HEAVY WATER REACTOR MODERATOR: ARE CHANGES TO DESIGN REQUIREMENTS NECESSARY?

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Abstract

Until the recent past, moderator chemistry has remained interesting but unexciting. Over the past two to three years moderator chemistry has become more interesting and far too exciting! This paper will discuss issues that have occurred and potential changes to design requirements of components, in order to keep moderator chemistry interesting rather than exciting.

1. Introduction

Since the early years of CANDU reactor unit operation, moderator chemistry has been, and continues to be, extremely important. Other than the requirement to efficiently thermalise neutrons and be a medium for soluble poison, moderator water has to be kept pure to avoid elevated dissolved deuterium. This is because the deuterium will end up in the moderator cover gas (Henry's Law). Should the deuterium concentration in moderator cover gas reach or exceed 4% by volume, the units(s) must be shut down in order to avoid the possibility of a deflagration or detonation event. Years of experience have taught us how to minimise elevated deuterium in moderator water and therefore moderator cover gas.

NPD-2, Douglas Point and Pickering "A" reactors were designed to use boron (as boric acid) as the soluble poison. The Bruce, CANDU-6, Darlington and Pickering "B" reactors have been designed to use both gadolinium and boron as soluble poisons. Boric acid and gadolinium nitrate have historically been the compounds of choice.

2. Deuterium

Gadolinium, as the nitrate salt, is used primarily due to its high solubility in the liquid injection shutdown system (LISS). Unfortunately the nitrate ion interrupts the recombination of deuterium and oxygen formed from radiolysis; this results in a surfeit of deuterium dissolved in the moderator water. Sulphate does not have the same effect as nitrate, consequentially that there is no buildup of dissolved deuterium. Gadolinium sulphate, however, is not as soluble as gadolinium nitrate and as such cannot be used for LISS in the current configuration.

The firing of shutdown system number "2" and injection of gadolinium nitrate from LISS, whilst the reactor is at power, results in a jump in dissolved deuterium. This increase in could be eliminated if gadolinium sulphate were used in place of gadolinium nitrate. A redesign of LISS may well be required to compensate for the lower solubility, therefore concentration, of gadolinium as the sulphate. Of course a review of assumptions and conservatisms with respect to LISS availability and duty may result in minimal redesign of LISS.

In 1997 gadolinium sulphate was used for the second time in a power reactor in Bruce Unit 7. The first use had been a test of gadolinium as a poison the Douglas Point reactor more than a decade earlier.

Currently the Bruce "B" units are the only CANDU plants using gadolinium sulphate. This salt (specifically the sulphate ion) has allowed excellent control of dissolved deuterium during all reactor power levels. Low dissolved deuterium results in lower transfer rates of deuterium into the cover gas, assuming all other parameters are normal. Whilst this low rate of transfer minimizes the rate of build-up of deuterium in the guide tube extensions it does not eliminate such build-up.

The turn of the millennium saw explosions at two boiling water reactors. There was a proposed explanation for the explosion at Brunsbuettel [Germany 14 Dec 2001], whereas that a month earlier at Hamaoka [Japan 17 Nov 2001] had no such explanation. INPO and WANO increased pressure on utilities to avoid elevated hydrogen/deuterium concentrations in nuclear process systems.

The BWR hydrogen/oxygen explosions caused a rethink with respect to the environment within CANDU moderators. This re-examination confirmed that deuterium could concentrate in guide tube extensions of vertical reactivity mechanisms. By design, guide tube extensions, have tubing connections that serve as pressure balance lines with the moderator cover gas system. Although deuterium and helium are extremely mobile (have long "mean free paths"), the ability to mix with moderator cover gas through the pressure balance lines is constrained.

Elevated deuterium concentrations have been demonstrated in the guide tube extensions in at least two CANDU plants. The saving grace would appear to be that although oxygen is produced in radiolysis, that oxygen, in my experience, is not observed in cover gas unless there is an oxygen addition system in service.

Of importance, however, is the internal diameter and internal geometry of the guide tube extensions with respect to detonation cell width; therefore these internal properties should be considered with respect to mitigation of deflagration and detonation events.

The on-line gas chromatograph allows for almost continuous monitoring of deuterium, oxygen and nitrogen concentrations in moderator cover gas. Recent experience has shown that gas chromatograph reliability and the availability of alternate sample points are in need of improvement.

The issues of, and following, the Fukushima (Japan 11 March 2011) events will no doubt renew the issue of deuterium deflagration/detonation in CANDU reactors.

2.1 Design Requirements to be considered to reduce deuterium issues

- I would advocate that future design requirements specify that the environment within guide tube extensions must be able to be mixed and/or recirculated with moderator cover gas. This will allow deuterium concentration to be reduced by mixing with oxygen (from oxygen addition) and passing the mixture through the recombiners, forming (heavy) water.

- Examine diameter and internal geometry of the guide tube extensions to determine if changes might be appropriate to further mitigate deflagration and detonation events.
- Refreshment and/or recirculation of guide tube extension gas spaces along with moderator cover gas to minimize deuterium build-up.
- Improve the reliability and redundancy of gas sampling and analysis systems.
- Use of gadolinium sulphate, in place of gadolinium nitrate, for xenon simulation[1] to minimize dissolved deuterium loading.

3. Radwaste

Boric acid is a weak acid and therefore weakly ionized. This leads to inefficient removal of boric acid by ion exchange resin and, in my opinion, the consumption of more ion exchange resin, therefore radwaste, than if gadolinium sulphate were used.

Carbon-14 (C^{14}) is produced in the moderator [$O^{17}(n, \alpha)C^{14}$] and presents a potential emissions problem. The C^{14} will be present in water as bicarbonate which, like borate, is weakly ionized and can be removed by ion exchange. Prevention of build-up of C^{14} is required to minimise emissions to the environment.

I believe that the use of gadolinium sulphate for xenon simulation will simplify ion exchange resin deployment, minimize radwaste and maximise C^{14} control.

3.1 Design Requirement to be considered to minimize radwaste and environmental emissions

- Replace boric acid with gadolinium sulphate for xenon simulation to minimise radwaste and maximize C^{14} control.

4. Annulus Gas Leaks

Gadolinium is used as a soluble poison in CANDU reactors in both moderator liquid poison and LISS. One CANDU unit experienced a leak of carbon dioxide from the annulus gas system into the moderator system. Even though this leakage had continued for some years without incident, also during guaranteed shutdown states, the eventual consequence was the precipitation of gadolinium as the oxalate. Gadolinium oxalate is very insoluble. Conditions arose to allow carbon dioxide radicals to combine forming of oxalate. The consequence was a very long and costly shutdown for the unit.

[1] $Xenon^{135}$ is a fission product having a huge neutron capture cross section (2.665×10^6 barns). When a reactor starts up following a shutdown $xenon^{135}$ is absent, having decayed (half-life 9.39 hours). A soluble poison is added to capture neutrons, in effect simulating the neutron absorbing of the $Xenon^{135}$ until the $Xenon^{135}$ concentration has equilibrated in the fuel.

4.1 Design Requirement to avoid annulus gas leakage into moderator

- New and/or replacement calandria tubes must not allow leakage of carbon dioxide (annulus gas) into the moderator;
- any (suspected) leakage of annulus gas into the moderator is grounds for immediate shutdown of the Unit.

5. Oil

5.1 Fuelling Machine Oil

Pickering is unique in that its fuelling machines have hydraulic systems using oil. This oil has occasionally found its way through heavy water recovery systems into moderator water. At Pickering "A" the consequence was minimal partially because gadolinium (nitrate) has not been used and partially because the cover gas volume, due to the dump tank, is huge. Fuelling machines at other CANDU plants use electric drives and therefore eliminate hydraulic oil ingress from fuelling machines.

5.2 Pump Lubricating Oil

In 2009 at Bruce Unit 3 a volume of moderator pump thrust-bearing oil was inadvertently introduced into the moderator. The consequence was the production of plastic (oil cross-linking) and deuterium/hydrogen (radiolysis of oil). Both the plastic and hydrogen/deuterium produced resulted in significant operational challenges.

5.3 Cover Gas Compressor Oil

In another CANDU unit oil, from one or both, moderator cover gas compressors found its way into the moderator system head tank. The head tank is located outside the significant radiation fields existing in-core. Since oil floats on water there was no immediate consequence.

A drain of the moderator, required for essential maintenance, resulted in the oil being sucked from the head tank into the calandria. The oil was transferred into a high radiation field environment and spread thinly over a large surface area along with water. The consequence was radiolysis of the oil and formation of oxalate and subsequent precipitation of gadolinium oxalate.

5.4 Reactivity Mechanism Oil

5.4.1 Motors

The vertical reactivity mechanisms have motors that, by current design, require lubrication. The lubrication is provided by oil. There are documented cases identifying that oil had leaked from these motor housings. The design should be such that, preferably, oil is not used. If oil must be used then leakage must not be able to enter the moderator.

5.4.2 Dampers

Some vertical reactivity mechanisms have dampers that contain oil. It is not beyond the realms of reality that this oil could leak into the moderator resulting in potential upsets.

5.5 Design requirements to avoid oil ingress into moderator systems

- Use rotating equipment (pumps, compressors, reactivity mechanisms) associated with moderator systems that do not use oil (or grease that can run);

- Avoid oil use of in other components (vertical mechanisms) to prevent oil from entering moderator systems;
- This is, in my opinion, a safety issue; should an upset occur, the increased complications from either elevated deuterium or precipitation of gadolinium must be prevented.

6. Considerations for revisions to design requirements

- Eliminate oil from rotating equipment (pumps, compressors, reactivity mechanisms) associated with moderator systems.
- Avoid use of oil in other moderator (related) components.
- Use of gadolinium sulphate for xenon simulation;
- Use of gadolinium sulphate in place of boron for reactivity shim;
- Use of gadolinium sulphate for LISS;
- Incorporate guide tube extension gas spaces into the recirculated moderator cover gas path.
- Improve reliability and redundancy of cover gas sampling and analysis systems.
- Change guide tube extension internal diameter and geometry.
- Eliminate leakage of annulus gas into moderator.

7. Summary

Should the recommended considerations of design requirements be reviewed, deemed acceptable and adopted they will result in:

- Reduced deuterium generation in the moderator;
- Reduced probability of a deflagration or detonation due to high concentrations of deuterium;
- Reduced probability of reactivity upset events as a result of precipitation of gadolinium oxalate;
- Reduced radwaste as IX resin whilst maximizing control of C^{14} releases.
- Reduced complexity of a unit in the absence of oil ingress.