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A review of structural material requirements and choices for nuclear power plant

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A nuclear power plant is a highly complex installation. Its design is a response to many drivers, including neutronic efficiency, thermal efficiency, economic efficiency, radiation safety, structural integrity, ease of monitoring and maintenance. The correct selection of structural materials has been key in achieving long term structural integrity, as different plant designs and operating conditions impose different combinations of requirements on the materials. This paper describes the reasoning behind structural materials choices and the changing perspectives on the interplay with other design imperatives in historic, current and future plant designs. It also describes the campaigns of materials development put in place to meet novel materials challenges specific to nuclear plant.

1 Introduction

Nuclear power plants (NPPs) constitute complex arrangements of connected components. Identifying materials capable of withstanding the operational lifetimes of exposure to the particular stress ranges, temperature ranges, radiation doses and chemical environments for each individual component is not trivial. Balancing the imperatives for different components, so that suitable and compatible materials may be found for each component in a NPP, is even less so. Materials selection is, thus, a significant aspect of initial reactor design. Changing any given reactor property (e.g., power output, operating temperature, operating life, reactor size) will affect the exposure histories of multiple components. As a result, knowledge of the properties of the selected materials must cover all plausibly relevant conditions. For the assurance of safety in NPP operation and development, this baseline data must be supported by component monitoring during service and material modelling to allow interpolation between and extrapolation beyond intermittent measurements.

Extensive experimental and modelling campaigns have been carried out around the world in support of component reliability in current NPP. These have involved: obtaining

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Abbreviations: AGR, Advanced gas-cooled reactor; AISI, American Iron and Steel Institute; ASME, American Society of Mechanical Engineers; ASTM, American Society for Testing and Materials; FR, Fast reactor; HTGR, High Temperature Gas-Cooled Reactor; LAS, Low alloy steel; LFR, Lead or lead-bismuth cooled Fast reactor; LWR, Light water reactor; MSR, Molten salt reactor; MTR, Materials Test Reactor; MWh, Megawatt hours; NPP, Nuclear Power Plant; ODS, Oxide dispersion strengthened; PWR, Pressurised water reactor; RPV, Reactor pressure vessel; SCC, Stress-corrosion cracking; SCWR, Supercritical Water cooled reactor; SFR, Sodium-cooled Fast Reactor; VHTR, Very high temperature gas-cooled reactor.

engineering data on plausible candidate materials, to show whether the properties required for a given component can be achieved consistently at start-of-life; down-selection among candidate materials; identifying the rate at which material properties degrade under operational conditions; further optimisation of compositions and thermo-mechanical treatments in the light of operational experience. Campaigns started in the 1940s, preceding the construction of the first power reactors in the late 1950s and early 1960s. Work has since continued through 60 years of development of NPP. This has required significant physical infrastructure in terms of inactive test facilities, active handling facilities and Materials Test Reactors (MTRs). The intellectual infrastructure involved, in terms of trained, qualified and experienced personnel, has also been wide-ranging in topic areas and extensive in the size of the workforce.

Plant operators face a continuous need to modify operational parameters e.g., to enhance safety, to extend plant life, to meet more fluctuating electricity demands. The timescales on which they require supporting material information is much shorter than that involved in initial plant construction, while the facilities available for materials research have diminished. More than 800 MTRs were built to facilitate materials and fuels characterisation in support of Generation II–Generation III + NPP, mostly in the 1960s and 1970s (Jawerth and Mattar, 2020). Fewer than a third are still in operation, and the great majority of MTRs today are more than 30 years old and in need of renewal. Timescales supporting Generation IV plant design and construction may be more relaxed, but the demands of Generation IV NPP are wider ranging. There is, thus, international interest in optimising efforts and reducing the time to qualify new materials, both to enhance current reactor operation and to allow new designs of reactor to be constructed.

This paper reviews some of the work that has been involved in materials research underpinning NPP operation. The review is not meant to be exhaustive. Indeed, the number of components and operating conditions involved in a single plant is enormous, and each component is susceptible to several degradation mechanisms. The detail in the review is intended to be sufficient to illustrate the reasoning behind different historic choices for structural materials, and the changing perspectives on the interplay with other design requirements in historic, current, and future plant designs. The aim is to provide insights which may be useful in the direction of structural materials development campaigns for future (Generation IV) NPP. More specific information on the operational conditions and materials used in many components of different reactor designs can be found in reviews such as (Was et al., 2007; International Atomic Energy Agency, 2012; Cattant, 2014; Shirzadi and Jackson, 2014; Motta et al., 2015; International Atomic Energy Agency, 2019; Odette and Zinkle, 2019; Konings and Stoller, 2020 Ahn, 2021).

2 Materials choices made in historic plant

2.1 Generation I—Magnox

The first nuclear power station to generate electrical power on an industrial scale was of the Magnox design. Magnox reactors were

dual-use, producing plutonium for military purposes as well as electricity for civilian use. The core design was, thus, developed from that used in the military-use Windscale piles. The fuel, like that available for the Windscale piles, was natural uranium and the moderator was graphite. These choices were constrained by material availability. For the fuel, no enrichment facilities were available in the United Kingdom. For the core, much of the preliminary materials research required to produce high-purity ("nuclear grade") graphite and characterise its behaviour under irradiation, had already been performed for the piles. The new feature required for a power reactor was heat extraction and, hence, the higher operating temperature. This necessitated the use of a gaseous, pressurised coolant and the development of a new fuel cladding alloy (Harris, 1985). The choice of CO_2 as a coolant rather than He was also influenced by material availability at the time. The primary requirement for the cladding was neutronic efficiency. Mg was known to possess a low neutron absorption cross-section and, by the 1950s, there was already industrial experience of Mg alloys, with production routes available, and many properties known. Be, for example, was known to reduce the alloys' tendency to rapid oxidation. Investigation found Mg to show little chemical interaction with U metal. This led to the production of a solution-strengthened Mg-0.8Al-0.004Be alloy for the majority of fuel can requirements, with precipitation-strengthened Mg-0.5/1Mn alloys where higher creep strength was required (at some cost to neutron transparency). The drawback of the Magnox alloys was their low melting point, which restricted their operating range to below 450°C. This limited the coolant temperature and, hence, the thermal efficiency of the reactor. Some improvement to heat transfer was achieved by engineering design (attaching fins to the cladding cans and utilising a high CO_2 flow rate) (Dawson and Phillips, 2012), but the core had to be made relatively large to achieve economic power production rates.

In the earliest Magnox plant, the core was contained in a C-Mn low-alloy steel (LAS) reactor pressure vessel (RPV). This material was chosen because it had a track record as a pressure vessel steel under inactive conditions. The boilers and gas ducting were outside the RPV and the containment. As the power outputs required of later plants were increased, so were the core sizes and the coolant temperatures and pressures. The number and thickness of the C-Mn steel plates involved in the construction of the RPV increased, making consistent quality less easily guaranteed, particularly in the thick-section welds. In later Magnox reactors the steel RPV was replaced with a prestressed concrete pressure vessel encompassing the entire primary circuit. This allowed the higher pressures and also reduced the radiation dose to the workers and general public.

The low coolant temperatures imposed by the use of Magnox cladding meant that the requirements on the secondary, steam-raising side were well within the capabilities of materials used in fossil fuel plants.

Material characterisation programs in association with monitoring and post-irradiation examination (PIE) of used fuel cans ran in parallel with Magnox plant construction and operation, continuing for entire lifetime of the reactor fleet. The degradation mechanisms for the fuel components identified within the first 10 years of Magnox reactor operation included: irradiation growth in the U metal, creep cavitation in the cladding, increased

oxidation of cladding in contaminated CO_2, fatigue failures in cans with certain fin designs and diffusional creep deformation. These degradation processes reduced the lifetime of the early fuel cans below that anticipated from burnup considerations alone. The development of mechanistic understanding and modelling of both irradiation growth and diffusional creep formed a significant part of the programs and aided in developing ameliorations. Grain size refinement via thermo-mechanical treatment and (for the fuel) composition changes, reduced both Irradiation growth and creep cavitation in later fuel cans.

Radiation embrittlement in the cladding was not a major cause of degradation, given the high homologous temperature at which the cladding operated. It was of more interest for the RPV LASs, which operated at temperatures between 170°C and 390°C, mostly receiving doses <0.1dpa. RPV embrittlement was followed using surveillance programs, as summarised in Wootton et al. (2008). The embrittlement rate was soon observed to be unexpectedly high and laboratory campaigns were devoted to developing a mechanistic understanding of the process (essentially precipitation of copper) (Fisher et al., 1985). Understanding the embrittlement process permitted more accurate prediction of the embrittlement rates in different plant, but no remedial action could be taken as the RPV was not designed to be replaceable. When an additional embrittlement mechanism was observed, and found to be due to P segregation, some amelioration could be achieved by reducing operational temperatures (Wootton et al., 2008).

An example of interacting and conflicting component requirements is provided by the various chemical interactions with the coolant. The CO_2 coolant breaks down under irradiation to produce CO which induces graphite corrosion and also leaves carbonaceous deposits on fuel pins and boiler surfaces. Graphite corrosion can be life-limiting, while carbon deposition reduces heat transfer efficiency. The addition of CH_4 to the coolant helps to control the CO level but also breaks down under irradiation, producing water, which greatly increases the corrosion rate of the steel components (Dawson and Phillips, 2012). Oxidation of rimming steel components (in particular, fasteners and bolts) in contaminated CO_2 also contributed to the derating of some Magnox NPP (Shropshire, 2004).

2.2 Generation II–AGR

The Advanced Gas-Cooled Reactor was the UK's Generation II development of the graphite-core, CO_2-coolant reactor. It was a purely power-generating design, intended to exhibit higher thermal efficiency and to operate with a coolant outlet temperature equivalent to that of a coal-fired power station (640°C). This, again, was to avoid the necessity for new designs and new materials in the secondary circuit. In the event, the 9Cr-1Mo steel used for the steam tubes in the "dry-out" temperature range was found to exhibit breakaway oxidation after a long incubation time (Foster, 1975) and replacements were required.

A new cladding material was required for the AGRs, capable of retaining structural strength and resistance to corrosion above 500°C. Austenitic stainless steel was chosen for its track record as a high-temperature structural material. Increasing Cr above that in conventional AISI 300 series stainless steels, while increasing Ni and

Nb improved high temperature strength and creep resistance. The resulting 20-25-Nb (20Cr-25Ni-0.5Nb-2Mn) cladding material has a significantly higher neutron absorption cross-section than Magnox alloy. Thus neutronic efficiency was sacrificed for thermal efficiency. This was offset by increasing the fuel enrichment (by then possible). AGRs use uranium dioxide fuel enriched to 2.5%–3.5%.

As in the later Magnox plant, a prestressed concrete pressure vessel contained the AGR primary circuit and was lined with stainless steel for leak tightness. The operating temperature of the vessel was kept low by a gas baffle directing incoming coolant first around the liner.

AGR construction, unlike Magnox construction, can be separated into a prototype AGR (operating from 1962) and a commercial phase (operating from 1974). The cladding composition was essentially settled during the prototype phase. There was significant overlap elsewhere, however, with construction of various designs of commercial plant beginning in the mid-1960s with limit input from prototype experience.

The more rigorous operational conditions of the AGR led to different materials problems appearing during service. Pellet-cladding interactions, helium embrittlement of the cladding, graphite corrosion, carburisation, sensitisation of the cladding, oxide spalling from steel components transporting active material and depositing dust in the core.

A major perceived advantage of the AGR was its high thermal efficiency (41%). The complexity of the plant, however, offset the economic advantage offered by the thermal efficiency such that the average cost per MWh was greater than that of the less thermally-efficient PWR (Wooldridge and Druce, 2019).

2.3 Generation II/Generation III+: PWR

The development of Pressurised Water Reactors (PWRs) was greatly accelerated by their choice for naval nuclear propulsion in the US in the mid-1940s. A key factor in the choice was the need for a compact design, which was not the case for the gas-cooled reactors. Keeping water liquid at high temperatures requires higher pressures than in the gas-cooled NPP. The PWR choice for the RPV was, therefore, for Mo-containing LASs, as these are stronger than simple C-Mn steels. The RPV steel composition developed from Mn-Mo (A302B) in early plant to Mn-Mo-Ni (A533B and A508) to improve toughness further. Even though these steels had a track record as pressure vessel steels, the precise form in which they were to be used (thick section plates, forgings and welds) required characterisation. This was especially the case as PWR construction spanned the decades in which fracture toughness was developed as a means of describing structural integrity. The extent of work involved may be assessed by the description of the ASME reference curve, describing the bounding toughness *versus* temperature for eleven ferritic LAS base metals and weldments as the "million dollar curve". As with the C-Mn steels of the Magnox RPVs, the Mn-Mo and Mn-Mo-Ni steels of the PWRs were found to embrittle via precipitation of Cu and associated elements under doses of <0.1dpa (Odette, 1983; Odette and Lucas, 1998). Given the extended period over which LWRs have been built (much longer than for Magnox or AGRs), it has been possible to incorporate these insights into changes in the material specifications for later plant, as well as into predictions of

component embrittlement rates. Later LWR RPVs were manufactured using lower-Cu, lower-P steels in the more highly-irradiated regions. Some NPP were built with further increases in the Ni content of the RPV, to reduce the initial ductile-to-brittle transition temperature, anticipating that this would allow for higher neutron doses (longer lifetimes) before the transition temperature became undesirably high. This proved only moderately successful, as higher-Ni steels were found to harden via the precipitation of Mn-Ni-Si-containing features at lower doses and, thus, to embrittle more rapidly than lower-Ni steels (Efsing et al., 2007). Alloy development programs indicate that high-Ni, low-Mn (very low-Cu) RPV steels would combine the advantages of low initial transition temperature and low embrittlement rates (Burke et al., 2012), but no commercial LWR yet uses such steel.

The lower toughness of welds has led to designs in which large ring forgings have replaced plates, to reduce the overall number of welds required in construction, and to avoid welds in the most highly-irradiated part of the RPV (the beltline surrounding the core). The expectation that building larger plant will lead to economies of scale and the increasing demand for electricity have led to the design of Generation III + LWRs with larger cores. This has significantly reduced the number of manufacturers around the world capable of producing the large beltline ring forgings with a guarantee of consistent quality.

The high-temperature water is more corrosive than CO_2, so the PWR ferritic steel RPVs are almost all clad internally with austenitic stainless steel for corrosion protection, as are the pressuriser and the steam generator channel head. This composite structure represents a balance between material cost (the LASs being significantly cheaper) and more complex construction, in providing a component capable of withstanding both stress and corrosive attack. The austenitic stainless steels used for vessel cladding, core internal structures, the coolant pipework and pump/valve casings were chosen for their track record in corrosion resistance, integrity at moderately elevated temperatures and fabricability. Both cast and wrought stainless steels are used. As with the RPV LAS, monitoring programs have shown that radiation-induced degradation becomes significant as the dose increases. Unlike the RPV itself, many of the austenitic components are too small in at least one dimension for standard toughness tests to be performed on portions of extracted components. Extensive databases underpin the current ASTM standards concerning size limitations for samples which indicate whether/how measurements on samples of a given geometry may be extrapolated to the geometry of components.

In the light of the accumulated data, modifications have been made to austenitic steel component compositions and thermomechanical treatments either by changing materials selected for later plant construction or via the replacement of components. Since not all of the components requiring replacement were designed for regular replacement, this increased operational costs above those anticipated originally.

For higher-temperature locations, Ni-based alloys were chosen, mainly the Ni-Cr-Fe Alloy 600. After observations of stress-corrosion cracking in Alloy 600 steam generator tubing increased in the 1970s and 1980s (Jones, 1996; Scott and Combrade, 2019), many research institutions investigated alternative alloy compositions. The higher-Cr Alloy 690 was found to resist stress corrosion and irradiation-assisted stress-corrosion cracking better than Alloy 600 (Sui et al., 1997).

Since it was qualified for use in NPP, it has progressively replaced Alloy 600 in high-temperature components, both in new plant and via upgrading programs during outages of operating plant.

No commercial structural material of the late 1950s combined neutron transparency with resistance to corrosion in high temperature water and radiation damage. Some zirconium alloy properties were known, but it was not in widespread use. The determination that pure zirconium, uncontaminated with Hf, exhibited neutron transparency, taken in combination with evidence for reasonable corrosion resistance and a high melting point, then led to the choice of a Zr alloy as the cladding material. At the time of the choice there was no process available by which to produce high-purity zirconium in bulk, and minimal information on alloy behaviour. Memoirs covering the US development programs show that those involved were aware that a choice requiring concurrent programs in metal production and alloy development were very risky. The authors are clear that obtaining funding, obtaining and exercising the authority to plan and coordinate the programs, and finally utilising constant monitoring and feedback to make sound engineering judgements to drive progress, required a leader with great determination and skill, a very dedicated workforce and a clear, consistent requirement from government (Rickover et al., 1975; Gordon, 2000).

Commercialising the supply chain for zirconium and its alloys required governmental assurances of guaranteed purchases, and other methods of de-risking the setting up of production facilities. Once these facilities were in place, civilian LWR constructors could take advantage of them.

The speed of the program necessitated parallel activities in material development design and construction with continuous feedback. The Mark 1 prototype reactor for the US navy utilised Zircaloy-1 but, as laboratory tests showed its corrosion rate to increase with time, the first submarine utilised Zircaloy-2. Further testing showed that Zircaloy-4 was less susceptible to hydrogen (or hydride-induced) embrittlement, so later naval reactors utilised this alloy (Rickover et al., 1975). Civilian reactors utilised the advances made in the naval program and, while some early civilian PWRs chose stainless steel for fuel cladding, the improvements in the corrosion behaviour of the Zircaloys led to this becoming the standard (Locke, 1975). Over the last decade, more Nb-containing alloys, such as ZIRLO and M5 have been replacing Zircaloy-4. These all oxidise less rapidly and pick up less hydrogen than the Zircaloys. Since the Fukushima-Daiichi accident, further efforts have been made to reduce oxidation and associated hydrogen evolution. Lead assemblies with Cr coatings have been introduced into a number of reactors, but no material has yet been identified which combines the resistance to corrosion in high-temperature water of the Zr-Nb or Zr-Sn-Nb alloys with a higher resistance to oxidation in steam under accident conditions.

3 Generation IV—past present and future

Generation IV NPP have been designed to meet requirements that have become more evident as more Generation I-III NPP have operated and the energy landscape has changed. These additional criteria include.

i. Improved passive safety
ii. Closing the nuclear fuel cycle
iii. Producing high temperature output coolant to support decarbonisation
iv. Higher neutronic and thermal efficiency in electricity production
v. Minimising active waste

Most Generation IV designs require structural materials to operate at higher temperatures, to higher doses and/or in more chemically aggressive environments than Generation I-III NPP. Achieving microstructural and dimensional stability, retaining necessary levels of ductility and toughness, while resisting cracking, corrosion, erosion, and degradation via combined mechanisms (stress-corrosion cracking, irradiation-assisted stress corrosion cracking, creep-fatigue, corrosion-fatigue, flow-asisted corrosion …) under these more extreme conditions, represent challenges shared all Generation IV NPP designs. Where the materials available in the near term are not capable of withstanding these conditions over the anticipated lifetime of the reactor, then either the designs must be adapted to reduce the demands on the materials or programs must be set up to develop materials capable of withstanding operational conditions. Experience with the AGRs illustrated how complexity in the design can increase initial costs and operational difficulties. Modern simulation aids to design are vital in reducing such problems for Generation IV NPP. Similarly, the example of LWR cladding development illustrates the level of cost and effort involved in developing new alloys, let alone new classes of materials.

Some materials development campaigns have been running since the 1980s, with a resurgence of interest in the last decade. A key factor in materials development for a particular component is the way in which the component will interact with the remainder of the plant and its management. Focussing too closely on meeting one requirement may have undesirable knock-on effects. For example, high-Ni alloys are the structural material of initial choice for high temperature applications, but will activate in a high-dose environment, increasing the dose to maintenance staff and the active waste volume. Depending on a high-Ni alloy in a high-dose environment would thus hinder the ability of the plant to meet the safety and waste design criteria above, negating some arguments in favour of the new design.

It is not possible to discuss the materials requirements and choices made for all components in each design, but the following sections highlight some key requirements and decisions.

3.1 Materials developments for fast reactors

Examples of Sodium-Cooled Fast Reactors (SFRs) and Lead (or Lead-Bismuth)-Cooled Fast Reactors (LFRs) have been built and have shown how meeting some of the requirements to improve on Generation I-III designs has increased the demands made on component materials. For the Fast Reactors, the key feature was meeting the requirement to close the nuclear fuel cycle, thereby reducing waste, although the operating temperature are 100°C–200°C higher than LWRs, offering the opportunity for higher thermal efficiency.

For SFRs, the austenitic stainless steels have, again, been the structural materials of choice for vessels, pipework, pumps and heat exchanger shells, based on their qualification for high-temperature operation. Within the heat exchangers, materials problems were associated with 321 stainless steel (which was progressively replaced), with design flaws which led to fatigue cracking or cracking at welds (Guidez and Martin, 2008; Guidez et al., 2008), and poor quality materials.

The most significant increase in the demands made on the structural materials was in the fluxes seen by the core internals and the integrity of the steam boilers. In the earliest prototype SFRs, AISI 316 austenitic stainless steel was used for fuel cladding and ducts but this steel was found to swell considerably under the high operational doses (10s–100 dpa). This was a new and unexpected degradation process. Since fuel assemblies are designed to be removable, however, it was possible to investigate multiple modifications to the fuel assembly materials over the cumulative centuries of SFR operation. The modifications involved both incremental changes to the austenitic steels (cold-working the 316, increasing its Ti content, developing the 15Cr-15Ni-Ti alloy) and more discrete changes towards the use of ferritic Fe-Cr-Mo steels (such as HT9, the ferritic-martensitic T91 or martensitic EM10) (Guidez and Martin, 2008; Yvon et al., 2015). These latter steels were less ductile and more prone to radiation embrittlement than the austenitic steels, but far less prone to swelling. All of these compositions are less neutron transparent than the Mg and Zr alloys of Generation I-III NPP. The possibility of introducing fine dispersions of oxide particles to increase strength and creep resistance and radiation resistance of ferritic steels was investigated, primarily in Japan and France. This would have reduced the neutron absorption penalty by permitting thinner claddings to be used.

Most SFRs were built in the 1970s–1980s and have since closed, with no new construction as public support for nuclear power in general waned and accidents/shutdowns associated with sodium leaks were perceived as particularly dangerous. Modern SFR designs benefit from the lessons learnt concerning material choices, material quality and the necessity to design out demands made on vulnerable components. New SFRs have been built in Russia, China and India, while Japan, France and Canada are considering construction. Research is continuing into producing oxide-dispersion strengthened (ODS)-steels in commercial quantities and developing methods of welding them. This includes improvements to welding techniques and the use of advanced manufacturing processes to reduce the number of joints required to produce complex shapes.

LFRs have been utilised in Russian submarines. LFRs nominally have better passive safety features than SFRs, but the Pb or Pb-Bi eutectic coolants present an additional demand on primary components, being very much more aggressive towards structural metals than Na or NaK. The coolant must therefore contain sufficient oxygen to ensure that the structural material produces a stable, protective oxide even at the highest temperature (plausibly 700°C at hot spots on the cladding) and coolant flow rates, while low enough to avoid Pb oxidation even at the coolant inlet temperature (350°C–400°C). Russian designers have focused on the development of higher-Si austenitic stainless steels to provide a silica-containing oxide layer, though still exercise strong oxygen control. Elsewhere,

alumina-forming austenitic stainless steels have been investigated extensively, as this would permit further expansion of the acceptable coolant oxygen range. Alumina-forming austenitic steels have not been incorporated into a functioning NPP: small heats of such steels have been produced, but they are not commonly commercially available. Difficulties in avoiding the internal oxidation of these silica and alumina-forming steels have also led to investigations of surface alloying with higher-Al material and high-Al or oxide coatings.

This experience with FRs shows how long a developmental phase can last when engineering design does not minimise the known demands on materials, when novel operating environments introduce new demands on materials, and when material consistency and quality are not fully assured. Modern simulation aids to design will play an important part in avoiding the unnecessary demands on components in future NPP designs, and in ensuring that plants permit access for component inspections, especially of welds. The importance of quality assurance in the initial materials, in component manufacture and in construction has been recognised in many industries and improved quality monitoring is a major contributor to the increased availability of modern NPP. Ensuring consistency and quality in large-scale production of base metals and in joining techniques must thus play a major part in the development of new materials for Generation IV NPP.

3.2 Material and code developments for high temperature gas-cooled reactors

Like the Fast Reactors, HTGRs have also exhibited a long developmental phase, with an intermediate period of low activity. The many different HTGRs built since the 1960s differ significantly in size, operating temperature, fuel type and, hence, choice of structural materials (Scheuermann et al., 2017; Iwatsuki et al., 2021). Experimental or prototype HTGRs currently operate in Japan and China. The high gas outlet temperatures have been presented as increasing thermal efficiency, while the He coolant and large thermal inertia of the graphite core represent safety improvements over earlier thermal reactors.

The HTGR pressure vessel experiences lower pressures and radiation doses than those of LWRs, but higher temperatures, even when the design incorporates a dedicated vessel cooling circuit. For example, the Japanese High Temperature Test Reactor (HTTR) utilises a cooling circuit, but the vessel operates around 440°C, higher than the LWR RPVs. The HTTR vessel material is 2¼Cr-1Mo ferritic steel, which achieves greater strength than Mn-Mo-Ni steels at the operating temperature (Tachibana and Iyoku, 2004). Conversely, the Chinese HTR-10 is able to utilise SA516-70 C-Mn-Si LAS without a cooling circuit because the lower power output of the plant requires a lower overall coolant temperature (<300°C). Some proposed HTGR designs, both with and without RPV cooling circuits, are considering the use of the same Mn-Mo-Ni steels used in LWR RPVs. While there is extensive experience of Mn-Mo-Ni steels in NPP RPVs, it is necessary to keep to an appropriate vessel temperature range to take full advantage of the experience. These examples are intended to illustrate the close interplay between power requirements, operational conditions and materials choices. The construction costs and overall complexity of plant design are also involved in this interplay.

The most obviously demanding conditions in the HTGR are those seen by the intermediate heat exchanger, the hot gas duct and the control rod sheaths, into which He passes from the graphite core at temperatures above 700°C. The Japanese HTTR has operated for short periods with a gas outlet temperature of 950°C. Few conventional structural materials have a track record of operating at such high temperatures for extended times. Indeed, few design codes are available to describe the data which engineers need to predict material behaviour during operation at these temperatures: the French RCC-MR covers materials to 700°C and the ASME Code Section III Subsection NH to 950°C (Buckthorpe et al., 2001). As of 2020, only Types 304H and 316H austenitic stainless steel, 2¼Cr-1Mo ferritic steel, modified 9Cr-1Mo, and the high-Ni Alloys 800H and 617 had been qualified against the ASME code, severely limiting the possible choices among readily available structural materials. More recently austenitic steel A709 has also been qualified. Alloy 800 and 800H were used in a number of prototype HTGRs, while Hastelloy-XR is being used in the HTTR. 800H is qualified for nuclear components up to 760°C (with some interest in extending this to 850°C (Swindeman et al., 2006). 617 is qualified for nuclear use up to 950°C. Hastelloy XR is codified in Japan for nuclear use but is not ASME qualified. Programs to acquire sufficient information to qualify Alloy 230 are in place, particularly in France (Yvon et al., 2015). Alloys 230 and 617 and Hastelloy XR all contain Co., which activates severely under irradiation and can contaminate the primary circuit. This makes the very low-Co Alloy 800H (or the as-yet-unqualified 625) a preferred material for the control rod sheath, which experiences a high dose, while material choices are less constrained by the very low doses anticipated for the intermediate heat exchanger and hot gas duct. Expanding the range of materials choices by obtaining appropriate data to qualify new materials would be useful for designers. As with the development of zirconium alloys, it requires significant investment by the interested parties. The campaign to qualify A709 required $15M and 12 years of effort supported by the US Department of Energy (Office of Nuclear Energy, 2020).

The design of the hot gas duct is such that the stresses on the highest-temperature components are kept low, minimising the severity of the operating conditions. Interactions with the coolant, however, cannot be avoided, and are not covered by ASME qualification. The chemical reactions can range from loss of mobile species such as C, N, and O from the surface into high-purity He to surface carburisation or oxidation after the coolant has picked up compounds from the graphite core or degassing seals etc. (Cabet et al., 2006; Cabet, 2008). The effect of surface modification during long component exposures on time-dependent properties such as creep and fatigue has been the subject of extensive laboratory testing (Huchtemann, 1989; Bhanu Sankara Rao et al., 1998; Kurata et al., 1998a; Kurata et al., 1998b; Shankar and Natesan, 2007). As in all designs, coolant composition control affects multiple components: A more oxidising environment will increase degradation rates in the graphite core, but a more carburising environment hardens the near-surface regions of the intermediate heat exchanger and RPV reducing ductility (Ennis et al., 1984). To date, 617 appears to have better creep properties than Hastelloy XR, but poorer corrosion resistance and a higher propensity to activate (higher Co.); 230 has similar creep properties to 617 and is possibly less prone to oxidise

(Yvon et al., 2015). Once corrosion is a factor, then the possibility of environmentally-assisted mechanical failures develops. There is minimal data on environmentally-assisted cracking in these materials.

In the case of Hastelloy XR, the lack of international qualification may be merely procedural, and there may even be commercial, proprietary arguments against publishing details of an alloy into which an organisation has invested a great deal of development time. In more general terms, however, without formal qualification of a material against a recognised Code, it is necessary for a design engineer or safety authority to locate and check, for each component and material choice, sufficient data to support safe operation for the required time under the required conditions. This would be a time-consuming process, and it is up to the prospective NPP designers to decide whether their efforts are better devoted to Code development and material qualification or to more extended discussions with Regulatory and engineering safety bodies. Similarly, it is up to these bodies whether they choose (or can afford) to maintain staff capable of making the necessary judgements or more simply require the use of appropriately qualified materials. Obtaining sufficient data to qualify materials for high-temperature nuclear applications is a long procedure. This is understandable when a component is being relied on to last for the lifetime of the NPP and failure has significant safety, financial and reputational costs.

The use of the latest statistical procedures in designing experimental campaigns and analysing trends in the data will be invaluable in maximising the information acquired from campaigns supporting Generation IV materials development. Even so, the amount of data required to down-select among candidate materials to a short list, and then to provide sufficient characterisation to support engineering decisions, will always be large enough that major experimental campaigns will be required to develop and qualify new materials. Empirical models are commonly used to interpolate data and to permit limited extrapolation (e.g., use of the Larsson-Miller parameter (Larson and Miller, 1952) in unifying creep data), but mechanistic modelling at all length/time scales is likely to provide a more secure basis for greater extrapolations, reducing the risks associated with data extrapolation in Generation IV programs.

The operating conditions of current HTGRs have permitted materials choices for primary circuit components which include currently-qualified materials, materials which are currently unqualified but for which a body of appropriate data exists, and materials requiring conceivable incremental development via optimisation of compositions and initial heat treatments. The advances made in computational thermodynamics [as discussed in (Zinkle, 2017)] can render such material development a much more efficient process than the essentially trial-and-error campaigns of the late 1950s.

3.3 New families of materials for other generation IV designs

The state of development of materials for other Generation IV designs is less advanced. This section highlights some of the challenges posed by these designs, together with some of the most urgent requirements for materials development if plant are to be built.

Very high temperature gas-cooled reactors (VHTRs) are frequently discussed as though they constituted incremental developments of HTGRs but increasing the gas outlet temperature above 900°C not only takes VHTR components out of the temperature range of most current design Codes but also takes them out of the operational ranges achievable by incremental improvements to well-known families of structural materials. New families of materials must be utilized.

The supercritical water reactor (SCWR) is intended to operate over a similar temperature range to the SFR, with components generally experiencing lower doses. This again leads to option of choosing standard structural materials with a track record in many of the required properties. The key factor in material choice thus becomes resistance to the aggressive super-critical water. Experience in non-nuclear applications indicates that the high-Ni alloys and stainless steels are resistant to general corrosion in supercritical water, but susceptible to stress-corrosion cracking (SCC), while ferritic-martensitic alloys are more resistant to SCC but exhibit faster general corrosion. Irradiation-assisted stress-corrosion cracking is more likely in, but not limited to, materials which exhibit SCC. Further information is thus required to characterise the susceptibility of all these materials to irradiation-assisted SCC (Zinkle and Was, 2013).

Molten salt reactors (MSRs) require the core structural materials to exhibit resistance to high temperatures (700°C–1,000°C), high doses (up to 200 dpa) and a highly aggressive environment. Ni is somewhat less soluble in the fluoride salts than Fe or Cr, so material choices have focused on the high-Ni alloys rather than steels, despite the significant increase anticipated in problems associated with activation. The low-Cr alloy Hastelloy-N has been developed for corrosion resistance in molten salts (DeVan and Evans, 1962). Methods of depositing Ni coatings and oxide coatings have also been investigated (Olson et al., 2011; Audigie et al., 2018). Even so, the additional demands of high temperature strength maintained to high doses are unlikely to be achieved, so investigations have expanded to ODS-strengthened Hastelloy-N. Japanese researchers are currently investigating the possibility of producing larger-scale, commercial quantities of ODS Hastelloy N. Campaigns are also required to characterise the material's radiation response over operationally-relevant conditions.

The limitations placed by the common structural materials on the operating conditions of Generation IV reactors have led to new families of materials being considered for structural material applications. In addition to the ODS steels and ODS- Ni alloys, high entropy alloys, MAX phase materials and ceramics have been investigated, based on their anticipated resistance to radiation damage and high temperatures. Monolithic ceramics tend to be too brittle and anisotropic to be viable candidates, but composites have received a great deal of attention. SiC fibres in a predominantly SiC matrix (plus fillers, and interlayers) have been investigated most extensively, although other carbides (TiC, ZrC), nitrides and graphite (C/C) composites have also been investigated (Yvon et al., 2015). If successful, these materials would expand the options for e.g., HTR and VHTR control rod sheaths, gas-cooled FR fuel cladding, SFR wrapper tubes. In parallel, work is proceeding on developing design rules and codes for coated or clad materials so that the adequacy of such combinations of materials can assessed and exhibited (Barua et al., 2020).

Materials from these newer families have mostly not yet been produced in sufficient quantities to judge whether component-sized sections can be manufactured, whether large sections will exhibit consistent properties and whether their desirable properties can be retained in jointed/welded components. SiC/SiC tubes have been produced and several Standard Practices have already been developed to test their mechanical properties, leak-tightness (with and without metallic liners) and joint strength (Deck et al., 2015).

Where materials are available in less-than-component quantities, development is essentially at the screening stage. This description is applicable to various ODS materials, high entropy alloys, MAX phases and other ceramics. The question then arises as to whether the screening procedures possible with small amounts of material are properly representative. General corrosion tests and investigations of microstructural stability are clearly interpretable. Mechanical tests and stress-corrosion tests are less consistent. The use of hardness indentations and shear punch tests to follow yield stress changes is well-attested for steels and Ni-based alloys; small punch tests somewhat less so. The property exhibited in indentation tests is different for ceramics which crack under the indenter but, again, there is a background against which the response of a new material can be assessed. Micropillar compression tests are increasingly providing information on the deformation mechanisms of ultra-small samples. In general, the standardisation of practices is supporting greater reliability in the use of small scale techniques to compare different families of materials.

Fusion reactors represent an extreme case, with materials being required for multiple novel exposure conditions. The technological readiness of different components varies widely. For example, tungsten is generally agreed for the diverter. Supply routes for this material exist and there are generally-available design rules governing the use of refractory materials in different regulatory regimes. Work is still required to extend the appropriate standards and rules to the diverter conditions, and to identify suitable tests to allow quality assurance and material acceptance. Optimising methods of joining the W diverter to other structural materials and assessing the quality of the weld/joint are still in progress. In contrast, materials for the breeder blanket and neutron absorbers are still being scoped. Breeder blanket structural materials will be required in large quantities, requiring suitable supply routes and reliable techniques for both homogeneous and heterogenous joints. The current candidates range from standard structural materials, such as P91 steel, coated with a lithium-resistant oxide coating (e.g., erbia) to non-standard metallic alloys, such as those of vanadium, which may be intrinsically Li-resistant, but are associated with minimal engineering data. The V alloys are also more brittle than current structural materials, requiring designs which are not vulnerable to limited ductility, and are not associated with mature supply chains. Candidates with even lower technological readiness levels also remain under consideration. An engineering solution such as separating the tritium breeding facility from the tritium burning facility would also reduce the complexity of the materials requirements. Overcoming the materials challenges involves large-scale effort over long periods. For the last 50 years this has predominantly supplied by governments, but investments from private companies are now significant.

3.4 Small modular and micro-reactors

Small modular reactors are frequently modifications to the Generation III and IV designs used for standard-sized NPP. This allows the operational experience accumulated with different materials in relevant environments to minimize the materials development required. The materials choices thus tend to start with the materials used in standard NPP. Nonetheless, SMR environments do generally differ from standard NPP environments in ways that increase demands on components: components may be placed closer to the core, resulting in higher fluxes and temperatures than seen by their equivalents in standard NPP; components may be smaller experiencing higher stresses; faster coolant flows may be required imposing higher stresses, erosion/corrosion rates or different vibrational (fatigue) regimes. Responses to such modifications require the acquisition of new engineering data at least, and plausibly further materials development.

4 Insights from historic materials choices

This very brief summary of some of the material choices made and issues encountered in operational NPP highlights some key factors in historic choices of materials.

- Material availability and limitations on development time have played a major role in constraining materials choices for particular NPP designs.
- Even before reactor designs have been finalised, the requirements for component properties have been scoped, such that most component materials can be defined at an early stage.
- The ability to achieve consistent manufacturing quality at large scale has been a key material requirement. The problems associated with the essential inhomogeneity of welds and joints have been reduced by design changes to reduce the numbers of welds/joints or reduce the severity of the environment in which they are placed.
  ○ Modifying new NPP designs to reduce the demands on materials, and increase the operating margin, will be cheaper than developing new materials.
  ○ Ease of preparing high quality joints is a baseline requirement for new materials.
- Materials with known track records have been used wherever possible (e.g., low alloy pressure vessel steels, 300 series stainless steel liners and internal support structures, high-nickel alloys for high temperature components).
  ○ The existence today of large, readily-interrogated materials databases should make such materials choices simpler than in the 1950s and 1960s.
- Operational experience has shown that the new aspects of the NPP operating environments have introduced or enhanced degradation processes. As new information emerged from operational experience and laboratory campaigns, further modifications of these "known" materials have been made

(e.g., reductions in residuals in RPV steels, shift from Alloy 600–690).

- ○ This highlights the importance for new NPPs of designing in monitoring campaigns, to allow unexpected problems to be observed at an early stage, and for plants or fleets of plants to support materials investigations to identify degradation mechanisms and find answers to the problems.
- ○ Improvements in on-line monitoring and non-destructive examination techniques should permit increasingly thorough condition monitoring of susceptible components.

- Where new materials have been required, some have been developed from conventional materials known to possess at least some of the desired properties (e.g., Magnox and AGR cladding).
  - ○ There has been an enormous expansion in the modelling of equilibrium phases, rates of phase transformations and thermo-mechanical processing since the 1960s. Taking advantage of these different aspects of modelling should speed up incremental alloy developments required for future NPP.

- Entirely new materials have been developed to meet particular component demands, although this has been considered a risky choice. Very large and well-focused programs were required to take new materials from concept to component manufacture (e.g., Zr alloy cladding). This option has been used only when the components involved were readily replaceable.
  - ○ Selecting NPP designs which minimise the number of irreplaceable components should allow engineers to be more flexible in their materials choices and make the plant more resilient in the face of unpredicted degradation processes.

- Ensuring security of supply for the testing of new materials and their use in component manufacture has required collaboration between the material manufacturers, plant designers and plant constructors. Government support has been used to de-risk the building of the associated large-scale production facilities.

5 Considerations for future plant

It is imperative that supporters of designs for future NPP appreciate the lessons learnt from historic materials choices and experience. NPPs involve complex combinations of interacting components. When materials capabilities limit the operating conditions of a particular component, this has knock-on effects for the requirements to be met by other components, for the power output of the plant and for its economic efficiency. Material characterisation, component monitoring and materials development campaigns have, thus, been part of every NPP's design, construction and operation phases and will be vital in supporting the evolution of new designs.

Using intelligent design to reduce the extent and complexity of demands on components has been successful in the past, and will be the first choice for any NPP designer. If suitable structural materials (i.e., materials available at scale, well-characterised for the operating environments of each component and inducing only benign

interactions between components) are not available, then the history of materials choices for NPP built to date shows that campaigns looking for incremental advances in materials capabilities have been preferred to those attempting to produce step changes in materials. Developing entirely new alloys to the stage that they can be used as structural materials requires a very well-funded, well-focused and dedicated campaign. Obtaining a new class of materials to expand the extend the operational envelope of components is very desirable, but is likely to take campaigns over several more decades to bear fruit.

It is self-evident that those Generation IV designs that can be modified to utilise currently-available materials, while still operating to provide the advantages claimed over Generation III designs (mostly enhanced safety, reduced overall waste, greater economic efficiency, flexibility in decarburisation), are the most likely to lead to near-term NPP construction. The more advantages that can be met by designs which require only current materials, or materials which will be characterised and available at scale in the next decade, the more difficult it will be to retain a market niche for the remaining fission designs. It will be necessary to make hard decisions, down-selecting reactor designs and material types if the purpose of the materials development campaigns is truly to support safe, reliable, low-carbon electricity production before 2050. Justifying the risk of materials development campaigns which, even if successful, will require more than a decade to initiate large-scale manufacturing will rely on the possibility of using the materials outside the nuclear area, as well as for the associated Generation IV NPP design. Fusion NPP are likely to occupy a unique niche in the energy supply landscape, regardless of the quantity of electricity supplied by fission plant. It is, therefore, possible to justify much longer campaigns to develop materials capable of withstanding the extreme conditions that will be seen by fission reactor components.

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