INTERNATIONAL NUCLEAR SAFETY JOURNAL
vol. 3 issue 1, 2014, pp. 13-37

ISSN 2285 – 8717
ISSN–L 2248 – 8717
http://www.insj.info

HEALTH PHYSICS ASPECTS OF GENERATION IV REACTORS

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Received: *12 September 2013*; accepted: *29 November 2013*

Abstract
The health physics challenges encountered by power operations in existing Generation II and III reactors are also present in the planned Generation IV systems. Generation IV reactors will present additional health physics challenges because the core coolants include lead, sodium, and molten salt in addition to light water. These materials create new activation products and these coolant media present unique operational challenges associated with refueling operations, primary coolant component maintenance, and reactor cleanup systems. The extent of these challenges depends on the performance of Generation IV fission product barriers. This is particularly important for molten salt reactors that contain fuel in a eutectic mixture with the coolant.

Keywords
Generation IV reactors; health physics; fission product barriers; operational issues; radiological protection

1.0 Introduction

Advanced fuel cycles will incorporate Generation IV fission power reactor designs [1-14]. The various stages of power reactor evolution are specified in terms of Generations. Generation I reactors were experimental devices used to investigate and verify design concepts. Most operating systems are Generation II designs that utilize pressurized water reactors (PWRs) or boiling water reactors (BWRs). These reactors utilize active safety systems. Generation III reactors are similar to Generation II designs but incorporate passive safety systems. A number of Generation III systems are operating and more are under construction [5,6,8,15].

An advanced fuel cycle must necessarily consider the long-term resource potential, energy supply and demand projections and, the potential deployment of advanced reactors including the planned Generation IV systems. Generation IV technologies are targeted for deployment by 2030, and will include a 60-year design lifetime. The Generation IV goals of sustainability, safety and reliability, and economic viability require consideration of the entire fuel cycle [5,8,16,17]. The health physics aspects of this advanced fuel cycle are considered in this paper.

Emerging technologies and changes in deployment strategy affect the operation and inherent structure of the 21st Century nuclear fuel cycle. Emerging technologies include the laser enrichment of uranium and use of Generation IV reactors to minimize the quantity of high-level waste [5,6,8]. Changes in strategy including a renewed interest in fuel reprocessing are also likely.

This paper reviews these and other issues and challenges that face advanced reactors in the 21st Century. The health physics challenges will likely be severe as new regulatory requirements emerge and economic considerations place a premium on operational efficiency with limited resources [10].

2.0 Generation IV Reactors

Operating Generation II and III reactors are dominated by light water systems. Light water reactors dominate the reactor fleet because they were developed first in the 1940-50s as a compact power source for naval vessels and these designs were scaled to commercial sizes. Problems with Generation II light water reactor designs were illustrated during the Three Mile Island and Fukushima Daiichi accidents [6,8,18,19]. In addition, the light water design does not fulfill the original vision of nuclear power generation that incorporated reprocessing spent fuel to extract the maximum energy from this technology.

Reprocessing has been limited by cost and proliferation concerns. The Generation III designs improve Generation II safety performance, but do not minimize high-level waste or effectively integrate the fuel cycle to operate in an optimum environmental manner [20-30].

Generation IV reactors strive to achieve these goals. The Generation IV reactors are conceptual and in the development phase. Currently, Generation IV is used to categorize these evolving designs that incorporate promising and innovative concepts that require significant research to achieve a final design that is capable of being licensed by a regulatory body.

As used in this paper, nuclear proliferation is the spread of nuclear weapons and associated information and technology [9,14,26,31,32]. Technologies associated with nuclear weapons and developing or acquiring their key components such as weapons grade uranium and plutonium are aspects of nuclear proliferation. Nuclear reactors also produce plutonium and use uranium that can be diverted to weapons use.

Reactors are classified into two broad categories: thermal and fast. A thermal reactor is a reactor that primarily operates using the thermal neutron fission of fissile nuclides (e.g., ^{233}U, ^{235}U, and ^{239}Pu). Fast reactors utilize fast neutrons to fission fertile nuclides (e.g., ^{232}Th and ^{238}U). Reactors utilizing fast neutrons also produce fissile materials including ^{233}U and ^{239}Pu. The presence of the nuclides ^{233}U, ^{235}U, and ^{239}Pu are important considerations when evaluating the proliferation potential of fuel cycle technologies. Nuclear proliferation is addressed in more detail in subsequent discussion.

Generation IV reactors have the potential for being proliferation resistant and incorporate reprocessing as part of their integrated design concept. To advance this reactor cycle concept, the Generation IV International Forum (GIF) [2,7,12] was established in 2000 and included the United States, Argentina, Brazil, Canada, France, Japan, South Korea, South Africa, Switzerland, and the United Kingdom. Additional GIF members now include China, Russia, and the European Union. These nations committed to the joint development of the next generation of nuclear technology. The ten nations agreed on six Generation IV nuclear reactor technologies for deployment in the 2030 timeframe. Some of these reactors operate at higher temperatures than the Generation II and III reactors, four are designated for hydrogen production, the characteristics of these reactors are summarized in Table 1, and the primary activation products are listed in Table 2. These activation products are either unique to the Generation IV designs or have production mechanisms that differ from their creation in previous Generation II or III designs.

The six design concepts offer the potential for improved economics; safety, reliability and proliferation-resistance. These designs also maximize the utilization of fissile resources and minimize high-level waste. Generation IV reactors addressed by the GIF include: Gas-cooled fast reactors (GFRs), lead cooled fast reactors (LFRs), molten salt epithermal reactors (MSRs), sodium-cooled fast reactors (SFRs), supercritical water-cooled reactors (SCWRs), and very high temperature, helium-cooled, graphite moderated thermal reactors (VHTRs). Lead-bismuth cooled fast reactors (LBFRs) were previously considered as a Generation IV candidate [1,2]. An advantage of the Generation IV design is the capability for full actinide recycling using a closed fuel cycle concept. Open fuel cycles do not incorporate actinide recycling and are addressed in subsequent discussion.

The principal goals of the Generation IV systems are to achieve high levels of safety and reliability, sustainability, proliferation resistance and physical protection, and economic competitiveness [7,12]. GIF member states collaborating in the development of the six reactor concepts are listed in Table 3. Collaborating states are required to sign a formal agreement that governs intellectual property rights and associated reactor technology. These arrangements have been signed for the SFR, VHTR, GFR, and SCWR reactors. Limited studies are governed by a memorandum of understanding for the LFR and MSR systems.

The GIF plans to construct an SFR and VHTR in the near future following the development of safety design criteria. These criteria must be met while meeting four main challenges for the Generation IV systems. These challenges are the acceptability, sustainability, economic competitiveness, and proliferation resistance of the Generation IV reactor design. An acceptable design must not only achieve the highest safety standards, but the enhanced safety must be clearly communicated to the public. Sustainability includes the long-term viability of these systems as related to fuel design, waste generation, and potential design enhancements [7,12].

The development of the six Generation IV systems is divided into four phases that include viability, performance, demonstration, and commercialization. Design viability and performance require substantial research and development that will involve collaboration of GIF signatories. The demonstration and commercialization phases require significant resources and will require funding commitments. The GIF has categorized the six Generation IV designs in terms of these phases.

Over the next decade, four systems (GFR, MSR, SCWR, and VHTR) will be assigned to the viability phase. The LFR system will move from the viability phase to the performance phase, and the SFR design will transition from the performance to the demonstration category [7,12].

The basic characteristics of these Generation IV systems are summarized in subsequent discussion. Before reviewing the Generation IV reactor types, an examination of expected activation products is provided. These activation products are summarized in Table 2 and focus on the unique Generation IV specific isotopes associated with fuel, coolant, and primary system components. The production mode for the activation product is also provided. Subsequent discussion outlines the unique Generation IV materials that lead to radionuclides not normally associated with Generation II and III reactors. The Generation IV reactors also produce the activation products encountered in Generation II reactors. These common fission and activation products include ^{3}H, ^{16}N, ^{54}Mn, ^{59}Fe, ^{58}Co, ^{60}Co, ^{85}Kr, ^{90}Sr, ^{90}Y, ^{95}Zr, ^{131}I, ^{133}Xe, ^{135}Xe, and ^{137}Cs.

Generation IV graphite-moderated reactors produce other isotopes due to impurities in this moderator. These isotopes include ^{38}Cl [$^{37}Cl(n, \gamma)$], ^{46}Sc [$^{45}Sc(n, \gamma)$], ^{82}Br [$^{81}Br(n, \gamma)$], and ^{152}Eu [$^{151}Eu(n, \gamma)$]. The specific isotopes and activity levels produced in graphite-moderated reactors will depend on the acceptable impurity content utilized in the graphite designs.

2.1 Gas-Cooled Fast Reactors (GFR)

The reference Generation IV GFR [7,12] utilizes a fast neutron spectrum and a helium cooled reactor core. It uses a closed cycle process that incorporates a direct cycle helium turbine for electricity production, and uses process heat for the thermochemical production of hydrogen. With a fast neutron spectrum and full actinide recycle, the GFR minimizes the production of long-lived radionuclides.

Operating in conjunction with a closed fuel cycle, GFRs enhance the utilization of uranium and minimize waste generation. Actinides waste is limited by incorporating full actinide recycle as part of the closed fuel cycle. To improve efficiency, the GFR facility is co-located with other fuel cycle facilities for onsite spent fuel reprocessing and fuel re-fabrication. The closed fuel cycle more closely links the power production, reprocessing, and waste disposal options in order to limit the generation of high-level waste.

The GFR concept has a number of characteristics that support health physics design objectives. These include the fuel composition that enhances fission product retention and actinide recycle that permits operation of a closed fuel cycle. GFR fuel incorporates a number of enhancements including advanced coatings and ceramic fuel composites (e.g., SiC, ZrC, TiC, NbC, ZrN, TiN, MgO, and $Zr(Y)O_2$) to facilitate fission product retention. These fuel characteristics enhance the fuel fission product barrier and limit the probability of a release of radioactive material to the environment.

Full actinide recycle eliminates long-term waste disposal and associated radiation dose concerns. In addition, the closed nature of the fuel cycle limits the occupational doses associated with waste disposal and storage. The extent of these health physics advantages depends on GFR fuel performance and the development of actinide recycling technology. Accordingly, near-term GFR activities focus on the development of severe accident mitigation, demonstration of performance, and design of a small experimental reactor.

2.2 Lead Cooled Fast Reactors (LFR)

The LFR system [7,12] utilizes a fast neutron spectrum, in-vessel steam generators, and a core that is cooled passively through natural convection with a liquid lead coolant. This reactor type is an inherently safe system and has the potential for significant waste volume reduction relative to advanced light water reactors. A key advantage of the liquid metal reactors is the potential to recycle essentially all of the actinides. LFR applications include the generation of electricity, hydrogen production, and desalination of sea water.

The LFR design must demonstrate successful proliferation resistance and economic viability. Economics are improved through simplification including modularization of the design. Proliferation issues are minimized if the design is

successful in efficiently recycling actinides, particularly ^{239}Pu.

Core lifetimes are projected to approach 15 – 20 y. The LFR system offers considerable flexibility and facility options include a 20 – 180 MWe fabricated reactor module, a 300 – 1,200 MWe modular design, and a 600 - 1,200 MWe base load facility.

The LFR concept utilizes a closed fuel cycle with the supporting facilities residing in a central or regional location. Within the closed fuel cycle, LFR facilities provide efficient utilization of uranium resources and management of actinides.

A number of issues must be resolved for the LFR concept to become a commercial viability. Chemistry criteria are needed to facilitate the control of oxygen and ^{210}Pb. The development of fuel and reactor materials and achieving acceptable corrosion properties for these materials are additional issues.

The liquid metal design initially incorporated a lead bismuth cooled fast reactor. However, there were a number of design issues that suggested a lead coolant would provide better performance. One health physics issue associated with the LBFR was the capability of the lead-bismuth eutectic to retain fuel and fission products during all operating conditions.

The limitations regarding fuel and fission product retention in the lead-bismuth coolant can be mitigated using lead. According to the Generation IV International Forum [7,12], the lead-cooled fast reactor system has excellent materials capabilities and fission product retention. In addition, the LFR's molten lead coolant is relatively inert which should lead to improved safety performance and reliability.

From a health physics perspective, a lead coolant has several positive aspects that can minimize the potential for a fission product release. In particular, lead has a high boiling point, low vapor pressure, and provides an efficient gamma-ray shield. Lead is compatible with the fuel material and retains fission products. Although these characteristics enhance the fuel fission product barrier, they must be demonstrated under accident conditions including design basis and beyond design basis events.

The lead coolant also enhances reactor safety by contributing to a low core damage probability. This result is supported by lead's heat transfer characteristics, high specific heat and thermal expansion coefficient, and inherent negative reactivity contribution to the LFR core. These characteristics also support good heat transfer from the core to lead coolant and the capability for natural circulation of the reactor coolant during emergency conditions. From a release perspective, lead reduces the risk of a recriticality following a core melt event.

The use of a liquid lead coolant creates a number of operational difficulties that will be encountered during routine outage activities. For example, the high temperature lead coolant presents a challenge during refueling operations. The coolant must remain in a liquid state for refueling to occur. There are engineering solutions that resolve the refueling issue including the use of a cool cover gas to facilitate access to the fuel assemblies. In addition, a number of operational requirements and maintenance activities involving primary system components in a liquid lead environment must be addressed for the LFR design to become viable.

The LFR is primarily envisioned for electricity and hydrogen production, and actinide management. Since the LFR system is transitioning into the performance phase, research and development will focus on reactor safety and ensuring that the fuel, reactor materials, and associated corrosion control measures perform as anticipated.

Two LFR reactor concepts are currently being designed [7,12]. These are the 20 MWe Small Secure Transportable Autonomous Reactor (SSTAR) developed in the USA and the 600 MWe European Lead-cooled System (ELSY), developed by the European Union. Most of the engineering and materials challenges are projected to be addressed by the ELSY design configuration. Separate designs for a small, transportable LFR with a long core life and a moderate-sized power plant will incorporate the operating experience derived from the SSTAR and ELSY demonstration facilities. Larger facilities are dependent on the success of these designs.

2.3 Molten Salt Epithermal Reactors (MSR)

Molten salt reactors [7,12] have potential advantages in terms of proliferation resistance attributable to the lower fuel inventory and plutonium buildup, and a reduced source term associated with the online separation and removal of fission products. The circulating molten salt fuel is a mixture of zirconium, sodium, and uranium fluorides. Other molten salt options include lithium and beryllium fluoride with dissolved thorium and ^{233}U. The molten salt/fuel flows in channels through the core's graphite

moderator. The MSR reference power level is 1,000 MWe.

Since the fuel is in a liquid state, fuel processing is performed while the reactor is operating. The produced actinides and fission products form fluorides in the liquid coolant. This chemistry permits the fuel cycle to be tailored for the destruction (burnup) of minor actinides and plutonium and the removal of fission products. Since the MSR fuel cycle allows full actinide recycle, waste issues are dominated by fission products. However, the MSR concept requires refinement and developing of high-temperature structural materials, establishing and demonstrating appropriate fuel characteristics, and resolving nuclear and hydrogen safety issues.

Since the fuel is dissolved in the coolant, the MSR design only has two fission product barriers. This is a significant departure from the current safety philosophy based on three fission product barriers. Any primary coolant leakage leads to the release of fission products to the facility. If leakage occurs in the containment building, it is the only remaining fission product barrier. Auxiliary building (AB) leakage merits special attention because no containment exists in Generation II and III AB structures. Therefore, health physics activities at a MSR will be strongly influenced by the ability of the coolant to retain fission products, activation products, and actinides in the molten salt coolant.

Although there are possible radiation safety issues associated with the MSR's liquid fuel, there are also positive nuclear and radiological safety characteristics. The liquid fuel has an advantage that it is recirculated and fission products can be continuously removed to minimize the source term. Recirculating fuel also facilitates the destruction of minor actinides.

The MSR design incorporates a unique reactor vessel design that incorporates a hole in its lower head. This hole is plugged with solidified fuel material. The fuel material remains in a solid state and is cooled by a refrigeration unit. When the plug is solidified it preserves the fission product barrier's integrity. If the reactor loses power during an emergency, the refrigeration unit becomes inoperable. Without power to maintain the solidified material, the plug melts and the fuel drains into underground holding tanks. These tanks provide a stable, safe shutdown condition to prevent the release of fission products.

Given the level of development required for the MSR design to become fully mature, additional health physics issues may emerge. A potential area of concern is the capability of the liquid fuel/coolant to retain fission and activation products during a severe accident. The capability of safety systems to preserve fission product barriers during design and beyond design basis accidents must be demonstrated. In addition, a number of technical issues must be addressed including demonstrating a safety approach, completing a fuel reprocessing flow sheet, characterizing the properties and behavior of the liquid salt coolant, and developing robust reactor materials.

2.4 Sodium-Cooled Fast Reactors (SFR)

SFRs operate with a fast neutron spectrum and utilize a liquid sodium coolant [7,12,33]. The SFR design is associated with a fuel cycle that incorporates full actinide recycle. With design improvements, the SFR can also be used for electricity production. Three SFR reactor concept designs are currently envisioned.

The first design is a large scale 600 to 1,500 MWe loop-type sodium-cooled reactor using mixed uranium-plutonium oxide fuel. Its fuel cycle is based upon advanced aqueous fuel reprocessing technology. The second design is an intermediate sized 300 to 600 MWe pool-type reactor and the third is a small-scale 50 to 150 MWe modular-type sodium-cooled reactor utilizing uranium-plutonium-minor-actinide-zirconium metal alloy fuel. Design options 2 and 3 are supported by a fuel cycle based on fuel reprocessing in facilities that are integrated with the reactor.

The SFR system benefits from considerable Generation II reactor operating experience with sodium cooled reactors. If the SFR capability to efficiently consume plutonium and other minor actinides were achieved, it would significantly reduce the actinide loadings in high-level radioactive waste. These actinide reductions would reduce the SFR's radioactive waste disposal requirements and enhance its non-proliferation characteristics. Reducing capital cost and improving passive safety system performance under transient conditions are the major challenges for implementing the SFR design concept.

Given existing experience with Generation II sodium cooled reactors, the health physics concerns are better defined than in other Generation IV systems. These health physics issues are similar to

the Generation II issues [5,6,8], but are complicated by the potential for the sodium-water chemical reaction to mobilize fission and activation products [33]. Health physics issues could also arise from the implementation of the closed fuel cycle with full actinide recycle. Experience with 20th Century reprocessing suggests waste storage, environmental concerns, maintenance of heavily contaminated equipment, and decommissioning issues merit thorough evaluation.

The SFR is advancing to the demonstration phase. Planned reactors supporting the SFR concept include the Russian BN-800 scheduled for operation in 2014, the French ASTRID with an operation date near 2023, and Japanese and Korean designs that are in development [7,12]. Research and development are focusing on enhanced safety options.

2.5 Supercritical Water Cooled Reactors (SCWR)

The SCWR system [7,12] is similar to the BWR design. It is primarily designed for efficient electricity production, with an option for actinide management. The SCWR is based on either a thermal spectrum or a fast spectrum. The thermal neutron version uses once through uranium dioxide fuel and has the same waste management issues associated with a Generation II and III single pass fuel cycle. From a health physics perspective, a fuel cycle without actinide recycle is not a desirable Generation IV alternative.

SCWRs have a thermal efficiency about one-third higher than current Generation II and III light-water reactors [4,7,12]. The plant design is considerably simplified because the coolant does not change phase in the reactor and is directly coupled to the energy conversion equipment. As with other light water systems, the fuel is uranium oxide. Passive safety features are similar to those utilized in Generation III simplified boiling water reactors [5,6,8].

The fast spectrum version permits actinide recycle using conventional reprocessing technology. However, the fast reactor version must overcome materials development issues. Both SCWR options utilize passive safety systems, and operational characteristics similar to those of the simplified BWR [3,5,11,15]. The full actinide recycle version is based on advanced aqueous fuel reprocessing. The fuel reprocessing facility may support individual or multiple SCWR reactors and must be integrated with these reactors for this Generation IV concept to achieve economic viability.

Based on initial design efforts, the SCWR concept has a number of reactor safety issues [7,12]. First the design has a tendency to have a positive void reactivity coefficient that limits the capability to reach a stable configuration during a severe reactor transient. There is also the potential for design basis loss-of-coolant accidents to occur. These two characteristics complicate the development of the SCWR concept. Other challenges for the SCWR are the development of a viable core design, accurately estimating the heat transfer characteristics, and developing fuel and core structural materials that are corrosion-resistant during the various SCWR normal and transient operating conditions.

The SWCR facility should have health physics issues that are similar to those encountered in Generation II and III BWRs [5,6,8]. Additional health physics issues will arise if reactor materials fail to achieve the desired lifetime and reliability goals.

2.6 Very High Temperature Reactors (VHTR)

The Very High Temperature Reactor is a high-efficiency, graphite-moderated, helium-cooled reactor that operates with a thermal neutron spectrum [7,12]. It can be utilized for the cogeneration of electricity and hydrogen and to provide process heat for industrial applications. The basic technology for VHTR systems has been established in Generation II high temperature gas cooled reactors [5,6,8].

VHTR fuel consists of coated particles using materials such as SiC and ZrC that are formed into pebble elements or prismatic blocks. The plant uses once through uranium fuel or U/Pu fuel to produce electricity, hydrogen, or process heat. Waste disposal issues associated with long-term spent fuel storage are not resolved by the VHTR's open fuel cycle.

Since the basic technology for VHTR systems has already been established in Generation II high temperature gas cooled reactors [5,6,8], the Generation IV design is an evolutionary development. However, the system's aim of operating at 1000°C presents challenges in terms of fuel and materials development and in maintaining reactor safety under transient conditions.

Technology advancements in fuel performance and high-temperature materials development are required for the VHTR to be a viable technology. Shortcomings in either of these areas would

potentially weaken the fuel and the primary coolant system fission product barriers. If these issues are resolved, the health physics issues will resemble those at a Generation II HTGR facility [5,6,8].

Current GIF estimates [7,12] suggest a prototype startup around 2015. The design is currently focusing on achieving the desired high outlet temperatures. Developing advanced materials and fuel designs will govern the long-term viability of the VHTR.

2.7 Radionuclide Impacts

The extent to which the radionuclides of Table 2 will dominate effective doses at a Generation IV facility ultimately depends on the reactor's operational characteristics. Based on Generation II and III experience, a number of health physics considerations will apply to Generation IV systems.

Internal radiation hazards are presented by ^{3}H in the HTO form and ^{14}C as CO_2 particularly during refueling operations and primary system maintenance. The extent of the hazard depends on allowable leakage and primary system performance characteristics. Traditional Generation II and III activation products and fission products including ^{60}Co and ^{131}I also present an internal radiation hazard.

Submersion hazards exist for short-lived radioactive gases (e.g., ^{15}O, ^{16}N, ^{17}N, ^{19}O, and ^{23}Ne). The noble gases produced in the fission process also present a submersion hazard, and these are primarily comprised of isotopes of Kr and Xe.

External hazards exist for a variety of nuclides including the coolant activation products ^{16}N and ^{24}Na. The extent of the external radiation hazard is dependent on the magnitude of the production of fission and activation products that decay via beta and gamma emission. It is likely that ^{58}Co and ^{60}Co will significantly contribute to worker effective doses, which is consistent with Generation II and II operating experience [5,6,8].

Reactor coolant leakage in the MSR, LFR, and SFR designs introduce new hazards that were not routinely encountered in Generation II and III reactors. The leakage of MSR coolant containing entrained fuel and fission products presents a significant source term that is much greater than encountered in Generation II and III light water reactor coolants. These coolant activity levels may be comparable to the levels encountered during the TMI-2 and Fukushima Daiichi accidents [18,19]. High activity levels will require changes in operating practices compared to contemporary PWR auxiliary building or BWR reactor building maintenance activities in Generation II and III reactors. The retention of fission gases and dispersion of entrained fission and activation products could require that routine maintenance and spill cleanup be accomplished remotely or using robotic techniques.

The LFR and SFR coolant activity levels are governed by the fuel integrity. However, SFR leakage presents a challenge because the energetic sodium-water reaction has significant potential to disperse radioactive material. LFR leakage must address the toxic characteristics of lead. The consequences of liquid lead and liquid sodium leakage in an industrial environment require controls to mitigate their effects. Radiation work permits and personal protective equipment must address these leakage issues in an operating Generation IV reactor.

Offsite releases of radioactive material from a Generation IV reactor are expected to be similar to those from Generation II and III facilities. The offsite release source term is dominated by radioiodine and noble gas activity. MSRs present potential health physics issues because there are only two fission product barriers. The MSR source term will require further investigation and characterization in terms of the capability of the coolant to retain fission products. However, noble gas and iodine will be a major portion of the MSR release source term.

Open fuel cycles present additional health physics concerns. The open fuel cycle associated with SCWRs (thermal option) and VHTRs have negative waste storage and associated effective dose impacts. These concerns include the long-term storage of high-level waste with the potential for the release of fission products and actinides to the environment. Closed fuel cycle options have positive nuclear proliferation and waste disposal aspects since actinides are destroyed during reactor operation.

2.8 Hydrogen Production

Hydrogen production for use as an alternative fuel is another application of advanced reactors, and four of the six Generation IV design concepts have hydrogen production as a design goal. Three basic approaches have been advanced for the nuclear energy production of hydrogen [5,7,12]. The first (nuclear-assisted steam reforming of natural gas) uses nuclear heat to reduce the amount of natural gas needed to produce hydrogen. Hot electrolysis is the

second approach and it produces oxygen and hydrogen from water using heat, not electricity. Finally, thermochemical cycles use a series of chemical reactions and high-temperatures to convert water into hydrogen and oxygen. All three of these processes use reactor heat as the basis for hydrogen production. Of these three, thermochemical hydrogen production is currently viewed as the most cost effective hydrogen production method [7,12].

The health physics aspects of hydrogen production depend on the reactor design used to generate process heat. Since only high-temperature reactor designs are candidates for hydrogen production, the optimum design matches the reactor output and hydrogen generation requirements. In addition, the nuclear reactor and chemical hydrogen production facility must be physically separated. Preliminary design studies suggest that a separation distance of at least a kilometer may be necessary to ensure that potential accidents in one facility do not affect the other [7,12].

2.9 Deployment of Generation IV Reactors

Generation IV reactors are projected to be deployed in the 2030s. The sodium-cooled fast reactor has the most optimistic deployment outlook which is somewhat expected since there is scalable operating experience from Generation II SFR designs. These deployment dates are contingent on the development of the Generation IV reactor types and resolution of the issues previously identified.

2.10 Generation IV Radiological Design Characteristics

From a radiological perspective, the Generation IV facility design should ensure that effective doses to plant workers and to members of the public are as low as reasonably achievable (ALARA). This is achieved through the design of systems, structures, and components (SSCs) that are reliable, easily maintained, and do not contribute to the radiological source term. The proposed radiological design characteristics are common to a variety of organizations involved in advanced reactor regulation and standards. This includes the US Nuclear Regulatory Commission and the International Atomic Energy Agency [5,7,12].

With these considerations, Generation IV SSCs should limit their radioactive source term. This entails the reduction in the concentrations of cobalt and nickel for materials in contact with the primary coolant to minimize the production of the ^{58}Co and ^{60}Co activation products. These isotopes are the major sources of radiation exposure during shutdown, maintenance, and inspection activities at Generation II and III light water reactors [5,6,8]. Exceptions to this design specification may be necessary to enhance component or system reliability and minimize component maintenance. However, the decision to utilize materials that produce ^{58}Co and ^{60}Co activation products must be made in a deliberate manner using ALARA considerations as a guide.

The resulting reduced radiation fields allow operations, maintenance, and inspection activities to be performed in a manner that leads to minimizing effective doses. Effective doses can also be maintained ALARA by incorporating the use of robotic technology to perform maintenance and surveillance in high radiation areas. The design should also accommodate remote and semi-remote operation, maintenance, and inspection activities to reduce the time spent in radiation fields. Reach rods and motor operators should be evaluated for incorporation into valves located in high radiation areas.

Generation IV SSCs should attain optimal reliability and maintainability to reduce the frequency and duration of maintenance requirements. This is particularly true for systems in contact with fluids cooling the reactor core. These requirements will reduce access, repair, and equipment removal times to limit the time spent in radiation fields. Adequate equipment spacing and job preparation areas facilitate access for maintenance, repair, and inspection. Modularized components facilitate their replacement or removal to a lower radiation area for repair.

The SSC design should facilitate the physical separation of radioactive and non-radioactive systems. High radiation sources should be located in separate shielded cubicles. In addition, equipment requiring periodic servicing or maintenance (e.g., pumps, valves, and control systems) should be physically separated from sources with higher radioactive materials concentrations including tanks and demineralizers.

The accumulation of radioactive materials in equipment and piping should be minimized. This is often accomplished using flushing connections to facilitate the removal of radioactive materials from system components. Locating drains at low points enhances the achievement of this design aspect. Piping should be seamless, and the number of

fittings minimized to reduce the accumulation of radioactive materials at seams and welds.

Systems that generate radioactive waste should be located close to waste processing systems to minimize the length of piping carrying these materials. The potential for pipe plugging is minimized by routing lines that carry resin slurries vertically. Large-radius bends should be used instead of elbows to limit the potential for pipe plugging.

The general design considerations lower worker effective doses and facilitate maintenance and surveillance activities. These considerations also foster plant modifications that will further reduce worker doses.

The aforementioned radiological design considerations are most easily met for water or gas cooled reactors, which have considerable Generation II and III operating experience. SFR designs also benefit from Generation II operational experience, but sodium systems do not have as much operational system experience as the light water systems. The MSR and LFR designs have limited operating experience and require significant development to achieve the performance levels currently available in operating water and gas cooled reactors.

2.11 Economic Considerations

At the most basic level, nuclear power plants utilize the fission of uranium, thorium, or plutonium to provide a heat source to boil water and produce steam that drives a turbine generator to produce electricity. As such, nuclear energy must compete with other energy sources and is ultimately judged by its production cost and public acceptance. Nuclear power has an added regulatory overhead that adds to its cost profile. Other energy sources such as natural gas do not have the regulatory burden attached to a nuclear power plant. Nuclear power also is associated with radiation and its deleterious effects. In the 20th Century, this association created at atmosphere of fear among a portion of the public that was reinforced by the accidents at Three Mile Island and Chernobyl.

In the first decade of the 21st Century, nuclear power appeared to be undergoing a renaissance with numerous organizations expressing interest in new Generation III plants. In the US, new plant designs were certified and a streamlined licensing approach contributed to a positive outlook for nuclear power. In addition, low interest rates promoted investment by nuclear utilities.

These positive conditions began to erode with declining world economic conditions and a major recession in the US. A combination of increasing capital costs (five to ten billion US dollars per reactor), additional regulatory requirements, and eroding public confidence following the 2011 Fukushima Daiichi accident further dampened the outlook for new nuclear power growth. In addition, recently discovered natural gas reserves and low natural gas prices have soured the outlook for a nuclear renaissance in the US.

The Fukushima Daiichi accident also led to a number of governments deciding to eliminate the nuclear option from their future energy plans. Although these conditions are not as favorable as those of the early 21st Century, the nuclear option still exists in the US and at least 60 nations have expressed interest in developing nuclear generating capability [3].

3.0 Health Physics Hazards

The essential health physics aspects of Generation II and III reactors are applicable to the Generation and IV systems. Under normal operating conditions, activation products including ^{58}Co and ^{60}Co will dominate worker effective doses. Assuming the effectiveness of health physics programs, internal doses will not be limiting. Accident releases will be dominated by noble gases and iodine.

It is also expected that maintenance and surveillance activities continue and that normal and outage activities are similar to contemporary operating reactors. Given these assumptions, subsequent discussion outlines generic and specific activities of health physics concern at Generation IV reactors.

3.1 Generic Health Physics Hazards

The power reactor health physicist must deal with a variety of issues. Internal and external dose control are not unique to the power reactor environment, but their implementation is dependent on the specific reactor environment and its operating characteristics. Examples of expected activities to be encountered in Generation IV reactors are provided.

A summary of the health physics concerns associated with generic power reactor activities are summarized in Table 4. Examples of these work activities include primary component maintenance

during outages and power operations, steam generator surveillance and repair, recirculation pipe replacement, spent fuel pool work activities, refueling operations, containment at power inspections, waste processing operations, component decontamination, and spill cleanup. The activities of Table 4 involve both internal and external exposure pathways [34,35]. Activation products, fission products, and hot particles (APFPHP) are common health physics concerns in many of these activities. The availability of fission products depends on fuel integrity.

LFR, MSR, and SFR Generation IV reactors have lead, molten salt, and sodium core coolants that will adhere to primary system components. The core coolant will contain activation products and fission products (APFP) and possibly transuranic isotopes. These radionuclides and their associated coolants present a unique maintenance challenge that will require component removal for conventional maintenance or development of specialized robotic repair methods.

From Generation II and III operating experience, activation of the core's ^{16}N coolant presents an operational concern in advanced PWR and BWR systems. ^{16}N doses are an operational concern near primary system piping. A similar coolant activation concern exists in Generation IV reactors. Isotopes of concern for the various Generation IV reactors include: GFR (^{15}O, ^{16}N, ^{17}N, and ^{19}O), SCWR (^{15}O, ^{16}N, ^{17}N, and ^{19}O), MSR (^{16}N, ^{18}F, ^{19}O, ^{20}F, ^{22}Na, ^{23}Ne, and ^{24}Na), and SFR (^{20}F, ^{22}Na, ^{23}Ne, and ^{24}Na).

3.2 Specific Health Physics Hazards

The generic descriptions of Table 4 provide an overview of the radiation hazards that affect task performance at Generation IV reactors. Knowledge of these generic hazards facilitates the introduction of specific Generation IV hazards. For specificity, selected tasks and facility conditions are chosen to illustrate the health physics hazards. These tasks and conditions are the buildup of radioactive material in components such as filters, demineralizers, and waste gas decay tanks; activation of reactor components; fuel damage; reactor coolant system leakage; hot particles; and effluent releases.

3.2.1 Buildup of Activity in Filters, Demineralizers, and Waste Gas Decay Tanks

The reduction of activity concentrations in radioactive fluids is an important consideration in minimizing worker doses. Filters, demineralizers, and waste gas decay tanks are often used to reduce fluid activity levels.

Air filters trap airborne radioactive material, liquid filters remove suspended particulates, demineralizers use an ion-exchange technique to retain radioactive material from liquid streams, and waste gas decay tanks collect fission gases and iodine removed from the primary coolant [36]. Light water reactor experience has demonstrated the effectiveness of these components to reduce the activity levels that also affects the source term available for release. Similar techniques will be developed for the MSR, LFR, and SFR designs. This is important from a health physics perspective, because all power reactor types benefit from minimizing their radioactive source terms.

The activity that accumulates in filters and demineralizers are primarily activation products. Fission products accumulate if fuel damage has occurred. MSR designs must contend with fission products and actinides since the fuel forms a eutectic mixture with the salt coolant. Metal cooled reactors involve complex interactions between the coolant, fission products, and fuel form.

Filters are commonly used to reduce effluent concentrations. A variety of air filter types (e.g., high-efficiency particulate air and charcoal) remove airborne activation products, fission products, and iodine. Liquid filters vary in construction and composition, but all types mechanically remove radioactive material suspended in fluid streams.

Filter performance is affected by the coolant medium and interaction with fuel. Although similar fission products and actinides are generated in both light water reactors and metal cooled reactor types, their chemical interactions with the metal coolant and release to the containment are unique to the specific coolant type. Since most metal research has been performed on liquid sodium coolant, the discussion focuses on that material [33].

In a severe sodium cooled reactor accident, the noble gases would be immediately released to the containment. The volatile halogens (iodine and bromine), alkali metals (cesium and rubidium), alkali earths (strontium and barium), and chalcogens (tellurium and selenium) are highly soluble in liquid sodium metal, some of which form soluble sodium compounds. These compounds are released from the sodium coolant by vaporization from the liquid

surface or though aerosol production if the sodium is burning.

Sodium aerosols tend to agglomerate into rather large fluffy particles that quickly separate from their environment. In addition, sodium chemically reacts with several fission products, forming compound aerosols that settle quickly. Sodium aerosols tend to agglomerate into large particles that tend to precipitate close to the release point during atmospheric dispersion. These properties must be considered when analyzing fission product transport and retention on filters.

Demineralizers remove activity from fluid systems using an ion-exchange process. In Generation II reactors, the radiation levels inside demineralizer cubicles associated with spent fuel cleanup systems can exceed the US regulatory criteria for a very high radiation areas (5 Gy/h at 1 m from the source) [37,38]. Following fuel damage, demineralizer radiation levels increase dramatically with the release of fission products through the fuel fission product barrier.

MSR demineralizer or equivalent systems will be unique because the fuel and salt coolant form a eutectic mixture. Activation products and fission products are removed from the coolant as part of the facility's design. Demineralizer loading and change out are unique aspects of the MSR, and the selection of ion exchange or equivalent media requires careful selection to avoid radiation degradation of the media.

Waste gas decay tanks accumulate fission gases and iodine that are removed from the reactor coolant. The radioactive material is stored and retained in these tanks until it meets the criteria for release to the environment. These tanks and supporting systems to strip fission gases from the coolant should be integral Generation IV systems.

The buildup and decay of radioactive material in a system is described in terms of production equations [36]. Production equations describe a variety of physical processes, and are important in a number of health physics applications [36]. The activity deposited into a filter, demineralizer bed, or waste gas decay tank is described in terms of production equations.
The buildup of activity of isotope *i* (A_i) on a filter, in a demineralizer bed, or in a waste gas decay tank is determined from the system properties and isotopes present in the fluid entering these components. As noted previously, additional components and physical mechanisms must be considered with metal and salt coolants, but these systems are also described by production equations. The following discussion assumes a constant rate of production and the buildup and removal terms have an exponential form. Other factors introduced by changes in chemical composition, particle interactions, or chemical interactions are described by production equations if their associated removal terms are exponential.

Important parameters impacting the buildup of the activity of isotope i in filters, demineralizers, or waste gas decay tanks include the concentration of the isotope in the fluid entering the device (C_i), the system flow rate (F), the time the filter or demineralizer is operating or processing influent (t_{op}), and the time the system is isolated (t_{decay}) from the influent stream:

$$\mathbf{A_i = \frac{C_i e_i F}{\lambda_i}\left(1 - \exp(-\lambda_i t_{op})\right)\exp(-\lambda_i t_{decay})} \quad (1)$$

where e_i is the efficiency of the filter or demineralizer for removal of isotope i and λ_i is the radioactive decay constant of isotope i. Fluids containing multiple isotopes require the application of Eq. 1 for each nuclide present in the influent stream.

The types of radioactive material deposited in filters, demineralizers, and waste gas decay tanks vary with the specific design. Activation products are design specific as noted in Table 2. These activation products vary considerably and depend on the coolant type, materials used in the construction of the primary system, fuel type, and the reactor's neutron spectrum (i.e., thermal or fast).

Fission product generation depends on the specific fuel composition and neutron spectrum incorporated into the design. For example, fission products are derived from a variety of nuclides including ^{233}U, ^{235}U, ^{239}Pu, and ^{241}Pu for thermal fission and ^{232}Th and ^{238}U for fast fission reactor fuels.

3.2.2 Activation of Reactor Components

The direct irradiation of reactor components and the activation of corrosion products are also described in terms of production equations [36]. Corrosion or wear products dissolved or suspended in the primary coolant are subjected to the core's neutron fluence. Activation occurs by a variety of neutron-induced reactions, and the nuclides produced depend on the neutron spectrum and fluence impinging upon the

material in the core region. Specific activation mechanisms are illustrated in Table 2.

The activity resulting from an activation reaction has the specific form [36]:

$$\mathbf{A_i = N_i \sigma \phi \left(1 - \exp(-\lambda_i t_{irr})\right) \exp(-\lambda_i t_{decay})} \quad (2)$$

where N_i are the number of target atoms that are activated, σ is the microscopic cross section for the activation reaction, ϕ is the fluence rate or flux inducing the activation reaction, t_{irr} is the time the target is irradiated or exposed to the core flux, and t_{decay} is the decay time or time the target was removed from the reactor's core region or activating flux.

Activated material presents an internal as well as external dose concern. In a power reactor environment, external radiation sources are dominated by ^{60}Co in most Generation II reactors and ^{58}Co in late Generation II systems and many Generation III PWRs and BWRs. External doses are dominated by activation sources that emit beta and gamma radiation types.

3.3.3 Fuel Damage

With the exception of MSRs, a nuclear reactor contains three barriers to prevent fission products from escaping from the reactor core to the environment. These barriers are the fuel matrix and fuel element cladding or coating, the reactor coolant system and included piping, and the containment building. A breach of any of these barriers enhances the probability that radioactive material will be released to the environment.

The robustness of the fuel fission product barrier depends on its specific composition. In PWRs, BWRs, and Canadian Deuterium (CANDU) reactors, the fuel fission product barriers consist of UO_2 pellets enclosed within a stainless steel or zirconium alloy tube. In VHTRs, the fuel is coated in a ceramic, and the fuel fission product barrier is the SiC or ZrC fuel coating and the fuel material.

Fuel barrier damage facilitates the release of fission products contained between the fuel pellet and cladding (gap activity) or between the ceramic coating and fuel and increases the primary coolant activity. Noble gas activity entering the primary coolant is either released to the containment atmosphere via leakage paths or to off gas systems. These gaseous fission products are an early indication that a fuel cladding/coating failure or mechanical damage to the cladding/coating has occurred. BWRs normally detect fuel failure by detection of fission gases in the off gas system. However, PWRs normally monitor the primary coolant line or letdown filter lines for these fission products or monitor the containment atmosphere for released noble gases (e.g., xenon and krypton) and their daughter products. The analysis of primary coolant samples by gamma spectroscopy is a routine confirmatory action.

Reliable fuel performance in a Generation IV system must be demonstrated. GFR, VHTR, SCWR, and SFR systems derive fuel performance experience based on Generation II and III designs that operated at lower temperatures. The effects of increased temperatures can be significant since chemical reaction and corrosion rates tend to increase as temperatures increase. Fuel performance in these systems must be achieved to ensure a reliable fission product barrier.

There is significantly less data regarding the fuel performance in LFRs and MSRs. The ability of the LFR fuel and lead coolant to retain fission products during normal operations and transient conditions have yet to be demonstrated.

The MSR concept does not have a conventional fuel fission product barrier because the fissile material and the molten salt coolant form a eutectic mixture. If the salt effectively retains the fission products, then the salt solution could be considered a type of barrier. However, the retention of fission gases in the salt coolant appears to be an open issue for MSRs.

A severe accident involving MSR fuel has a different character than light water uranium dioxide fuel. In a MSR accident in which core cooling capability is lost and the solidified fuel plug melts (Section 2.3), the fuel storage tanks are assumed to provide a barrier equivalent to that of the reactor coolant system. If the storage tank barrier fails, a unique accident condition exists that requires additional evaluation. Its severity depends on the capability of the MSR coolant to retain fission products when its temperature is elevated and the capability of the containment to withstand the stress induced by a breached fuel storage tank. If the containment is breached, the release of fission products to the environment will depend on the capability of the MSR coolant to retain fission products and the type of aerosols generated during the accident.

3.3.4 Reactor Coolant System Leakage

Since reactors are electro-mechanical systems, leakage from the primary coolant system is an undesirable but inevitable problem [5,6,8]. This leakage occurs in Generation II and III systems and will also occur in Generation IV reactors. Generation II and III leakage is well quantified, but some Generation IV systems use metal and salt coolants instead of light water, and their leakage characteristics during operating conditions have yet to be demonstrated.

Value stems, pump seals, value packing, and instrument line connections provide pathways for small leaks that contaminate local areas. This contamination must be controlled in order to limit station external and internal doses. In addition to primary system leaks, health physicists must address leakage from the primary to secondary systems for reactors using steam generators.

Leakage of primary coolant from steam generator tubes to the secondary system presents a health physics concern because additional plant areas become contaminated. Since the secondary components are clean systems, the presence of contamination has a negative impact on facility operations and expands areas requiring stringent radiological controls.

Secondary coolant contamination has a number of negative health physics aspects. The secondary activity tends to concentrate in components such as the main steam isolation valves and high-pressure turbine piping resulting in surface contamination areas and local hot spots. Secondary ion exchange resins and filters become contaminated which adds to the facility's contamination control requirements and increases the volume of radioactive waste. Steam generators cleanup systems also become contaminated. Contaminated secondary system areas increase health physics survey requirements and associated decontamination activities.

Primary to secondary leakage increases the likelihood of a release radioactive material (e.g., noble gases and iodine) to the environment. The most likely release pathways are through a secondary system relief valve or through the condenser air ejector.

Leakage of metal or salt coolants present additional challenges beyond those encountered in light water reactors. In addition to the presence of fission products and their dispersion by a decay heat source, metal and salt coolants present the possibility for chemical reaction and phase transition energies to enhance the dispersion of fission products. Although liquid sodium reactions have been studied, less is known about other metal and salt coolants and their reactions with fission products and construction materials encountered as the coolant leaks onto surfaces supporting the reactor coolant system. These reactions and their ability to mobilize fission products will govern the health physics consequences of these leakage events in LFR and MSR Generation IV systems.

3.3.5 Hot Particles

The maintenance of pumps, valves, and primary system components and piping create small particles during the process of testing, cutting, grinding, and welding. Operation of valves and pumps leads to wearing of active surfaces and this wear produces small particulate material. Cladding erosion and failures or erosion of control rod surfaces contribute additional particulate matter to the reactor coolant system. This material is often too small to be removed by the reactor coolant system's filters, and it passes through the core and is activated by the neutron fluence. The result of this activation is the creation of highly activated, microscopic material or a *hot particle* [39,40]. Given the nature of the MSR coolant-fuel eutectic, hot particles could present an interesting health physics challenge in these Generation IV reactors.

Hot particles are composed of activation products, fuel fragments, and fission fragments depending upon the integrity of the fuel fission product barrier. Particles may contain either single isotopes or a large number of radioisotopes. Hot particles present a skin dose hazard. Beta radiation is the dominant contributor to the skin dose, but gamma contributions can approach about 30% of the beta dose contribution [6,8,39]. Hot particles can also be deposited in the ear and eye, and inhalation and ingestion are additional pathways of entry into the body [6,8,39,40].

The absorbed dose to the skin from a hot particle is given by the relationship:

$$\mathbf{D} = \frac{\mathbf{t}}{\mathbf{S}} \sum_{\mathbf{i}} \mathbf{A_i F_i} \qquad (3)$$

where D is the absorbed dose to the skin from the hot particle, A_i is the particle activity for radionuclide i, F_i is the dose factor for radionuclide i (Gy-m^2 /MBq-h), t is the residence time on the skin,

S is the area over which the dose is averaged, and *i* is the number of radionuclides in the hot particle. Similar relationships can be developed for the hot particle dose to the eye, ear, human respiratory tract, and human alimentary tract.

3.3.6 Environmental Releases

The effluents that characterize a facility depend on the core materials, reactor materials, and specific design aspects of the Generation IV system. Examples of the unique nuclides that will likely appear in a facility are summarized in Table 2.

Light water and heavy water reactor effluents are primarily isotopes generated through the activation and fission processes. Although off gas systems are designed to trap most gaseous effluents, quantities of noble gases, ^{3}H, ^{14}C and iodine isotopes are available for release. Their release is facilitated by defects in the fuel clad/coating or failure of the MSR coolant to retain fission products.

Isotope production mechanisms are design dependent. For example, tritium arises from the neutron activation of the light water coolant [$^{2}H(n, \gamma)^{3}H$] and from tertiary fission. Tritium production is enhanced in CANDU reactors that use a D_2O coolant. In a PWR, tritium is also produced from neutron capture in ^{10}B used for reactivity control [^{10}B (n, 2α) ^{3}H] and from neutron capture in ^{6}Li used for chemistry control [$^{6}Li(n, \alpha)^{3}H$]. ^{14}C is usually produced from the $^{14}N(n, p)^{14}C$ and the $^{17}O(n, \alpha)^{14}C$ reaction in a CANDU reactor. In GFRs and VHTRs, tritium is produced in the gas coolant reactions $^{4}He(\gamma, p)^{3}H$ and $^{4}He(n, d)^{3}H$. MSR salt coolants produce tritium via $^{6}Li(n, \alpha)^{3}H$.

Liquid effluents include fission and activation products as well as tritium. Tritium is the dominant liquid effluent in PWRs. The quantity of fission products in liquid waste depends on the integrity of the fuel fission product barrier. Liquid waste cleanup systems, including filtration and demineralization, remove most of these radionuclides from the effluent stream. Similar effluents are expected in Generation IV reactors. However, the high temperature SFR, LFR, and MSR reactors are departures from light water and gas cooled reactors and their liquid release characteristics are not well defined.

Fission product radionuclides generated from binary fission include ^{85}Kr, ^{87}Kr, ^{88}Kr, ^{133}Xe, ^{135}Xe, ^{137}Xe, ^{131}I, ^{137}Cs, ^{137}Ba, ^{141}Ce, ^{144}Ce, ^{103}Ru, ^{106}Ru, ^{103}Rh, ^{106}Rh, ^{90}Sr and ^{90}Y. Activation products are produced by neutron capture in core materials and in materials in the vicinity of the nuclear core including chemical control agents dissolved in the primary coolant, stainless steel or stellite corrosion and wear products resulting from system maintenance or operation, primary coolant system piping, the reactor vessel, and core structural material. Unique Generation IV activation products for the various reactor types are summarized in Table 2.

Generation IV reactor activation products are produced from a variety of reactions including: $^{54}Fe(n, p)^{54}Mn$, $^{58}Fe(n, \gamma)^{59}Fe$, $^{57}Co(n, \gamma)^{58}Co$, $^{58}Ni(n, p)^{58}Co$, $^{59}Co(n, \gamma)^{60}Co$, and $^{94}Zr(n, \gamma)^{95}Zr$. The aforementioned (n, γ) reactions are normally induced by thermal neutrons, and the (n, p) reactions are initiated by fast neutrons. The specific activation products will vary with specific reactor type and generation. As an illustration, the activation products and associated effluents in gas cooled reactors are briefly reviewed.

Since gas cooled reactors have different materials of construction than water cooled reactors, different radionuclides inventories and effluents are expected. As an illustration, the effluents from CO_2 and ^{4}He gas cooled reactors are outlined in subsequent discussion [5,6,8].

Advanced CO_2 gas cooled reactors developed in the United Kingdom are graphite moderated facilities. Activation of the CO_2 primary coolant produces ^{14}C, ^{16}N, and ^{41}Ar, and activation of the graphite moderator yields ^{3}H, ^{14}C, and ^{35}S. Fission products similar to those noted for PWRs, BWRs, and CANDUs are also produced. Their possibility of release depends on the integrity of the fuel fission product barrier.

The graphite moderator may contain trace sulfur and chlorine impurities that lead to ^{35}S production through the $^{34}S(n, \gamma)^{35}S$ and $^{35}Cl(n, p)^{35}S$ reactions. Graphite may also contains lithium impurities that upon capture of thermal neutrons produces tritium through the $^{6}Li(n, \alpha)^{3}H$ reaction.

One of the key features affecting the effluent releases in helium cooled reactors is the concentration of impurities in the graphite moderator. These impurities vary with the type of graphite used in the design. It is expected that a variety of elements will be found in the graphite moderator including boron, cesium, calcium, carbon, chlorine, cobalt, helium, iron, lithium, nickel, nitrogen, niobium, and uranium [5]. The concentrations of these elements directly affect the effluent

concentrations of their activation products such as ^{3}H and ^{14}C.

Helium cooled reactor metallic materials are dominated by chromium, iron, and nickel with smaller quantities of cobalt and molybdenum. These elements lead to activation products including ^{55}Fe, ^{59}Ni, ^{60}Co, and ^{63}Ni.

The previous discussion illustrates the uncertainty involved in discussion of gas cooled reactor effluents. The specific design requirements including materials specifications govern the radionuclides produced and their abundance. As an illustration, the graphite specification controls the impurities and their concentrations. The allowance for impurities in the graphite has a significant impact on the production of activation products. Therefore, identical Generation IV helium cooled reactors could have different effluent radionuclide characteristics if their graphite specifications are not the same.

3.3.7 Advanced Reactor ALARA Measures

One of the Generation IV radiological goals is minimizing worker radiation doses [7-12]. Reactor components are designed to be nearly maintenance free and minimize the production of activation products. In particular, cobalt alloys are restricted. This minimizes a major source term of activation products that occurs in Generation II facilities (e.g., ^{60}Co).

Component arrangement and accessibility is optimized in Generation IV reactors. These features enhance task completion, minimize radiation doses, and facilitate operability testing of reactor components. For example, heat exchangers, tanks, and vessels are designed to minimize the collection of radioactive material and to facilitate the removal of any radioactive material collecting within their boundaries. Components are arranged to allow for sufficient room for maintenance, surveillance, and inspection activities.

Passive safety systems are incorporated to preserve fission product barriers and to minimize the consequences of events that led to the TMI-2 and Fukushima Daiichi accidents. Design enhancements include additional core cooling inventories associated with safety systems, improved materials to minimize corrosion of components such as steam generator tubes, enhanced control room instrumentation to provide indication of abnormal conditions, enhanced core cooling capability, and improvements in emergency electrical supply capabilities during loss of power events [10].

4.0 Nuclear Proliferation

The nuclear non-proliferation treaty provides for nations to acquire nuclear technology (e.g., fission reactors, fuel reprocessing facilities, and uranium enrichment systems), and most of these facilities are subject to monitoring [14,26,31,32]. These facilities are monitored and inspected by the International Atomic Energy Agency, which ensures that fissile material is not diverted for other purposes (e.g., military or criminal uses).

Nuclear proliferation concerns arise from the enrichment of uranium, reactor operation, and reprocessing spent reactor fuel. The advanced centrifuges and laser enrichment technologies have the capability to produce highly enriched uranium. Reactor operation produces ^{239}Pu that if extracted through reprocessing can be diverted to weapons production. Generation IV reactors are designed to limit the proliferation impact of reactor produced ^{239}Pu.

Although Generation IV reactors have a proliferation resistance design goal, experience with the control of centrifuge technology suggests that reactor production of plutonium requires careful monitoring. Reactors produce copious quantities of ^{239}Pu through the neutron capture reaction

$$^{238}U + n \rightarrow {}^{239}U \xrightarrow{\beta^-} {}^{239}Np \xrightarrow{\beta^-} {}^{239}Pu$$

About one third of the power output of the reactor is derived from the fission of ^{239}Pu [6,8,16]. In addition, fuel discharged from a light water reactor (spent fuel) contains significant ^{239}Pu. The handling and processing of spent fuel creates a proliferation concern that requires oversight to ensure the ^{239}Pu is not recovered and diverted to illicit weapons production.

The options for a 21st Century fuel cycle depend on the acceptance, development, and deployment of new nuclear generating capacity. Although the development of a new generation of nuclear power plants in the US has lost momentum following the Fukushima Daiichi accident, proliferation resistance is likely to be a key issue in any global resurgence of nuclear power.

Any resurgence in nuclear power will ultimately depend on the resistance of new nuclear power facilities and their support facilities (e.g., uranium enrichment and fuel fabrication) to proliferation of

weapons-grade nuclear material. Nuclear power production is intimately linked to proliferation because the technologies used in power production overlap with those used in the production of fissionable material for nuclear weapons.

A 2005 American Physical Society (APS) report [26] makes a number of recommendations regarding the successful reestablishment of nuclear power operations in the US. These recommendations are related to ensuring the proliferation resistance of power reactors and fuel reprocessing activities. Four specific recommendations were provided in the APS report.

A strong research and development program on advanced safeguards technology is the first APS recommendation. The second recommendation calls for making proliferation resistance a high priority in the design and development of future nuclear energy systems. These systems should be open to international inspections and should exemplify the technologies that the US would suggest be implemented on an international basis. The third recommendation is to increase international nuclear security and safeguards cooperation. Expansion of US efforts similar to those in place with Japan and Russia would be warranted. The final recommendation focuses on spent fuel reprocessing, but no specific position was advocated.

Currently no option exists in the US to reprocess spent fuel in order to recover and recycle its valuable constituents. In order to implement reprocessing several technical decisions would need to be made. In addition, political support is required for spent fuel reprocessing to become an accepted national policy.

The proliferation concerns raised by the APS report are minimized with Generation IV reactors operating in a closed fuel cycle with the full recycling of actinides. Generation IV reactor concepts have the potential to meet power demands and proliferation concerns, but significant research and development is required to turn these candidate reactors types into members of an operating nuclear fleet.

5.0 Conclusions

Generation IV reactors offer the potential for enhancing reactor safety, limiting the production of high-level waste, and minimizing the proliferation potential for the reactor production of ^{239}Pu. These reactors employ unique core coolants and produce a variety of isotopes not usually encountered in Generation II and III reactors. The health physics challenges encountered in Generation IV reactors will be similar to Generation II and III designs, but lead, sodium, and molten salt coolants will challenge traditional health physics practices during refueling operations, primary coolant system component maintenance, and filter and demineralizer media replacement activities. These health physics challenges are manageable, but may require significant resources particularly for the initial Generation IV reactors.

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Table 1

Generation IV Reactor Concept Characteristics[a]

Reactor Technology	Power Rating (MWe)	Operating Temperature (^oC)[b]	Fuel Cycle Options	Economic Justification
Gas-cooled fast reactors	1200	850	Closed	Electricity and hydrogen are produced.
Lead-cooled fast reactors	20-180 300-1200 600-1000	480-800	Closed	Electricity and hydrogen are produced
Molten salt epithermal reactors	1000	700-800	Closed	Electricity and hydrogen are produced
Sodium-cooled fast reactors	50-150 300-600 600-1500	550	Closed	Electricity is produced
Supercritical water-cooled reactors (thermal and fast versions)	300-700 1000-1500	510-625	Open Closed	Electricity is produced
Very high temperature, helium-cooled, graphite moderated thermal reactors	250-300	900-1000	Open	Electricity and hydrogen are produced

[a] Derived from Gen IV International Forum Report (2009) and http://www.gen-4.org/Technology/roadmap.htm (2013).

[b] A range of values are presented by the Generation IV International Forum [7,9,12].

Table 2 Activation Products Produced in Materials Unique to Generation IV Fission Power Reactors			
Nuclide	**Half-Life**	**Decay Mode**	**Production Mode**
^{3}H	12.3 y	β^-	GFRs and VHTRs (^{4}He gas coolant): $^{4}He(\gamma, p)^{3}H$ and $^{4}He(n, d)^{3}H$ MSRs (Lithium fluoride salt coolant): $^{6}Li(n, \alpha)^{3}H$
^{10}Be	$1.56x10^{6}$ y	β^-	MSRs (beryllium fluoride salt coolant): $^{9}Be(n, \gamma)^{10}Be$
^{14}C	5715 y	β^-	GFRs, MSRs, and VHTRs (graphite moderator): $^{14}N(n, p)^{14}C$ and $^{13}C(n, \gamma)^{14}C$ GFRs (gas coolant) and SCWRs (water coolant): $^{17}O(n, \alpha)^{14}C$
^{15}O	2.037 min	β^+ γ	GFRs (gas coolant) and SCWRs (water coolant): $^{16}O(n, 2n)^{15}O$ and $^{16}O(\gamma, n)^{15}O$
^{16}N	7.13 s	β^- γ	GFRs (gas coolant) and SCWR (water coolant): $^{16}O(n, p)^{16}N$ MSRs (fluoride salt coolant): $^{19}F(n, \alpha)^{16}N$
^{17}N	4.174 s	β^- γ n	GFRs (gas coolant) and SCWR (water coolant): $^{17}O(n, p)^{17}N$
^{18}F	1.8293 h	β^+ γ	MSRs (fluoride salt coolant): $^{19}F(n, 2n)^{18}F$
^{19}O	26.9 s	β^- γ	GFRs (gas coolant) and SCWR (water coolant): $^{18}O(n, \gamma)^{19}O$ MSRs (fluoride salt coolant): $^{19}F(n, p)^{19}O$
^{20}F	11.1 s	β^- γ	MSRs (fluoride salt coolant): $^{19}F(n, \gamma)^{20}F$ SFRs (liquid sodium coolant): $^{23}Na(n, \alpha)^{20}F$
^{22}Na	2.604 y	β^+ γ	MSRs (sodium salt coolant) and SFRs (liquid sodium coolant): $^{23}Na(n, 2n)^{22}Na$ and $^{23}Na(\gamma, n)^{22}Na$

Table 2
Activation Products Produced in Materials Unique to Generation IV Fission Power Reactors

Nuclide	Half-Life	Decay Mode	Production Mode
^{23}Ne	37.1 s	β^- γ	MSRs (sodium salt coolant) and SFRs (liquid sodium coolant): ^{23}Na(n, p)^{23}Ne
^{24}Na	14.97 d	β^- γ	MSRs (sodium salt coolant) and SFRs (liquid sodium coolant): ^{23}Na(n, γ)^{24}Na GFRs (in core materials): ^{24}Mg(n, p)^{24}Na
^{25}Na	59.3 s	β^- γ	GFRs (in core materials): ^{25}Mg(n, p)^{25}Na
^{27}Mg	9.45 min	β^- γ	GFRs (in core materials): ^{26}Mg(n, γ)^{27}Mg GFRs (in core materials): ^{30}Si(n, α)^{27}Mg
^{28}Al	2.25 min	β^- γ	GFRs (in core materials) and VHTRs (fuel coating): ^{28}Si(n, p)^{28}Al
^{29}Al	6.5 min	β^- γ	GFRs (in core materials) and VHTRs (fuel coating): ^{29}Si(n, p)^{29}Al
^{31}Si	2.62 h	β^- γ	GFRs (in core materials) and VHTRs (fuel coating): ^{30}Si(n, γ)^{31}Si
^{35}S	87.2 d	β^-	GFRs, MSRs, and VHTRs (graphite moderator): ^{35}Cl(n, p)^{35}S and ^{34}S(n, γ)^{35}S
^{38}Cl	37.2 min	β^- γ	GFRs, MSRs, and VHTRs (graphite moderator): ^{37}Cl(n, γ)^{38}Cl
^{45}Ca	162.7 d	β^- γ	GFRs (in core materials): ^{48}Ti(n, α)^{45}Ca
^{45}Ti	3.078 h	β^+ γ	GFRs (in core materials): ^{46}Ti(n, 2n)^{45}Ti and ^{46}Ti(γ, n)^{45}Ti
^{46}Sc	83.8 d	β^- γ	GFRs (in core materials): ^{46}Ti(n, p)^{46}Sc GFRs, MSRs, and VHTRs (graphite moderator): ^{45}Sc(n, γ)^{46}Sc

Table 2 Activation Products Produced in Materials Unique to Generation IV Fission Power Reactors			
Nuclide	**Half-Life**	**Decay Mode**	**Production Mode**
^{47}Ca	4.536 d	β^- γ	GFRs (in core materials): ^{46}Ca(n, γ)^{47}Ca and ^{50}Ti(n, α)^{47}Ca
^{47}Sc	3.349 d	β^- γ	GFRs (in core materials): ^{47}Ti(n, p)^{47}Sc
^{48}Sc	43.7 h	β^- γ	GFRs (in core materials): ^{48}Ti(n, p)^{48}Sc
^{51}Ti	5.76 min	β^- γ	GFRs (in core materials): ^{50}Ti(n, γ)^{51}Ti
^{82}Br	1.471 d	β^- γ	GFRs, MSRs, and VHTRs (graphite moderator): ^{81}Br(n, γ)^{82}Br
^{88}Y	106.63 d	β^+ γ	GFRs (in core materials): ^{89}Y(n, 2n)^{88}Y and ^{89}Y(γ, n)^{88}Y
^{89}Sr	50.61 d	β^- γ	GFRs (in core materials): ^{89}Y(n, p)^{89}Sr GFRs (in core materials), MSRs (coolant component), and VHTRs (fuel coating): ^{92}Zr(n, α)^{89}Sr
^{89m}Y	15.7 s	γ	GFRs (in core materials): ^{89}Y(n, n')^{89m}Y
^{89}Zr	3.27 d	β^+ γ	GFRs (in core materials), MSRs (coolant component), and VHTRs (fuel coating): ^{90}Zr(n, 2n)^{89}Zr and ^{90}Zr(γ, n)^{89}Zr
^{90}Y	2.669 d	β^-	GFRs (in core materials): ^{89}Y(n, γ)^{90}Y and ^{93}Nb(n, α)^{90}Y GFRs (in core materials), MSRs (coolant component), and VHTRs (fuel coating): ^{90}Zr(n, p)^{90}Y
^{90m}Y	3.19 h	β^- γ	GFRs (in core materials): ^{89}Y(n, γ)^{90m}Y
^{92}Nb	3.5x10^7 y	γ	GFRs (in core materials): ^{93}Nb(n, 2n)^{92}Nb and ^{93}Nb (γ, n) ^{92}Nb
^{92m}Nb	10.13 d	γ	GFRs (in core materials): ^{93}Nb(n, 2n)^{92m}Nb and ^{93}Nb (γ, n) ^{92m}Nb

Table 2 Activation Products Produced in Materials Unique to Generation IV Fission Power Reactors			
Nuclide	**Half-Life**	**Decay Mode**	**Production Mode**
^{93}Zr	1.5x10^{6} y	β^- γ	GFRs (in core materials), MSRs (coolant component), and VHTRs (fuel coating): ^{92}Zr(n, γ)^{93}Zr
^{93m}Nb	16.1 y	γ	GFRs (in core materials): ^{93}Nb(n, n')^{93m}Nb
^{94}Nb	2.0x10^{4} y	β^- γ	GFRs (in core materials): ^{93}Nb(n, γ)^{94}Nb
^{94m}Nb	6.263 min	β^- γ	GFRs (in core materials): ^{93}Nb(n, γ)^{94m}Nb
^{95}Zr	64.02 d	β^- γ	MSRs (coolant component) and VHTRs (fuel coating): ^{94}Zr(n, γ)^{95}Zr
^{97}Zr	16.75 h	β^- γ	MSRs (coolant component) and VHTRs (fuel coating): ^{96}Zr(n, γ)^{97}Zr
^{152}Eu	13.54 y	β^- β^+ γ	GFRs, MSRs, and VHTRs (graphite moderator): ^{151}Eu(n, γ)^{152}Eu
^{203}Pb	2.164 d	γ	LFRs (lead coolant): ^{204}Pb(n, 2n)^{203}Pb and ^{204}Pb(γ, n)^{203}Pb
^{204m}Pb	1.12 h	γ	LFRs (lead coolant): ^{204}Pb(n, n')^{204m}Pb
^{205}Pb	1.5x10^{7} y	EC	LFRs (lead coolant): ^{204}Pb(n, γ)^{205}Pb
^{209}Pb	3.25 h	β^-	LFRs (lead coolant): ^{208}Pb(n, γ)^{209}Pb
^{210}Pb	22.3 y	β^- γ α	LBFRs (bismuth coolant): ^{209}Bi(n, p)^{209}Pb+n$\rightarrow$^{210}Pb LFR(lead coolant): ^{208}Pb(n, γ)^{209}Pb followed by ^{209}Pb(n, γ)^{210}Pb
^{210}Bi	5.01 d	β^- γ α	LBFRs (bismuth coolant): ^{209}Bi(n, γ)^{210}Bi

Table 2
Activation Products Produced in Materials Unique to Generation IV Fission Power Reactors

Nuclide	Half-Life	Decay Mode	Production Mode
^{210m}Bi	3.0x10^{6} y	α γ	LBFRs (bismuth coolant): ^{209}Bi(n, γ)^{210m}Bi
^{210}Po	138.38 d	α γ	LBFR (bismuth coolant): ^{209}Bi(n, γ)^{210m}Bi $\xrightarrow{\beta^-}$ ^{210}Po

Table 3
Generation IV Industrial Forum Agreements

Design	Canada	France	Japan	South Korea	South Africa	Switzerland	US	EU	China	Russia	UK	Brazil	Argentina
SFR		[a]	[a]	[a]			[a]	[a]	[a]	[a]			
VHTR	[a]	[a]	[a]	[a]		[a]	[a]	[a]	[a]				
GFR		[a]	[a]			[a]		[a]					
SCWR	[a]		[a]					[a]		[a]			
LFR			[b]					[b]		[b]			
MSR		[b]						[b]					

[a] Signatory of formal agreement.
[b] Memorandum of Understanding.

Table 4 Generation IV Power Reactor Generic Work Activities and Associated Health Physics Hazards		
Work Activity	**Reactor Types**	**Radiological Concern[a]**
Primary Component Maintenance During a Refueling or Maintenance Outage	All	APFPHP[b] personal contaminations APFP direct dose Core coolant adhering to primary system components (LFRs, MSRs, and SFRs)
Primary Component Maintenance During Power Operations	All	APFPHP[b] personal contaminations APFP direct dose Core coolant direct dose[c] Core coolant adhering to primary system components (LFRs, MSRs, and SFRs)[c] Neutrons
Steam Generator Eddy Current Surveillance and Tube Repair During an Outage	All Reactors with Steam Generators (Independent of type)	APFPHP[b] personal contaminations APFP direct dose Core coolant adhering to steam generator tubes in LFRs, MSRs, and SFRs.
Recirculation Pipe Replacement	SCWR Systems Depending on Final Design	APFPHP[b] personal contaminations APFP direct dose
Spent Fuel Pool Activities Including Fuel Rearrangement, Control Rod Replacement, Fuel Assembly Reconstitution, and Clean-up Activities	All except MSR[d]	APFPHP[b] personal contaminations Criticality APFP direct dose
Refueling Operations	All	APFPHP[b] personal contaminations APFP direct dose Core coolant adhering to primary system components (LFRs, MSRs, and SFRs) Core coolant direct dose[c] Tritium Criticality

Table 4
Generation IV Power Reactor Generic Work Activities and Associated Health Physics Hazards

Work Activity	Reactor Types	Radiological Concern[a]
Containment at Power Inspections[e]	All	APFPHP[b] personal contaminations APFP direct dose Noble Gases Tritium Neutrons Core coolant direct dose[c]
On-line Refueling, Radioactive Waste Processing, and Actinide Recycle	MSR	APFPHP[b] personal contaminations APFP direct dose Core coolant direct dose[c] Criticality
Radioactive Waste Processing	All	APFPHP[b] personal contaminations APFP direct dose
Component Decontamination	All	APFPHP[b] personal contaminations APFP direct dose Core coolant direct dose[c] Core coolant adhering to primary system components (LFRs, MSRs, and SFRs)
Spill Cleanup	All	APFPHP[b] personal contaminations APFP direct dose Core coolant direct dose[c] Solidified core coolant (MSRs, LFRs, and SFRs)

[a] Derived from Refs 5-8,12.

[b] Activation products, fission products, and hot particles. The fission product activity levels depend on fuel integrity or ability of the MSR coolant to retain fission products and other radioactive materials.

[c] Core coolant activation products vary by reactor type and are discussed in the text.

[d] MSRs have no fuel fission product barrier since the fuel and coolant form a eutectic mixture. Refueling occurs while the reactor is operating.

[e] This is a Generation II and III activity that improved maintenance and outage planning. Operating experience and operating policy will determine if it is utilized at planned Generation IV facilities.