The results are listed in Table 3 IEAE radioactivity of each radionuclide in Australian coal

(CSIRO, 2017)

4 Radioactivity and potential technical enhancement in CSG production

CSG production is an unconventional gas production process. The process is illustrated in the following diagram (Figure 3). The mobility of radionuclides in CSG production is depending on the fluid chemistry in each production stage, which could be varied significantly. It is apparent that its technically enhancing path is different in each production stage.

4.1 Drilling

CSG well is well-known for its low yield rate, ranging from 0.5~ 5 TJ/day. Therefore, a large quantity of well needs to be drilled to sustain production capacity. During the drilling stage, all radionuclides in the borehole, including coal and overburden will be brought over to surface. Normally immobilized uranium and thorium isotopes will inevitably be carried with drilling cuts. Some radionuclides are likely enriched in the mud. As drilling fluid is continuously cycled down the drill string and back to the surface and reused from well to well, it gradually takes on the salinity and the radioactivity of the formation water. Moreover, as the drilling fluid saturates and coats the drill cuttings, the characteristics of the rock will be dominated by that of the drilling fluid and brine water (Thompson, et al., 2015). It is reported that ^{226}Ra is preferentially dissolved in brine with high chloride under temperature and pressure.

It was reported that the radionuclide concentrations were higher in horizontally drilled wells than those typically seen in conventional vertical bores. This was likely due to the greater surface area of exposure between the drilling fluids and the regions of the formation elevated in radionuclide concentration (Thompson, et al., 2015).

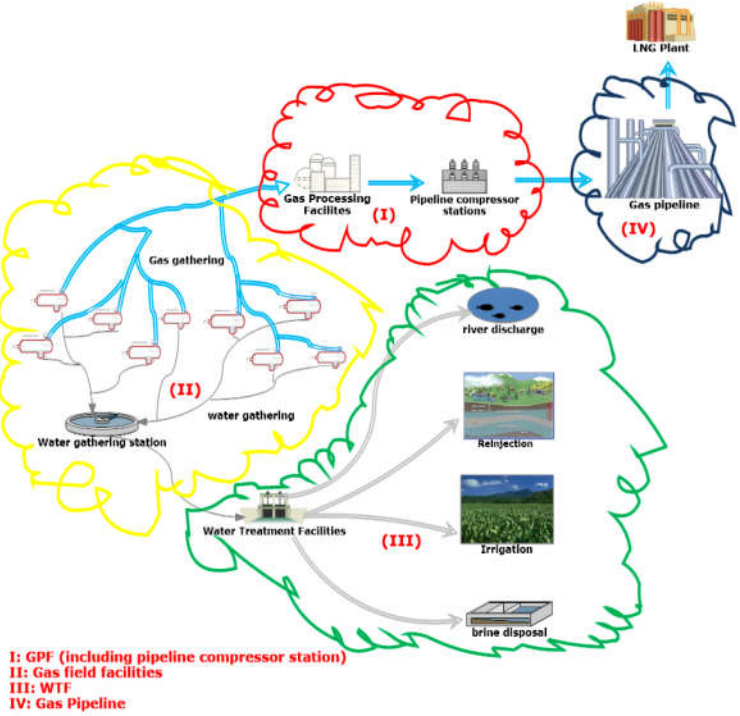

Figure 3 CSG production: process illustration

4.2 Field gathering and water treatment

Since gas is tightly bound to the coal and surrounding strata by hydraulic pressure and adsorption, de-watering is usually the critical step for the CSG production. The formation water and gas produced in each well are collected and transported through a field gathering system for further treatment. At this stage, U and Th, which have low solubility, are usually retained in the coal and overburden. Only high solubility radionuclides, mainly ^{226}Ra and its decay progenies, are carried by CSG formation water and gas as the primary source of NORMS (Varskog & Gellermann, 2012). Similar to conventional oil and gas production (^{222}Rn), a non-polar noble gas, may dissolve in produced water (Hylland & Eriksen, 2013). Rn is also carried in the gas stream.

The NORM in the coal formation water is well recognized (Skubacz, Michalik, & Wysocka, 2011). In CSG water stream, the scale is a major NORM enhance mechanism, it may form due to change of pH, temperature and pressure. The radionuclides including Radon decay products (^{210}Pb and ^{210}Po), along with sulphate and carbonate, precipitate as scale or sludge in pipes and related equipment (EL-Kameesy, Diab, Ramadan, & Megahid, 2017).

Accumulated Sediments in tank bottoms and sludges is normally considered as high potential for NORM, although there are no data available on CSG production. However, in conventional oil production, it is estimated that 5 million cubic feet (141,000 cubic meters) of accumulated sediment with NORM concern each year (Thompson, et al., 2015).

RO brine is another route of NORM enhancement. In Australia, stringent environmental regulation requires CSG produced water to be further treated before its discharge and/or beneficial utilization (Khan & Kordek, 2014). Reverse osmosis (RO) technology is the current industry standard practice of CSG produced water treatment (GasFields Commission Queensland, 2014). The NORM has the potential to be enhanced in the RO's brine stream.

4.3 Gas dehydration and transportation pipeline

CSG is dehydrated and transported through a pipeline system. The only potential NORM accumulation in this stage is Radon and its progenies. Although both ^{222}Rn and ^{220}Rn are potential presents, ^{220}Rn has a reasonably short half-life of 56s and of little significance in pipeline NORM. As a gaseous component, once ^{222}Rn passes through the compression station, its progenies can be settled and accumulated in the preferred location as particulate matters. This is the common reason that accumulation of ^{210}Pb and ^{210}Po in downstream piping and equipment. They may plate on gas valves, pipeline and accumulate at the particulate filter. In the aged pipeline, pigging trash is highly susceptible to high NORM activity due to the presence of ^{210}Pb and ^{210}Po (Varskog & Gellermann, 2012).

The overall presence of radioactive elements in different stages of CSG production is summarised in Table 4

Table 4 Potential presence of ^{238}U and its progenies in CSG production

	Potential presence of ^{238}U and its progenies in CSG production
Coal and Overburden	^{238}U; ^{226}Ra; ^{210}Pb
Drilling muds	^{238}U; ^{226}Ra; ^{210}Pb, ^{222}Rn
Drilling equipment	^{226}Ra; ^{210}Pb,
Field gathering Pipeline	^{226}Ra; ^{210}Po, ^{210}Pb
Produced water	^{226}Ra; ^{210}Pb, ^{222}Rn
Gas facility	^{210}Po, ^{210}Pb, ^{222}Rn
Gas pipeline	^{210}Po, ^{210}Pb, ^{222}Rn

5 Conclusion

The source of NORM in CSG and its technical enhancing mechanisms are explored. It is found that major contributors are ^{238}U and its decay progenies while ^{235}U and ^{232} Th have little contribution. The U and Th are only likely to present in drilling cuts and drilling mud, due to their low solubility. Radium and Radon are widely existing in the produced water stream. Radon has particular significance in CSG production: passing compressor station, the gaseous radionuclides can further decay into ^{210}Pb and ^{210}Po. These particulate matters are plated out in downstream pipeline and equipment. ^{222}Rn may enrich in the propane stream of the liquification plant due to the similar boiling point to that of propane and ethane.

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