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Source Term Evaluation for Advanced Small Modular Reactor Concepts

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ABSTRACT – In reactor safety analysis the "source term" is the amount of radioactivity available for release to the environment in the event of an accident. Knowledge of the source term is necessary to predict the radiological consequences of a postulated event. In turn, quantification of the source term requires knowledge on the amount of radionuclides present in the reactor and, most importantly, an understanding of the mechanisms in which they may be released to the environment. Mechanistic models have been developed to predict source terms in design basis accident and severe accident analyses of water-cooled reactors. However, several of the SMR concepts proposed for deployment in Canada are more advanced designs that eschew water coolant or even solid fuel. In this study three advanced SMR concepts were examined: a High Temperature Gas Reactor, a Lead-Cooled Fast Reactor, and a Molten Salt Reactor, to determine the mechanisms for radionuclide release and the potential limiting (worst-case) accident with regards to source term for each.

Introduction

Small Modular Reactor (SMR) and Very Small Modular Reactor (VSMR) concepts have been touted as competitive alternatives to large-scale Nuclear Power Plants (NPPs) by virtue of their smaller, simpler designs, through which they carry smaller financial risk and are more adaptable to growth and off-grid operation. The need for power and steam generation in remote locations (e.g., isolated communities, industrial sites, military bases, etc.) makes VSMRs of particular interest to Canada, and several vendors have been marketing VSMR concepts specifically to the Canadian market.

While many SMR concepts are based on mature Light Water Reactor (LWR) technologies, several of the proposed VSMRs concepts are more advanced in their use of high temperature gas coolant, lead coolant, or liquid fuel in the form of a molten salt. The nature and progression of many Design Basis Accidents (DBAs), Beyond Design Basis Accidents (BDBAs), and Severe Accidents (SAs) in such VSMRs will necessarily differ from contemporary water-cooled reactors. As a consequence, the limiting reactor accident scenarios with respect to source term (i.e., the amount of radioactivity that can be released to the environment) have not necessarily been identified.

In many cases, the technologies in the relevant VSMR concepts have had substantial conceptual development, and in some cases operating experience, in larger-scale NPPs or research reactors. Past analysis on DBAs and BDBA/SAs in these reactors may be extremely relevant to the VSMR concepts. This paper summarizes results from a literature review on accident phenomenology in three reactor types: a High Temperature Gas-cooled Reactor (HTGR), a Lead-cooled Fast Reactor (LFR), and a Molten Salt Reactor (MSR). The existing literature is used as a basis to postulate maximum credible accident scenarios for the corresponding VSMR concepts.

1. High Temperature Gas Reactor (HTGR)

HTGRs have been under consistent development for several decades and several experimental and demonstration reactors have been constructed and operated from the 1960s to present. Among the experimental reactors were Dragon in the United Kingdom [1], AVR in Germany [2], HTR-10 in China [3], and HTTR in Japan [4] (with both HTR-10 and HTTR currently operating). The demonstration reactors include Peach Bottom Unit 1 [5], Fort Saint Vrain [6] (both in the United States) and THTR in Germany [7]. HTGR designs have undergone substantial refinement based on the operating experiences at these facilities. Contemporary HTGR concepts now typically have two common features: an inert helium coolant with outlet temperature of at least 700°C (some as high as 950°C), and a core consisting of tristructural-isotropic (TRISO) fuel particles in a graphite matrix (which serves as the moderator) [8].

These TRISO particles, which are mixed with graphite to form cylindrical compacts or spherical elements, are an integral part of HTGR safety due to their ability to retain fission products. The SiC layer of the particle, which is both the primary load-bearing member and barrier to fission product release, has been demonstrated to remain intact up to 1600°C [9]. Limited amounts of some fission products (e.g., caesium) are released by diffusion through an intact SiC layer at elevated temperatures [9], but these are not usually considered to contribute significantly to the source term. The fundamental tenet of HTGR safety is thus ensuring that 1600°C fuel temperature is not exceeded in any postulated DBA or BDBA.

Potential accidents in HTGRs have been categorized in to three types: (1) core heat-up, (2) depressurization, and (3) air and/or water ingress [10,11]. Core heat-up refers to the rise in temperature resulting from an insufficient ability to remove heat from the core (either fission heat or decay heat). The scope of postulated heat-up transients includes unintentional control rod withdrawals (i.e., reactivity insertions) and loss of forced flow, which may or may not be followed by reactor shutdown (scram). Higher peak temperatures are obviously achievable in the anticipated transients without scram, although negative reactivity feedback from increasing core temperature rapidly reduces the power to a low level. Should the reactor remain in that state for a prolonged period, the decay of short lived fission products (e.g., xenon) and the cooling of the core will result in recriticality. Eventually, owing to the temperature reactivity feedback, the core will assume a steady power equivalent to its capability to reject heat through all available means. Analysis typically shows that the peak fuel temperatures will be achieved following the recriticality, not in the initial part of the transient [12,13]. Smaller HTGR cores (<400 MWth, including SMRs) experience much lower peak temperatures than larger cores owing primarily to the larger surface area to volume ratio, which allows them to more effectively reject heat through passive means (including radiation) to the reactor cavity [14,15]. In such small cores, peak temperatures below the 1600°C limit are reasonably achievable in any postulated core heat-up accident. Tests of loss of forced flow without reactor scram have been performed at AVR [16], HTR-10 [3], and HTTR [17].

Depressurization of the primary helium cooling circuit, such as through a pipe break, has been reported as the most important licensing based event (the severity of which will ultimately depend on the amount of air and/or water ingress) [8]. Conservatively, it can be assumed that all the activity present in the primary circuit will escape in to the environment. This includes any activation of the coolant gas as well as any releases from the fuel particles. Assuming the vast majority of fuel particles are intact (i.e., the peak core temperature is below 1600°C), the source of activity will primarily be heavy metal contaminants in the fuel particles left from the manufacturing process. Uranium contaminants outside the SiC layer, for example, will undergo fission and freely release the products in to the binding graphite and the coolant gas. The activity that existed inside the primary circuit prior to the depressurization, including that from contaminants and defective particles, is expected to dominate the total accident source term in design basis events [8,15].

Ingress of air in to the primary circuit is expected following depressurization. The most likely path for water ingress is leaks or breaks in a secondary steam circuit, as was experienced at AVR [2] and Fort Saint Vrain [18], however the potential for water leakage is limited if a specific HTGR concept uses a gas-turbine and not a steam generator. The concern with air ingress comes from the oxidation of the graphite structure (and to a lesser extent, the fuel particles themselves). It is not agreed whether or not the exothermic oxidation of graphite represents "burning" in the conventional sense [19,20], but there is generally agreement that the consequences of unlimited graphite oxidation would be detrimental towards HTGR safety. These consequences include the heat of oxidation contributing to fuel temperature increase, corrosion of the graphite support structures within the core, mobilization of contaminants retained in the graphite matrix, and the formation of burnable gases (e.g., carbon monoxide) [19]. Significant air ingress with large amounts of graphite oxidation is considered far beyond a DBA [15].

The oxidation rate of graphite above 650°C (i.e., at the normal operating condition, or in an accident without scram) is essentially limited only by access to free oxygen [20]. The flow path within the primary vessel can be designed so that there is no "chimneying" that rapidly brings air in to the core in the event of a pipe break (e.g., by locating both the flow inlet and outlet at the bottom). It is therefore necessary to postulate separate breaks/leaks at both the bottom (e.g., guillotine rupture of primary circuit piping) and top of the vessel (e.g., through control device penetrations) for there to be sufficient flow of air to result in substantial oxidation. If sufficient flow of oxygen is achieved, then the production of combustible carbon monoxide above its autoignition temperature must be considered. These additional heat sources may contribute to the fuel temperature exceeding the 1600°C limit. Past this temperature, it can be postulated that failure of the fuel particles would freely release fission products to the environment, aided by combustion or detonation of carbon monoxide which could have detrimental effects on the building structure.

2. Lead-Cooled Fast Reactor (LFR)

LFRs were identified by the Generation IV International Forum (GIF) as one of the six technologies that could meet their goals for the next generation of reactors. According to GIF, LFRs have great potential to meet small-unit electricity needs of remote sites while still possessing the ability to be scaled up for larger capacity generation [21]. LFRs below 100 MWe are considered especially attractive since the small size facilitates good breeding ratios, very small reactivity swings with burnup, and passive decay heat removal [22].

Past experience with LFRs is essentially limited to a Russian (Soviet) naval reactor program, which used lead-bismuth eutectic as the coolant rather than pure lead owing to the eutectic's lower melting temperature (124°C vs. 327°C for pure lead). The program consisted of two land-based facilities, one prototype submarine, and seven *Alfa* class submarines, totalling 15 separate reactors accumulating 80 reactor-years' worth of experience [23]. Lessons learned from this experience have been incorporated in to contemporary LFR concepts, primarily in the maintenance of the coolant chemistry to mitigate the corrosive nature of liquid lead and the production of lead-oxide slag, as well as use of an integral (or "pool-type") reactor design that eliminates primary circuit piping and its risk of failure [24].

The extremely corrosive nature of molten lead towards structural materials is mitigated by maintaining a concentration of dissolved oxygen in the primary coolant. This results in creation and maintenance of a protective oxide layer on the material surface [25]. A second aspect of lead coolant chemistry control is the removal of any lead-oxides (e.g., by reduction with hydrogen) [23]. Significant quantities of lead-oxide slag, primarily formed by interaction between molten lead and humid air, caused substantial degradation of fuel-to-coolant heat transfer in one lead-bismuth-cooled naval reactor. Subsequent withdrawal of the control rods (to increase reactor power) resulted in substantial fuel failure, with fuel and fission products being carried away with the lead in to the primary circuit [24]. While there is no energetic chemical reaction between lead and water or lead and oxygen (as occurs with sodium, another fast reactor coolant), maintenance of an inert cover gas and/or reduction with hydrogen is necessary to limit accumulation of this slag during operation [26].

LFRs, as with all liquid metal cooled fast reactors, generally have smaller reactivity feedback coefficients than thermal spectrum reactors. As a consequence, the reactivity worth of control devices is limited to be less than the delayed neutron fraction β so that no unintended withdrawal may make the core prompt-critical [27]. Void reactivity in high-leakage lead-cooled cores is typically, but not always, negative. Void in low-leakage cores, or localized void in the centre of a small core, can have a positive reactivity worth [28]. Given the high boiling point of pure lead at atmospheric pressure (1749°C), void from coolant boiling is extremely unlikely. One potential source of coolant void, however, is entrained steam from a failed steam generator tube [29]. The reactivity worth of entrained steam bubbles is of sufficient concern that in some LFR concepts the coolant flow path is specifically designed to eject steam bubbles to the cover gas system and prevent their ingress to the core. Such a flow path, however, limits the capability for natural coolant circulation in accident conditions, so the relative merits of each approach need to be weighed in specific designs [30].

Apart from noble gasses, lead is considered to have a high retention capability for fission products. Notably, iodine and caesium have been observed to form compounds with lead up to 600°C [31]. If a prompt-critical reactivity excursion is postulated (e.g., as a result of entrained steam bubbles entering the core, unintentional control rod withdrawal, or some combination thereof depending on the specific LFR design being considered) the result would be substantial cladding failure and release of fuel and fission products in to the lead coolant. Noble gasses would bubble out in to the inert cover gas. Fuel materials and other fission products are expected to be retained in the lead (either in solution or as lead compounds). Since the lead itself resides in a single vessel at low pressure, there is no obvious path for fission products retained in lead to be released. Volatilization of some fission products (e.g., iodine, caesium) at elevated temperatures may be possible, in which case they would also be released in to the inert cover gas. It is then necessary to postulate failures and/or leaks in the cover gas system to provide a path for these volatile fission products to escape.

Assuming all of the above, the total accident source term will thus be dictated by the total volatilized fraction of fission products.

3. Molten Salt Reactor

The MSR concept originates from work performed at Oak Ridge National Laboratory (ORNL) in the 1960s that culminated in the Molten Salt Reactor Experiment (MSRE) [32]. The unique feature of the MSR is that the fuel is in solution with a liquid fluoride salt that also functions as the primary coolant. In the primary circuit, this fuel salt typically flows through a graphite moderator (although fast-spectrum concepts now exist) and then through a heat exchanger, delivering heat to a secondary "clean" (i.e., without fuel) salt. MSRs are generally conceived of as breeder reactors, as was the original intent of the ORNL developers, but there is no reason that a MSR could not operate as a strict "burner" of fuel [33]. The attractive features of the MSR are generally considered to include [34]:

- The fuel salt / primary circuit operates at low pressure (atmospheric) but high temperature (700°C) with a large margin to boiling (1400°C);
- The molten salt undergoes no violent chemical reaction with air or water;
- The fuel salt retains most fission products in solution, except noble gasses which bubble out;
- Large, negative temperature and void reactivity coefficients.

The principal concepts in the MSR were conceived during the Aircraft Reactor Experiment (ARE) [35] and finalized during the construction of the MSRE [36]. The MSRE was designed to operate with all of the materials to be used in a potential breeder, except for simplicity it used a single fluid with no chemical processing, did not include thorium, and ran at lower power (8 MWth). It operated between 1965 and 1969, and is notable for being the first reactor to be critical on ^{233}U [36]. Contemporary MSR concepts are based heavily on the MSRE. This is especially true for "burner" MSRs with powers in the SMR or VSMR range.

Since the natural state of MSR fuel is a flowing liquid, the key aspect of MSR safety becomes retention of fission products inside the molten salt. The MSRE remains the principal source of knowledge of fission product behavior in molten fluoride salts [37]. There, it was observed that: (1) noble gasses (e.g., xenon, krypton) will bubble out freely in to an off-gassing system, (2) many fission products (including caesium) form stable fluorides and are permanently retained in the salt, and (3) certain noble metals and tellurium will not form fluorides nor remain in solution, but rather plate out on the surfaces of the primary circuit [37]. There was a notable gap between how much iodine was predicted to be produced and how much could be measured [37]. It was theorized that the missing inventory must be outside the primary circuit, since laboratory experiments showed that if iodine formed in contact with the fluoride salt, it would be well retained [38]. One source of iodine in the off-gassing system was theorized to be decay of a volatilized tellurium precursor [38].

Substantive summaries of different postulated accidents in MSRs have been presented in the original MSRE safety report [39] and contemporary literature [40,41,42]. While the effective delayed neutron fraction β in MSRs is very low, the extremely large fuel temperature feedback limits the potential consequence of any reactivity insertion accident [39,40]. Furthermore, contemporary MSR concepts include passive mechanisms for rejecting decay heat during accidents or prolonged station blackouts. There is thus no obvious mechanism by which the retention of fission products may be compromised by overheating. Integral designs that forego piping also substantially reduce the likelihood that molten salt may escape containment within the primary circuit (e.g., as a result of a pipe break). It is generally recognized that the most likely path for radiation release is a failure of the off-gassing system that leaks noble gasses [40].

In order for there to be any substantial release of activity from an MSR, it is first necessary to postulate a loss of primary circuit integrity. Failure of the vessel in an integral design is unlikely, and even so, there is no pressure that would drive molten salt out of any leak (whether or not there is a cavity to leak to would depend on the specific MSR design). More credible would be confinement failure through any penetrations at the top of the vessel (e.g., to the off-gassing system). Loss of primary circuit integrity would provide a path for water to come in to contact with the molten salt (assuming that there is a large leak of water above the core). While there is no violent chemical reaction, the interaction with water could be expected to: (1) remove some fission products from solution in the salt, (2) through the production of steam, provide a driving pressure for release of volatilized activity, and (3) produce corrosive vapours and solutions (e.g. fluorides and chlorides) which could eventually imperil system integrity. The credibility of such an event is evident in the MSRE safety report [39], where the maximum credible accident in the MSRE safety analysis was a spill of the entire molten salt inventory simultaneous with a large spill of water.

4. Conclusions

On the basis of a literature review on accident phenomenology for three reactor types (HTGR, LFR, and MSR), potential limiting accident scenarios for SMR and VSMR concepts based on these technologies have been identified. “Limiting accident” in this case refers to the event or sequences of events that would result in the maximum conceivable amount of fission products being released in to the environment.

- For the HTGR, in order to achieve the temperatures necessary for failure of the TRISO fuel particles, it is necessary to postulate a depressurization and air ingress accident without reactor shutdown, accompanied by substantial graphite oxidation and production of combustible gasses.
- For the LFR, it is necessary to postulate substantial fuel failure as a result of a prompt-critical excursion, followed by leaks in the cover gas circuit that allow escape of volatilized fission products.
- In the MSR, a substantial leak of water that comes in to contact with the molten fuel salt may result in release of volatilized fission products.

Many of the key phenomena in these events, including combustion of carbon monoxide produced by reactor graphite oxidation, volatilization of fission products from molten lead at elevated temperatures, and the interaction of fission products in molten salt and water, have not been well quantified in literature.

It should be noted that the sequence of events necessary for there to be substantial release of fission products in these reactors are nearly incredible. Further, it is possible to account for these specific scenarios in the conceptual design of an SMR, so that the likelihood of such release occurring may be so minimal as to be completely inconsequential. More detailed analysis on specific reactor designs will need to be performed in order to establish if the above events are credible. What this does not take in to account, however, is the effect of any external malicious action (which is typical of this type of analysis). It is possible that, given the inherent robustness of these designs towards limiting the release of radioactivity, it will be necessary to postulate such initiating events for the limiting activity releases.

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