Peter Hosemann, Jasmina Vujić, *Material issues for current and advanced nuclear reactor designs*
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Page 10 of 25

MATERIAL ISSUES FOR CURRENT AND ADVANCED NUCLEAR REACTOR DESIGNS

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Abstract: In all engineering applications, design and materials together determine the functionality and reliability of a device. This is particularly important in nuclear systems where the materials are pushed to their limits and phenomena not present anywhere else occur. In nuclear systems a combination of high temperature and pressure, stress, corrosive environment and high radiation environment combined causes significant materials challenges. Majority of commercial LWRs today are licensed for 40 years of operation, but many of them undergo lifetime extension to 60 or possibly 80 years. Materials degradation has always been a significant issue. However, due to the lifetime plant extension, finding materials that could sustain prolonged exposure to these extreme conditions has become a significant problem. In addition to the materials challenges in current LWRs, advanced reactors usually deal with even more difficult issues due to their operational requirements. Unusual heat transport media, such as liquid metals, liquid salts or other types of coolants, lead to a whole new set of material challenges. While corrosion has been the main issue, much higher operating temperatures create additional difficulties. In this paper, we present an overview of materials issues for current and advanced nuclear reactor designs.

Keywords: nuclear systems, extreme environments, liquid metals, liquid salts, corrosion, radiation effects.

1. INTRODUCTION

For efficient economic and social development, a basic necessity is access to affordable energy. In addition to food and water, electricity in particular is a major factor in quality of life. In the last decade, the world population has increased by more than 12% and is expected to rise from today's 7 billion to 9 billion people by 2050. At the same time, with the population growth, the primary energy consumption has increased by 20%, and electricity consumption has increased by 31.5%, while at the same time, nearly 20% of the global population lack access to electricity. [1] It is clear that the world is facing considerable energy and environmental challenges having in mind that the current energy systems, particularly in underdeveloped and developing countries, are highly dependent on fossil fuels, whose combustion accounts for over 80% of global greenhouse gas emissions. This trend will continue in the future, unless more affordable and environmental friendlier source of electricity could be supplied. With all energy production systems there are environmental issues to be considered, risks to be assessed, and challenges to be addressed. It must be emphasized that an ideal energy source that is efficient, cost-effective, environment-friendly, and at the same time risk-free does not exist currently. There are always some necessary trade-offs to be made, in order to ensure optimal use of energy resources, while limiting environmental, safety and health impacts.

It needs to be emphasized that nuclear power is currently the only technology with a secure base-load electricity supply and no greenhouse gas emissions that has the potential to expand to a large scale, and efficiently replace fossil fuel while it has a 50 year history of operation. However, there are issues that need to be addressed in the conventional light water reactor (LWR) and advanced reactor designs, such as the spent fuel management, safety, particularly in the case of natural disasters as shown during the Fukushima accident, and issues with materials degradation under extreme conditions present in nuclear system, such as a combination of high temperature and pressure, stress, and high radiation environments over extended periods of time.

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Current generation LWRs are coming of age and a life extension beyond the original design is going to be needed in order to bridge the time gap until new reactors or more alternative energy sources became widely available. Together with the reactors, operating the materials deployed in the reactor are coming of age. Long service of the materials deployed in this rather harsh environment can lead to degradation of the components ensuring safe and reliable power generation. The combination of radiation, pressure, elevated temperature and corrosive environment over time exceeding now 40 years lead to challenges of how to ensure safe and reliable operation of LWRs for an additional 20−40 years.

Novel reactor designs that have been under the development over many years pose a substantially larger challenge for materials in terms of even higher temperatures, higher radiation doses, higher pressures and novel cooling environments. It needs to be emphasized that *"all designs and engineering solutions are only as good as the materials available to the scientists and engineers"*. Therefore, thorough research in material science and classical metallurgy is needed in order to address these new challenges.

This paper will present the current situation in nuclear power generation in the world, the advanced reactor systems belonging to the Generation IV, and the Small Modular Reactors that are gaining in popularity due to simpler, standardized, and safer modular design by being factory built, requiring smaller initial capital investment, and having shorter construction times. We will outline some important materials issues in current generation reactors deployed as well as the research needed in order to address the issues arising. In addition we will discuss materials limits and requirements for future reactors pointing towards the main issues encountered. While this paper cannot cover all aspects of materials needs for

fission reactors due to the large number of critical components and the complex material science phenomena in a reactor and large number of reactor types it can give an overview of some critical aspects which should be considered in the future.

2. NUCLEAR POWER IN THE WORLD: THE CURRENT SITUATION

In 2013, 30 countries world wide operated 437 nuclear power reactors for electricity generation (with a total net installed capacity of 373,069 MWe), 1 nuclear power reactor was in long term shutdown, and 67 new nuclear power reactors were under construction in 14 countries [2]. The largest number of reactors under construction is in China (29), Russia (11) and India (7). In the meantime, additional countries decided to start with the construction of new power reactors, such as Belarus, United Arab Emirates, Vietnam, Poland and UK [3]. The percentage of electricity generation by nuclear power in the world is 13.8% and in the OECD countries is 21.4% [4].

The United States, with 104 currently operating nuclear power reactors in 31 states (with the total installed net capacity of about 101,000 MWe, and the capacity factor of 92%) that produce about 20% of the total electricity production in the U.S., is the country with the largest number of operating NPPs [5]. There are 5 nuclear power plants (NPPs) under construction in the U.S. at this moment, and the last order for NPP was in 1979. Figure 1 [2] summarizes the nuclear generation in the world by percentage. France has the largest percentage (close to 80%), and in additional 11 countries more than 1/3 of electricity production is nuclear.
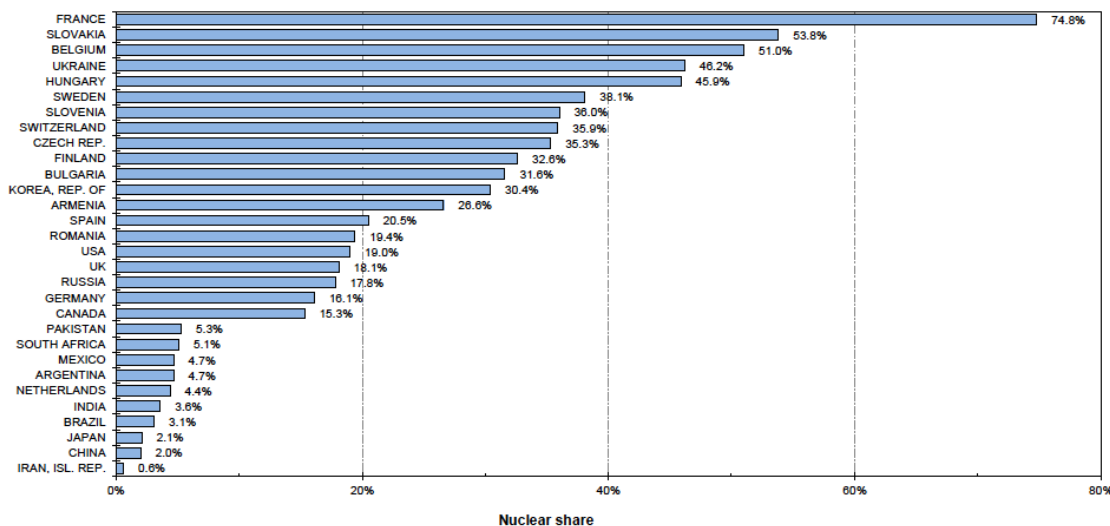

Figure 1. Nuclear Share of Electricity Generation, as of Dec 31, 2012 [2]

Since the beginning in the early 1950s, nuclear power technology has evolved through the following generations of system designs (Fig. 1) [6]: Generation I – mostly early prototypes and first-of-a-kind reactors built between 1950s and 1970s; Generation II – reactors built from 1970s to 1990s, most of which are still in operation today (such as PWR, BWR, CANDU); and Generation III – evolutionary advanced reactors with active safety systems built by the turn of the 20th century (such as General Electric's Advanced BWR and Framatom's EPR). In the U.S. 2 reactors began commercial operation in the 1960s, 50 in the 1970s, 46 in the 1980s and 5 in the 1990s.

2.1. Commercial Reactors of Generation II and III

Majority of today's commercial nuclear power reactors belong to either Generation II or Generation III. There are 274 PWRs (Pressurized Light-Water-Moderated and Cooled Reactors), 81 BWRs (Boiling Light-Water-Moderated and Cooled Reactors), 48 PHWR (Pressurized Heavy-Water-Moderated and Cooled Reactors), 15 GCRs (Gas-Cooled, Graphite-Moderated Reactors), 15 LWGRs (Light-Water-Cooled, Graphite-Moderated Reactors) and 2 FBRs (Fast Breeder Reactors, Sodium-Cooled) [2]. The majority of the new reactors that are under construction are also of PWR (54), BWR (4) or PHWR (5) type [2], which reflects a desire to use proven nuclear reactor technologies.

The newest Westinghouse AP1000 and GE's ESBWR designs that feature passive safety systems belong to the Generation III+. These reactors are yet to be built – the first four AP1000s are under construction in China. For example, the AP1000 reactor design [7] has passive safety features, simplified plant design and modular construction, and short engineering and construction schedule. It was the first and only Generation III+ reactor to receive Design Certification from the U.S. Nuclear Regulatory Commission. Some of the features include: dramatically safer and simpler design, smaller footprint (needs less concrete and steel per MWe), no safety-grade pumps, less maintenance required, much less reliance on operator action to mitigate accidents, independence of off-site AC power to operate reactor safety systems, ultimate heat sink is ambient air. The most important improvement is that the reactor safety functions are achieved without using any safe-ty-related AC power. Instead, the following processes are used: battery powered valve actuation, natural circulation, condensation, evaporation and compressed gases (nitrogen and air) [7]. In the category of advanced LWRs, the European Pressurized Reactor (EPR) which is an evolutionary Generation III reactor designed and built by a French company AREVA [8]. There are two EPRs currently under construction in Finland (Olkiluoto 3) and France (Flamanville 3). Another advanced PWR is a Russian designed VVER-1200, which features passive safety, and which is currently constructed at Lenigrad-2 and the Novovoronezh sites [9].

Generation IV is the next generation of advanced nuclear reactor systems currently under the development, with the goal to improve the performance of current reactors and fuel cycles, in terms of better economical efficiency, enhanced safety, minimization of waste and resistance to proliferation [6].

Having in mind that the majority of current operating commercial power reactors and those reactors that are under construction or planned for early deployment are and will be based on proven LWR technology, it is important to review the basic characteristics and parameters of these reactors as presented in Table 1. Extension of the existing LWRs nuclear power plant operating life up to 60-year license period must insure long-term reliability, productivity, safety and security. After the Fukushima accident, a large opposition appeared towards lifetime extension, stating that it is a better option to start constructing new advanced nuclear reactor systems which don't rely on refurbished aging plants, which might suffer of materials aging and degradation, as well as flaws in safety designs, etc.

Continuing research is necessary in order to develop the scientific basis for understanding and predicting long-term environmental degradation behavior of materials in nuclear power plants. This work will provide data and methods to assess the performance of systems, structures, and components essential to safe and sustained nuclear power plant operations. The results of research will be used to define operational limits and aging mitigation approaches for materials in nuclear power plant systems, structures, and components subject to long-term operating conditions, providing key input to both regulators and industry.
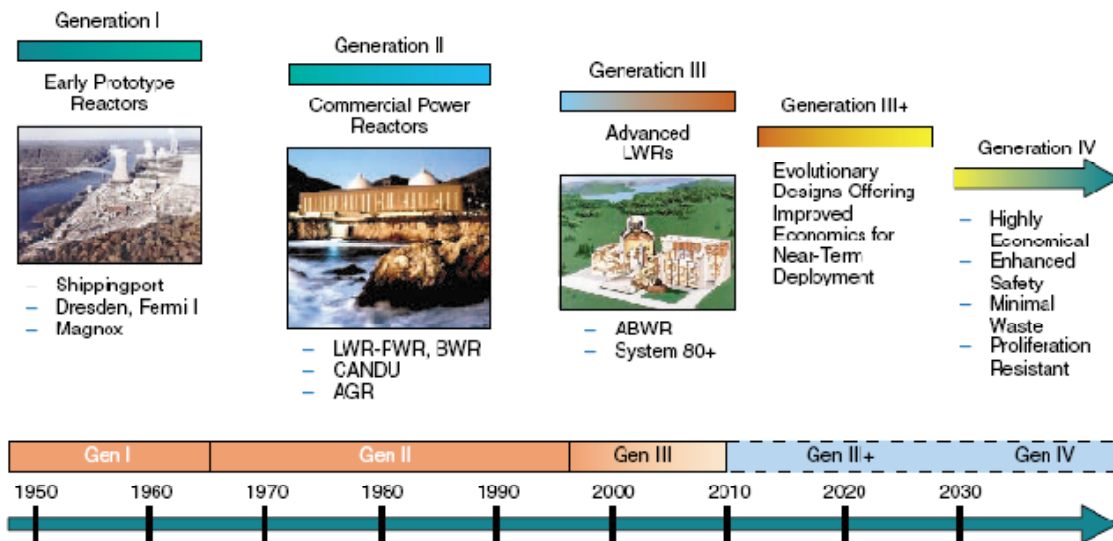
Figure 2. Nuclear Reactor Generations [6]

Table 1. Basic Characteristic for Current and Advanced Water-Cooled Commercial Power Reactors [10]

General Data	PWR	BWR	PHWR CANDU	LWGR RBKM	EPR	ABWR	AP1000	VVER-1200
Thermal Output (MWe)	3411 MWt	1130	600 – 900	4800	1600	1350 – 1600	1117-1154	1170
Pressure (MPa)	15.2	7.1	9.89 – 11.05		15.5	7.07	0.507	16.2
Coolant T-inlet (C)	275	278.3	266	260 – 266	295.9	278.3		298.2
Coolant T-outlet (C)	315	287.2	310	284	327.2	287.8	321	328.9
Fuel Type	UO_2	UO_2	UO_2	UO_2	MOX	UO_2	UO_2	UO_2
Enrichment (w/o)	2-5 w/o	2-5 w/o	Natural	2	Up to 5	4	2-5 w/o	Up to 5
Cladding	Zircaloy-4	Zircaloy 2 - 4	Zircaloy		M5		ZIRLO or M5	Alloy E-110
Coolant	H_2O	H_2O	D_2O	H_2O	H_2O	H_2O	H_2O	H_2O
Moderator	H_2O	H_2O	D_2O	Graph.	H_2O	H_2O	H_2O	H_2O
Burnup (yrs)	40 – 60	40 – 60		60	60 years	60 years	60 years	60 years
MWd/kgHM		56,000	7500		> 70,000	50,000	60,000	60,000

2.2 Advanced Reactors: Generation IV

Achieving the vision of sustainable growth of nuclear energy will also require transition from the current once through fuel cycle to an advanced fuel cycle that recycles nuclear materials. Advanced fuel cycles that are the part of Generation IV reactors have a goal of developing fuel cycle technologies (i.e., fuel, cladding, separations, fuel fabrication, waste forms, and disposal technology) to significantly reduce the disposal of long-lived, highly radiotoxic transuranic isotopes while reclaiming spent fuel's valuable energy.

The emphasis on fast reactors reflects their excellent potential to make significant gains in reducing the volume and radiotoxicity, and increasing the manageability of spent nuclear fuel. Fast reactors also hold the potential for extending the useful energy yield of the world's finite uranium supply many-fold for long-term sustainable nuclear energy. The principal issues in the development of a next-generation fast-spectrum reactor are its economic competitiveness and management of the overall risks to workers and the public from the deployment of a closed fuel cycle. The most promising fast-spectrum Generation IV systems are the Gas-cooled Fast Reactor (GFR), the Lead-cooled Fast Reactor (LFR), and the Sodium-cooled Fast Reactor (SFR).

The Generation IV Roadmap identified six most promising systems, four of which are mentioned above. The additional two are the Supercritical Water-cooled Reactor (SCWR) and the Molten Salt Reactor (MSR). The SCWR employs water above the critical temperature and pressure that affords a considerable increase in thermal efficiency as well as major simplifications and savings in the balance of plant. The MSR employs a circulating liquid fuel mixture that offers considerable flexibility for recycling actinides and may provide a favorable alternative to accelerator-driven systems for actinide destruction.

Below is Table 2 that summarizes main characteristics of Generation IV reactors [6]. The last column lists the challenges of the Generation IV designs, including the needed development of new fuels and other advanced materials, materials compatibility, advanced recycle procedures and safety measure.

Table 2. General characteristics of Generation IV reactor types.[6]

	Neutron Spectra//Coolant/ Fuel	Inlet/Outlet Coolant Temp/ Pressure	Fuel Cycle	Size/Power MWth	Applications	Research and Development
Sodium-cooled Fast Reactor (SFR)	Fast Sodium Metal Alloy or Oxide	550°C outlet 1 atm	Closed	Med to Large 1000-5000	Electricity, Actinide Mgmt. (AM)	Advanced Recycle
Lead-alloy Fast Reactor (LFR)	Fast Pb-Bi Metal alloy/ Nitride	550 − 800°C outlet 1 atm	Closed	Small to Large 125−3600	Electricity, Hydrogen Production	Fuels, Materials compatibility
Gas-Cooled Fast Reactor (GFR)	Fast Helium UPuC/SiC (70/30%)	490°C inlet 850°C outlet 90 bar	Closed	Medium 600	Electricity, Hydrogen, AM	Fuels, Materials, Safety
Very High Temp. Gas Reactor (VHTR)	Thermal Helium ZrC coated particles	640°C inlet 1000°C outlet high	Open	Medium 600	Electricity, Hydrogen, Process Heat	Fuels, Materials, H_2 production
Supercritical Water Reactor (SCWR)	Thermal, Fast Water	280°C inlet 510 − 550°C outlet 25 MPa	Open, Closed	Large 1700 MWe	Electricity	Materials, Safety
Molten Salt Reactor (MSR)	Thermal Fluoride salts UF	565°C inlet 700 − 850°C outlet	Closed	Large 1000 MWe	Electricity, Hydrogen, AM	Fuel, Fuel treatment, Materials, Safety and Reliability

Table 3 illustrates possible choices for fuel, cladding, and structural materials, as well as the operating temperature ranges for Generation IV reactor designs [6]. Choice for fuel includes oxide (SFR, SCWR, VHTR), metal (SFR, LFR, SCWR-fast), nitride (GFR, LFR), carbide (GFR) or fluoride – liquid fuel (MSR). Fuel will be exposed to higher temperatures, and longer burnup. Most of the designs will operate either in fast or epithermal neutron spectra, leading to the higher radiation damage. Choice of cladding and coolant also leads to the question of physical and chemical compatibility.

Very important are long-term irradiation testing of fuels and other reactor materials and demonstration of their safety.

2.3. Small Modular Reactor (SMR) Systems

Small Modular Reactors (SMRs) came into the focus over the last several years, primarily due to large initial capital investment requirements for large nuclear power plants. In the recently published paper on SMRs [11], it was pointed out that SMRs could offer simpler, standardized, and safer modular design by being factory built, requiring smaller initial capital investment, and having shorter construction times. The SMRs could be small enough to be transportable, could be used in isolated locations without advanced infrastructure and without power grid, or could be clustered in a single site to provide a multi-module large capacity power plant.

Table 3. Operating temperature of various Gen IV reactors and potential material choices [6]

System	Spectrum, T_{outlet} [°C]	Fuel	Cladding	In-Core	Out of Core
GFR	Fast; 850	MC/SiC	Ceramic	Refractory metals, Ceramics, OD, F/M	Prim Circ.: Ni based e.g. 32Ni-25Cr-20Fe.5W-0.05C Ni-23Cr-18W-0.2CF-M Thermal barrier turbine Ni-based; ODS
LFR	Fast; 550 and Fast 800	MN	High Si or Al F-M, Ceramic or refractory		High Si or Al austenitics, Ceramic or refractory
MSR	Thermal 700 −800	Salt	Not applicable	Cera., refract., High Mo Ni based alloys (e. g. INOR-8, Hastelloy–N) graphite	High Mo Ni based alloys e. g. INOR-8, Hastelloy –N)
SFR (Metal	Fast, 520	U-Pu-Zr	F/M (HT-9, T91, ODS)	F-M ducts 316l grid plate	Ferritics, Austenitic
SFR (MOX)	Fast, 550	UO_2	ODS	F/M duct, 316l grid plat3	Ferritics, Austenitic
SCWR thermal	Thermal, 550	UO_2	T91, HT,9 Fe-35Ni-25Cr-0.3Ti Inbc. 800, ODS, INc 690, 625&718	Same as cladding	F/M
SCWR fast	Fast 550	MOX, Dispersion	T91, HT,9 Fe-35Ni-25Cr-0.3Ti Inc-800, ODS, Inc-690, 625	Same as cladding options	F/M
VHTR	Thermal 1000	Triso, UOC in graphite compacts ZrC coating	ZrC coating and graphite	Graphite PyC, SiC, ZrC Vessel F/M	Ni based superalloys 32Ni-25Cr-20Fe-12.5W, 0.05C, Ni 23-Cr-18W-0.2C F/M; thermal barrier coatings on Ni based alloys or ODS

Abbreviation: F/M Ferritic martensitic steels typically T91 or HT-9; OSD Oxide dispersion strengthened alloy; MN (U,Pu), NMC: (U,Pu) C, MOX (U, PU)O_2

There are technical and institutional challenges to be addressed regarding broader deployment of SMRs: testing and validation of technological innovations in components, systems and engineering (especially testing and fabrication of fuel), fear of first-of-kind reactor designs, economy-of-scale, perceived risk factors for nuclear power plants, and regulatory and licensing issues. Other issues to be addressed are the cost of reactor decommissioning and spent nuclear fuel management. [11].

Today the U.S. is making major investments to develop small modular reactor (SMR) technologies that will feature major simplifications in their

design and operation, will have passive safety systems, and will use very high degrees of factory prefabrication of modular components. Multiple U. S. vendors have been competing in this process for years. Other countries have also developed various SMR designs, as shown in Table 4 [11].

Figure 3 shows several SMRs developed in the USA. In 2013, the mPower SMR was selected by the U. S. Department of Energy (DOE) as the sole winner, and the contract was signed between the B&W company and DOE regarding funding for the development and licensing of B&W mPower technology.

Table 4. Current SMR Designs [11]

Type	US	Russia	Japan	France	India	S. Korea	Argentina	China	Italy
Integral PWR	IRIS, NuScale, mPower		IMR	SCOR		SMART	CAREM		
Marine Derivative PWR		ABV, KLT-40S, VBER-300, 150		NP-300					
BWR/PHWR		VKR-MT	CCR		AHWR				MARS
Gas-Cooled	GT-MHR	BGR-300	GT-HTG-300					HTR-PM	
Lead/Pb-Bi Cooled	ENHS STAR/SST AR	BREST SVBR-75/100	LSPR						
Sodium-Cooled	PRISM ARC-100	BN-GT-300	4S RAPID			KALIMER			
Non-conventional	AHTR Hyperion TWR	MARS	MSR-FUJI		CHTR				

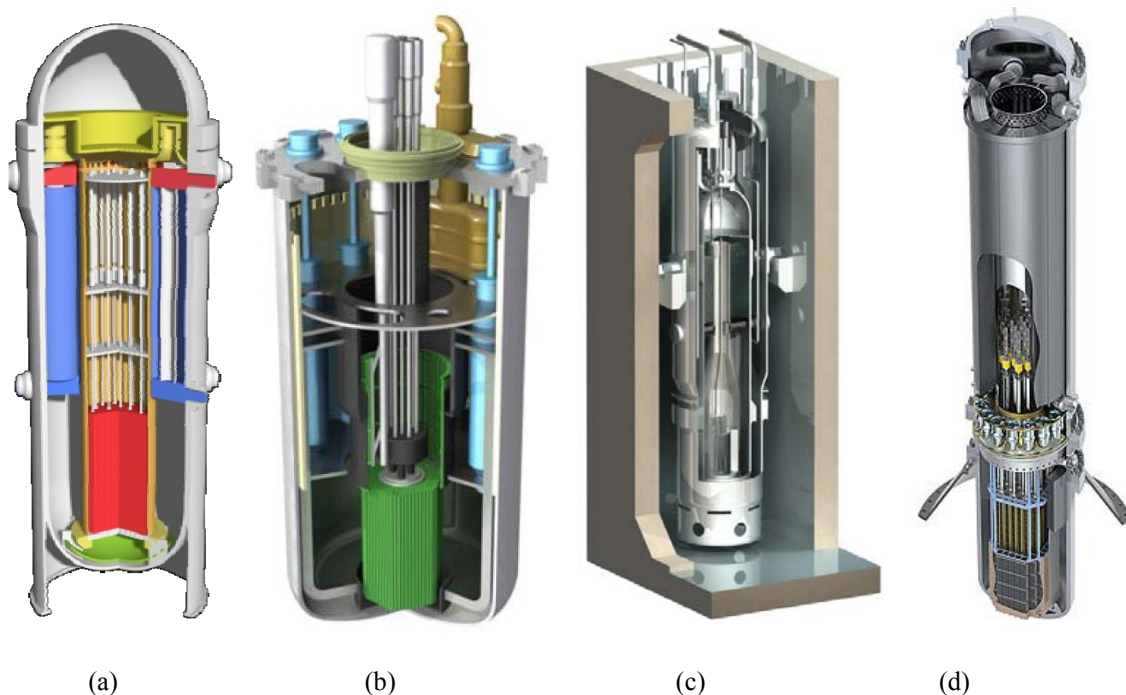(a)   (b)   (c)   (d)

Figure 3. Modular small reactor designs in USA: (a) IRIS [12], (b) PRISM [13], (c) NuScale [14], and (d) mPower [15]

The ideal SMR concept must satisfy requirements of sustainability, passive safety, proliferation resistance, simplicity of construction and operation, and affordability. SMRs could be broadly used by smaller utilities, by smaller countries with financial or infrastructural constrains, in isolated regions or for distributed power needs, and for various other non-electrical application (process heat, desalination, oil recovery for tar sends and oil shale, district heating). SMRs offer increased safety by eliminating

most of accident initiators (for example, large pipes in primary circuit), by improving decay heat removal and including more efficient passive heat removal from reactor vessel, more in-factory fabrications, transportability and site selection flexibility, smaller plant footprint and use of seismic isolators for increased seismic safety, as well as reduced investment risk. Still, there are technical and institutional challenges to be addressed with further R&D: testing and validation of technological innovations in components, systems and engineering (especially testing and fabrication of fuel), fear of first-of-kind reactor designs, economy-of-scale, perceived risk factors for nuclear power plants, and regulatory and licensing issues. Other issues to be addressed are the cost of reactor decommissioning and spent nuclear fuel management.

3. MATERIAL ISSUES AND CHALLENGES

In the following we are introducing the issues of materials degradation under radiation and discussing the materials issues and challenges in LWR's and advanced reactors from Section 2.

3.1. Materials issues in LWR applications

The most critical components in a reactor are the fuel (cladding and actual fuel), core internals, reactor pressure vessel and the building/concrete structure. Each component has its own specific set of issues associated with the environment the materials are exposed to. While the fuel assemblies are frequently replaced, the vessel and the building itself are not. The fuel in LWR is in general UO_2 a well proven and established fuel form. While the [235]U provides the actual fuel for the fission reaction and is responsible for the heat production and therefore power, it also leads to breeding of a large number of fission products in the entire fuel rod. These fission products contribute to the fuels degradation in combination with the strong thermal gradient within the fuel pellet itself due to the limited thermal conductivity of the oxide fuel form. The fuel swelling and decrease of thermal conductivity is due to the buildup of fission gases in the fuel. Other fuel forms (non-oxides) have been considered and discussed for a long time, but have not been widely deployed due to immediate issues such as manufacturability and costs (nitride or carbide fuels) as well as melting points (metal fuel). In addition to the fuel degrada-

tion itself, the fuel is in a direct contact with the cladding material. This can lead to the fuel–clad chemical interactions (FCCI), causing the disintegration of the fuel cladding from the inside out, and failure of the integrity of fuel pin [16]. New fuel forms, enhancing thermal conductivity, reducing swelling and FCCI, are under consideration but have not been deployed in commercial reactors yet. Novel ideas, like liquid metal bound fuel-cladding to enhance heat transfer, have been proposed [17] but a significant amount of research need to be conducted in order to make these materials concepts feasible.

The fuel cladding is probably the most critical component in the reactor. It is supposed to hold the fuel pellets in the pin and prevent leakage of fission products into the reactor under all conditions. In commercial reactors Zr based alloys are deployed (Zircalloy -4 or Zircalloy -2) because of their low neutron absorption cross section, corrosion resistance and strength under normal operation condition. However, in an accident scenario this material has the unfortunate property to react with water at elevated temperatures according to the reaction:

$$Zr + H_2O \rightarrow ZrO_2 + 2H_2 \qquad (1)$$
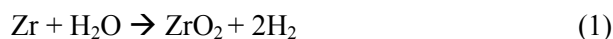
This leads to the rapid production of hydrogen at temperatures above 1000°C and releasing $6.057*10^5$ J/mol. Typically, a parabolic oxidation rate can be used to calculate the loss of Zirconium (W) due to oxidation with

$$W = (K(T)*t)0.5 \qquad (2)$$

and

$$K(T) = A \exp (B/RT) \qquad (3)$$

where A is a constant, B is the activation energy, t is the time, T is the temperature in Kelvin and R is the gas constant [18, 19]. Equations 2 and 3 allow estimating the amount of ZrO_2 developed during exposure of Zr to steam at various temperatures and times and are used to determine the amount of hydrogen buildup as shown in Figure 4.

It is the above mentioned chemical reaction and the subsequent generation of Hydrogen which leads to the explosions observed in Fukushima after the Loss of Coolant Accident (LOCA). Another unfortunate property of Zirconium alloys is that the alloys loss of strength at high temperature leads to ballooning [20] of the cladding and therefore reduction of the space between the cladding rods which prevents effective cooling and even enhancing the effects.
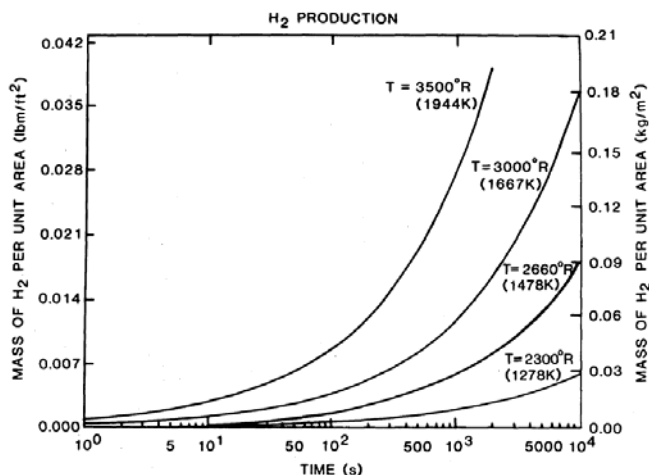
Figure 4. Hydrogen produced per unit area at different times and temperatures on a Zr cladding [19]

Due to the unfavorable properties of Zr and its alloys mentioned above the nuclear materials community is constantly searching for new materials deployable in LWR's as fuel cladding. Currently several alternative fuel claddings are proposed including but not limited to SiC/SiC composites, refractory based cladding and Fe-Cr-Al based claddings. While all proposed claddings mitigate the shortcomings of Zr based materials, new sets of challenges are introduced. SiC/is a ceramic materials and has an inherent low fracture toughness [21] which can be improved with the composite structure SiC/SiC (fiber matrix). In addition, manufacturing and costs are a challenge of these ceramic based materials [22]. Mo-based claddings are currently considered, but production of volatile MoO_3 leads to rapid oxidation of Mo at elevated temperatures and therefore additional coating of the cladding is required. Fe-Cr-Al steels have excellent corrosion properties but the comparable low melting point of steels in general leads to significant strength issues (creep). The similar unfavorable neutron cross section compared to Mo raises mechanical stability questions at accident scenario temperatures of 1000°C and above due to the high neutron cross section which requires a reduced cladding thickness. As always in materials science there is no "silver bullet" only decent materials for specific applications are available with a range of limitations.

The reactor pressure vessel (RPV) is maybe the most critical single component of a reactor and therefore close monitoring of the component and its degradation is conducted. Since it is uneconomically and often not possible due to building constraints and the vessel size to replace the vessel itself if failures or enhanced degradation occurs it is one of the lifetime limiting factors. Maybe the biggest single concern of RPV is its embrittlement over time [23]. Since reactors are in service for more than 40 years, one has to consider the metallurgical limitations of manufacturing from 40 years ago. Early RPV's were found to contain minor amounts of Cu and other impurities which, under normal circumstances are not an issue, but do cause significant embrittlement over the RPV's life due to the long time (> 40 years) exposure at elevated temperature (300°C) and radiation dose. In particular, the work performed by Odette et al. [24] shows that this condition leads to the formation of the so-called late blooming phases which significantly increase embrittlement to a level unacceptable for reliable operation. Figure 5 shows the shift in ductile to brittle transition temperature as a function of neutron dose on the RPV and how various impurities (Cu, Ni) affect it.

The main concern of embrittlement and a high DBTT is not during normal operation but due to emergency cooling in the event of a Loss of Coolant Accident (LOCA). Emergency cooling can cause thermal shock of the component and if the material has a relative high ductile to brittle transition temperature (DBTT) and lower upper and lower shelf energy rapid crack propagation will occur. Due to the above consideration, close monitoring of the RPV is conducted involving a large number of mechanical tests on surveillance samples from the core. Also, non-destructive monitoring of cracks and other degradation issues is routinely conducted. If embrittlement is estimated to limit the life of the reactor based on Nuclear Regulatory Commission (NRC) guidelines [23] so the reactor intended maximum operation time cannot be fully utilized, the utilities have several options. First, a complete heat treatment of the RPV can be conducted in order to mitigate some of the embrittlement issues as illustrated

in Figure 5d; second, the dose on the RPV can be reduced by rearranging the fuel such that the RPV experiences less dose for the remainder of its lifetime which usually reduces the fuel utilization; and

third, the reactor and therefore the plant will need to be shut down and the construction of a new unit need to be planned.
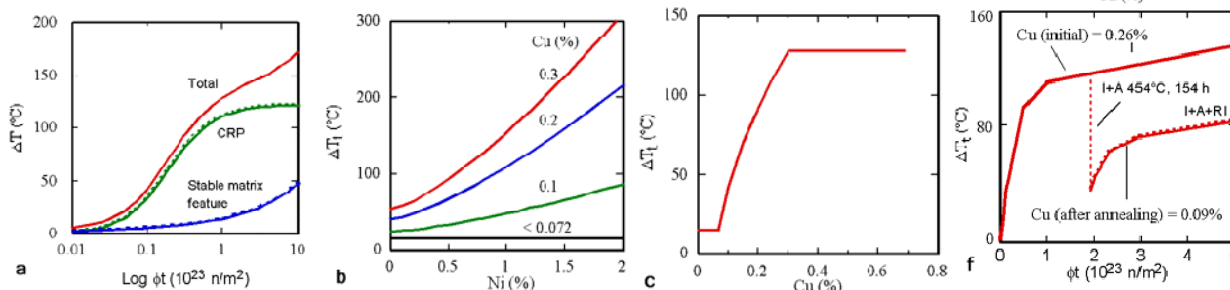

Figure 5. Ductile to brittle transition temperature change due to the formation of copper rich precipitates (CRP) and stable matrix features as a function of dose (a). Effect of Cu and Ni impurities on DBTT (b). Effect of shift in DBTT induced by Cu (c) and improvement of DBTT due to a heat treatment (d). Figure is reproduced after Odette et al [25]

The issue of late blooming phases (minor impurity precipitates after long time aging) is also mitigated in more modern RPVs due to the fact that lower amounts of impurities are present in the RPV materials deployed. However, also for modern RPV materials long term materials testing and close monitoring of the materials degradation are needed. It has to be stated here that RPVs are among the largest single components manufactured and deployed and operating the longest under such extreme condition of temperature, time and dose which all contribute to materials aging and difficulties with manufacturing.

The containment structure itself in an NPP experiences aging issues and is one of the few items which usually cannot be replaced. If the containment itself is compromised, a safe operation cannot be guaranteed in respect to potential accident scenarios. Therefore, close monitoring of the containment and the building structure must be conducted. It is known that concrete ages just like steel or other materials over time [26]. Three main mechanisms of concrete degradations are reported in [26], the well-known alkali-silica reaction causing the formation of a alkali-silica gel and further cracking of the concrete; corrosion of the steel reinforcement; and sulfate attack causing the loss of concrete strength. Radiation can also significantly damage concrete. In addition to neutron damage also gamma radiation can cause radiolysis in the water contained in the concrete leading to creep and shrinkage. [27].

Interestingly, despite the importance of the concrete for nuclear power applications, comparable little research is conducted if compared to structural core internal components.

3.2. Materials issues in next generation nuclear power (NGNP)

Due to the large number of potential reactor designs proposed it is impossible to discuss materials challenges for every single one of them in this paper. However, the challenges can be summarized into three main categories a) environmental (corrosive), b) high temperature and c) higher dose requirements for most of these designs.

As in all of material science and associated engineering, there is not a single factor responsible for difficulties, but rather the combination of all requirements due to comprehensive nature of an engineering challenge. Ideally, one has a material which can address all three main concerns listed above, but this is usually not feasible.

The need for higher temperatures is usually driven by the need of higher efficiency for the power generation or generation of process heat. High temperature usually calls for high creep resistance. While regular F/M steels are usable to temperatures of 600 C, higher temperatures ask for more advanced alloys such as oxide dispersion strengthen materials usable up to 800 C. Beyond this temperature only Ni based superalloys, refractories or ceramics are an option. Figure 6, reproduced after S. J. Zinkle [28], illustrates the operating temperatures for candidate fusion materials which also applies to fission materials.

While Ni based superalloys are a good choice based on temperature considerations, the high Ni content however tends to increase the materials radioactivity after exposure to neutrons and therefore makes service of the components more difficult while more He is generated in the alloy leading to severe embrittlement issues. The fcc nature of Ni alloys also leads

to enhanced void swelling of the components in question if no further defect sinks are provided like if additional oversized solute atoms are provided [30] or large number of interfaces due to precipitates. Currently, materials concepts such as SiC/SiC composites or ODS alloys are often discussed for a large number

of NGNP designed. Both options address the high temperature questions but are expensive to manufacture and difficult to implement. Simple tasks like welding can become a major issue with those rather unconventional material solutions.

a)   *b)*
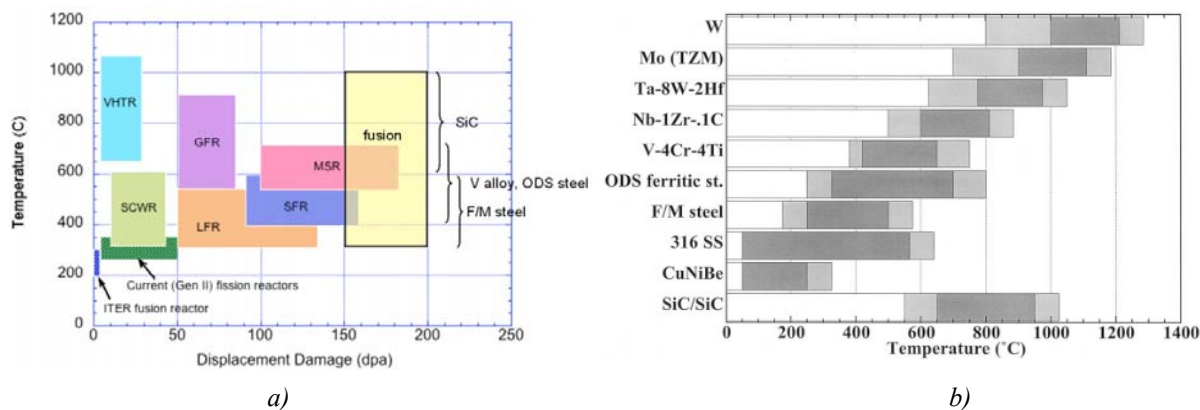
Figure 6. Temperature requirements for various reactor designed Gas fast reactor (GFR), Very High Temperature Reactor (VHTR), Lead Fast Reactor (LFR), Sodium Fast Reactor (SFR), Molten Salt Reactor (MDR), Supercritical Water Reactor (SCWR), and fusion reactors a) [28]. Usable temperature range on various potential structural materials considering dose and creep performance b) [29]

Unusual cooling environments, such as liquid salts or liquid metals and even gases, in combination with the large number of various materials deployed like metals and graphite together in the same system can lead to a large number of challenges. Liquid salts as well as liquid lead bismuth eutectic (LBE) can be rather corrosive if the chemistry is not controlled well and particular oxygen levels are not kept at a set target value. While liquid salts need a low oxygen level due to the fact that the corrosion mitigation strategy is immunity against metal ion dissolution [31 and 32. In general impurity control is very important for salt corrosion issues and grain boundary attack of steels is common. Similar is true for Sodium cooled reactors. The oxygen levels need to be kept to a minimum [33]

Lead bismuth eutectic (LBE) calls for oxygen concentrations between a low and a high set point going for passivation as the corrosion mitigation strategy [34] requiring precise oxygen control [35]. While several LBE cooled reactor systems have been realized, detailed studies of the oxidation and corrosion mechanism are still conducted using modern Atomic Force Microscopy techniques [36], nanoindentation [37] Transmission Electron Microscopy [38] leading to comprehensive models [39]. In fact, these studies showed that the oxidation process of steels in a liquid metal environment is far more complicated than the oxidation process in gaseous environments due to the combination of oxidation

and leaching growing leached out nano porous passive films with a spinodal decomposition type chemistry on a nano scale. Figure 7 shows TEM [38] and atom probe tomography (APT) [40] next to each other showing a local Ni segregation in a passive film grown on the austenitic stainless steel D9 in LBE over the course of 3000 h at 550 °C.

Again, the detailed studies like the ones mentioned above, provide a deep insight into the materials degradation mechanism and therefore allow us to make predictions of long term behavior in reactor applications.

One of the most prominent representatives of the liquid metal cooled reactors is the sodium cooled reactor. This reactor concept has been realized multiple times in the past. Bor 60, BN 600 are long term operating sodium fast reactors operating reliably since decades. Also in China and India sodium technology is deployed while the US shut down all relevant programs on this technology. While the deployment of sodium as a coolant has the inherent dangerous feature of potential sodium fires, corrosion is less of an issue in pure sodium without any impurities and corrosion rates are low. The phenomenon of liquid metal embrittlement (a structural degradation of a material due to exposure in a liquid metal environment) is not as dominant as it is for lead bismuth reactors [41].
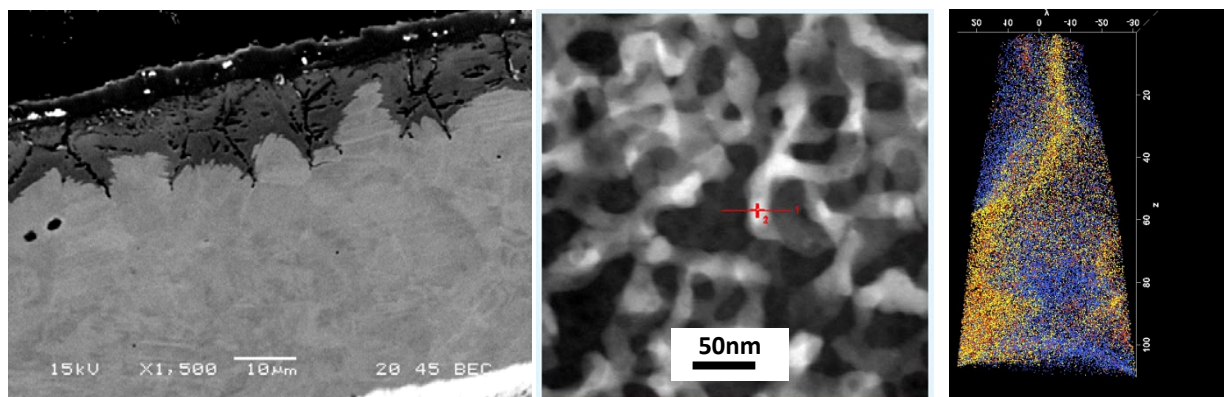
Figure 7. Typical double layer structure formed on D9 stainless steel in LBE at 550C over 3000h (left). High resolution Scanning Transmission Electron Microscopy image showing spinodal decomposition of the inner oxide layer (middle) [38] and Atom Probe Tomography image (right) of the inner oxide layer with Yellow – Oxygen, Red – Chromium and Blue – Nickel [40]

High temperature gas reactors usually call for He as a heat transport fluid. This can cause corrosion issues due to impurities in the gas, especially oxygen and moisture, which can cause the formation of CO CO_2 H_2O, H_2 and CH_2 [42 and 43] and therefore some solid/gas interactions and high temperature corrosion. In lower temperature systems the usage of certain non-ideal materials in environments is often allowed, simply because potential degradation mechanisms are too slow to take place. At these types of reactors however, kinetics is not the limiting factor to degradation mechanisms and the governing phenomena are thermodynamic based. One aspect often overlooked in various reactor designs is the question of solid-solid materials interaction. For example, graphite components utilized in direct contact with stainless steel can lead to rapid degradation and property changes of the stainless steel component due to diffusion which is rather sensitive to carbon content. Again, this article cannot focus on the detailed aspects of each one of the issues but can give a brief introduction to a number of issues and raise awareness of these issues.

High dose exposures of core internal components are maybe the most discussed issue for advanced reactors. One of the main difficulties with deploying advanced reactors is that there is simply no high dose data available on modern and advanced materials considered. Nuclear materials scientists deploy the unit of "displacement per atom" (dpa) to evaluate a material dose. A dpa attempts to cross compare different damaging spectra even bridging the gap between neutron irradiation and ion beam radian. However, it is a difficult unit to use and solid materials science background is needed to appreciate the full breath of this unit in combination with other factors such as temperature. While dpa serves as a

dose measure and assesses the instantaneous damage a material experiences under radiation condition, it does not say anything about the actual degradation damage since it is in general the remaining defects present in longer timeframes which cause the degradation of mechanical properties. Therefore, parameters like dose, dose rate, temperature, stress levels, microstructural features and others are all part of the equation assessing a material's sustainability to radiation damage. However, in general it can be stated that the main concerns induced by radiation damage are: void swelling (volume increase due to void growth), embrittlement, strength increase, enhanced creep at medium temperature ranges, enhanced corrosion under radiation due to enhanced diffusion and the production of aggressive radicals. In addition, changes in composition must be considered, especially in fuels or fusion materials where Tungsten breeds Re and as the result of long lasting fusion irradiation a tungsten Rhenium alloy is formed.

Some reactor designs call for dose levels well above a 100 dpa. Unfortunately, only very few materials such as HT-9 [44] have been irradiated to this level since irradiation campaigns achieving this can take decades to plan and conduct. In addition, most funding agencies today have not the foresight to invest heavily in long lasting programs addressing long term issues leading to a lack of data. In general it is to say that nuclear materials are an area of research requiring solid metallurgical work and continuous commitments. There simply is no real substitute for an engineering scale creep or irradiation tests which is a time consuming and costly endeavor.

However, to mitigate some of the issues stated above as well as the shortcomings in long term commitment the nuclear materials community deploys more ion beam irradiation work to obtain more

basic scientific data and understanding in combination with modeling efforts to potentially estimate materials behavior as a function of radiation exposure in a reactor environment. Again, this approach is science-based and all data obtained from these efforts need to be validated with true engineering scale measurements. Ion beam irradiations have been deployed since decades to address basic scientific questions and accelerated damaging of materials. In order to properly mimic reactor irradiations using ion beams, it has been realized that multi beam experiments need to be conducted after Tanaka et all [45] found that there seem to be synergistic effects especially in regards to void swelling between dpa dose, He and H, the two main transmutation products in metals. The Jannus facility in France, Duett in Japan and the facility currently under construction at the University of Michigan are facilities that allow studying this phenomenon and conducting the research.

While irradiating materials is clearly a bottleneck in materials studies under radiation, it is the post irradiation examination which is the most difficult one, especially with declining hot cell facilities around the world. Very few facilities have the capability and expertise to handle and properly examine irradiate materials from reactors. With the deployment of new materials characterization and micro manufacturing techniques one can overcome some of these limitations. It has been shown that if specimens are manufactured small enough that no radioactivity can be detected on them, they can be handled outside of hot areas putting the emphasize towards materials processing rather than materials examination in hot areas [46 and 47]. Hot focused ion beam (FIB) machining is maybe the single most pressing bottle neck today to examine irradiated materials. Facilities like the PSI in Switzerland, or CASE in Idaho and UC Berkeley realized this need and established user facilities for people to process their hot samples.

4. SUMMARY AND CONCLUSIONS

Nuclear engineering, including nuclear materials research, has been an established research and technology field for many decades, and therefore represents a mature field of research with a large amount of data and experience. One might think that the amount of new research conducted is limited due to the maturity of the field while quite the opposite is true. As conventional power plants age, the question of materials degradation becomes more and more important and solid material science needs to

be conducted in order to ensure the long-term plant safety. Also, the Fukushima event triggered a large amount of questions regarding the LWR plant design and materials choices, leading to new research directions with the aim of improving efficiency and safety of already deployed conventional power plants.

However, material issues also arise in the advanced designs for the future generations of nuclear power plants, where they are presenting the most serious limitations for the realization of new reactor designs, due to requirements for much higher operating temperatures, larger burnups, higher irradiation doses, incompatible coolants and various safety issues.

This paper gives a short overview of the current situation in the world regarding nuclear power. Most of the current commercial reactors belonging to Generation II and III were built in the 1960s and 1970s. They are exceeding 40 to 60 years of operation lifetime, with increasing challenges of how to ensure safe and reliable operation for an additional 20-40 years. The paper also addresses the main features of advanced reactor systems belonging to the Generation IV, and the Small Modular Reactors with simpler, standardized, and safer modular designs. The most important features that make LWR SMRs commercially attractive--rapid construction, low capital placed at risk, and less expensive and difficult project financing-also lower the barriers for building Gen IV demonstration reactors.

The second part of the paper deals with the materials issues in nuclear reactors (current and future) and a list of challenges that need to be overcome. As stated above, it is the materials available to the engineers and scientists that are the limiting factors in all new plant designs, and engineers need to consider the limitations of the materials available in order to obtain credibility for a new reactor design. Therefore, large sustainable investments need to be made in the materials aspects of nuclear engineering today.

It should be emphasized that the goal of this paper is not to analyze the detailed aspects of each of the materials issues, but rather to give a brief introduction to a number of issues and to raise awareness of those issues and the need for further research. However, the paper did cover in more details three of the most critical components in a rector: the fuel (cladding and actual fuel) core internals, reactor pressure vessel and the building/concrete structure. A specific set of issues were discussed which were associated with the environment the materials are exposed to.

For future reactor designs, the materials challenges could be summarized into three main catego-

ries: a) environmental (corrosive), b) high temperature and c) higher dose requirements for most of these designs. The paper discussed issues with superalloys, refractories or ceramics for very high temperature environments, the corrosion issues with Lead Bismuth Eutectic coolants, and the embrittlement, swelling, enhanced creep and corrosion for high-dose material irradiation. The need for experimental verification and validation of theoretical results was emphasized, as well as the investment in new experimental facilities that could speed up this crucial materials research.

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ПРОБЛЕМИ СА МАТЕРИЈАЛИМА ЗА ПОСТОЈЕЋЕ И БУДУЋЕ ДИЗАЈНЕ НУКЛЕАРНИХ РЕАКТОРА

Сажетак: У свим инжењерским дисциплинама, функционалност и поузданост једног уређаја највише зависи од дизајна и коришћених материјала. То посебно долази до изражаја код нуклеарних система, гдје се материјали доводе до граница издржљивости, и гдје се појављују ефекти на које се не наилази у другим примјенама. У нуклеарним системима материјали су истовремено изложени високим температурама и притисцима, стресу, и високим радијационим пољима. Већина данашњих комерцијалних лаководних реактора је лиценцирана за 40 година рада, али им се све чешће лиценца продужава на 60, а могуће и 80 година. Деградација материјала у нуклеарним реакторима је одувијек представљала велики проблем. У случају продужавања радног вијека, проналажење материјала који ће моћи да опстану под тако екстремним условима и то на дуже вријеме може да представља непремостиви проблем. Поред проблема са издржљивошћу материјала у данашњим нуклеарним електранама, на још веће изазове се наилази при дизајнирању будућих реактора, јер се пред њих постављају још тежи изазови. На примјер, као хладиоци будућих реактора четврте генерације појављују се течни метали и растопљене соли, што доводи до потпуно нових захтјева. Мада је корозија материјала увијек представљала проблем, много више температуре рада будућих реактора стварају додатне тешкоће. У овом раду ћемо дати преглед проблема са материјалима који се користе у дизајнима постојећих као и будућих нуклеарних реактора.

Кључне ријечи: нуклеарни системи, екстремни услови рада, течни метали, растопљене соли, корозија, радијациони ефекти.

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