PAPER • OPEN ACCESS

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To cite this article: Peter Trampus 2020 *IOP Conf. Ser.: Mater. Sci. Eng.* **903** 012044

IOP Conf. Series: Materials Science and Engineering **903** (2020) 012044 doi:10.1088/1757-899X/903/1/012044

On the necessity of domestic research on nuclear materials science

Peter Trampus

Professor emeritus, University of Dunaújváros, Dunaújváros, Hungary

Email: trampus@uniduna.hu

Abstract. Globally, systematic research is going on to better understand the operation induced degradation effects of reactor structural materials and to enhance the effectiveness of their in-service monitoring. In Hungary, there are four reactor units in operation and two new ones are soon to be built. Some of the construction features of the new units differ from that of the operating ones. In order to be prepared in time for efficiently managing materials science related questions, it is necessary to establish a coordinated research program utilizing the capabilities of domestic research institutions. This paper describes the major elements of this potential program. Implementation of the program can assist ageing management activities of the operating units.

1. Introduction

For more than 40 years, Hungary has belonged to that relatively small, elite group of countries, in which nuclear power plants (NPPs) are in operation. The Hungarian energy strategy plans on long term use of nuclear power; consequently, the currently operating units will be replaced by nuclear based capacity after their retirement. The four "old" reactors at Paks site has run for more than 30 years, i.e. the originally licensed term, thus, structural materials of mechanical components and structures are ageing. The term ageing means the continuous change in material properties due to service life and/or operation loading and the environment. Construction of two "new" reactors at the same site has already started; the components of these units are intended to serve for 60 years.

Having accumulated some twenty thousand reactor year experience worldwide, the civil nuclear technology is unquestionably mature. Despite of matureness, the prospects of nuclear energy expansion is not considered positive in every region of the world. Global climate change and energy generation are in strong connection because generation related emissions take a significant portion of greenhouse gas emission. NPPs can mean a vital option for decarbonisation of energy systems. As a consequence of all these global issues, the value of operating reactors is increasing and life management, service life extension – nowadays called: long-term operation (LTO) – are playing a central role in the world's nuclear power generation. We are close to the truth when saying that LTO is not on the agenda only in countries where it is hampered by political will. The major characteristic of the new nuclear power projects is that more and more developing countries plan to embark in nuclear energy utilisation.

2. The role and importance of materials science and engineering

To assess structural integrity, i.e. the safety of mechanical components and structures, knowledge in materials science is indispensable. The basic elements of materials science and engineering, which create an intellectual foundation are the following: (1) composition and structure, (2) synthesis/processing, (3) properties, and (4) performance under operating circumstances. Nature laws control the individual elements and their coherence. When building new NPPs and during long-term plant operation, the key for both engineering and operational safety is the integrated application of each element of materials science.

Since 1970s, the operating license of the nuclear reactors has been issued by regulators for a predetermined period. This period was typically 40, in the case of Soviet designed units 30 years, determined by design considerations (also called design life) or – in the USA – by antitrust law. The fact that a nuclear power plant can be operated beyond its originally licensed term, even for double period, is acceptable if it is properly justified technically. The world's nuclear industry has elaborated and routinely been using the service life extension methodology [1]. In the light of the accumulated experience, one could even state that there is enough knowledge available on structural materials' behaviour under normal operating and potential off-normal conditions. However, this statement may be questioned, because in the past decade more and more reactors entered into the LTO phase. Figure 1 illustrates the age of the world's operating reactors [2].

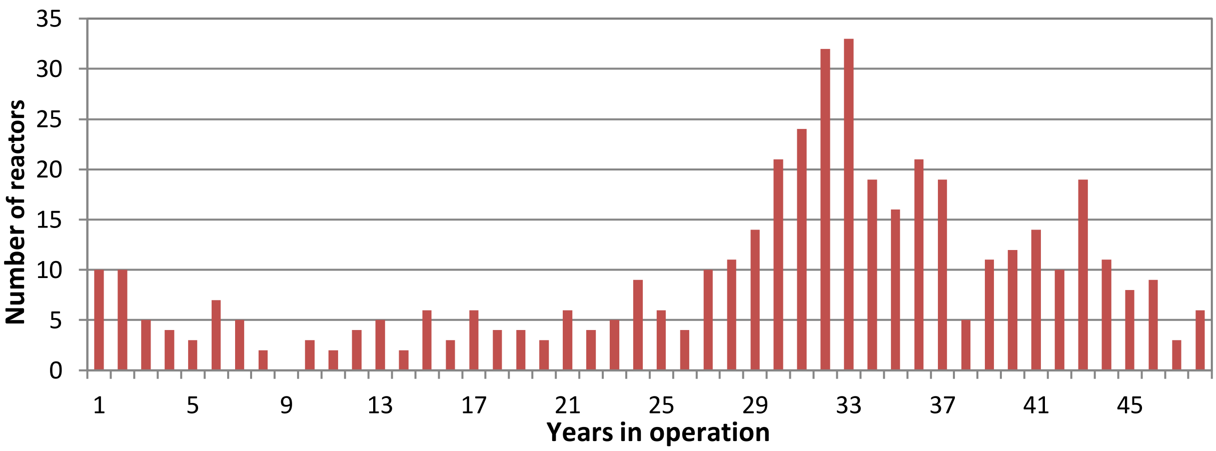

Figure 1. Distribution of operating reactors by age (as of December 12, 2016.)

The direct consequence of operation beyond design life is an increase of ageing effects on the components. Moreover, some ageing processes can accelerate and even degradations not considered by the designer may occur. Many factors influence reactor materials' ageing. Chemical composition and microstructure determine the initial mechanical properties (e.g. tensile strength, fracture toughness). Degree of loading (e.g. stresses, number of cycles) and its mode (e.g. static, dynamic, cyclic), as well as environmental effects (e.g. temperature, radiation, corrosive agent) can initiate the degradation processes, which lead to a gradual decrease in initial properties. Once the degradation exceeds a certain limit, the component fails. Cracks or crack-like flaws in the material have a significant influence on the degradation processes and thus on structural integrity. The aforementioned factors have different weight in components' degradation and usually a single factor leads to failure. It is possible, however, that more than one degradation processes are active at the same time, manifesting in a synergistic effect (e.g. environmentally assisted fatigue, irradiation accelerated stress corrosion cracking).

Mechanical components of NPPs are usually made of conventional structural materials (steels, nickel-based alloys). Materials development is evolutionary; enhancement of their operating features is achieved through the improvement of manufacturing processes or optimisation of operation and maintenance. Structural materials of the new reactors (so called fourth generation, fission based) most probably will not have significantly more rigorous requirements.

By extending plant life management, i.e. power uprate and/or LTO, the attention of research tends to focus on reactor structural materials' behaviour. In this context, one of the most challenging tasks is the determination of the actual components condition and, based on this, the estimation of their technically achievable service life. This requires an intensification of the knowledge on degradation mechanisms, development of physically based degradation models, improvement of extrapolation methods, as well as the improvement of destructive and non-destructive testing methods and techniques to monitor materials' condition and elaboration of new maintenance and repair technologies.

3. The new reactor project in Hungary

Hungary's four old reactors (Russian design, VVER-440/V-213, current electric output: 500 MW each) are planned to shut down between 2032 and 2037. The lost capacity will be replaced by two new reactor units (also Russian design, VVER-1200 with an electric output of 1200 MW each).

3.1. Historical retrospection

Before investigating the materials science related questions of the new reactors, let us briefly look back to the situation of the old units' construction phase. In 1980, the Hungarian Academy of Sciences (HAS) established a program on „Research tasks for serving safe operation of NPPs", under the leadership and coordination of the Central Research Institute for Physics. One of its sub-programs was dedicated to „Strength analyses and in-service inspections to determine components condition and estimated service life", led by the Research Institute for Ferrous Metallurgy. The sub-program budget was approximately 5 billion HUF (converted into current value). Within the framework of this sub-program, research was conducted in the following areas between 1981 and 1985 [3]:

- Modelling and investigating thermal ageing and low-cycle fatigue in structural materials of the reactor coolant system, and determination of crack initiations.
- Investigating neutron irradiation embrittlement of reactor pressure vessel (RPV) material and developing methodology for monitoring the embrittlement process.
- Investigating crack stability in the pressurised components of the primary circuit. Creating data bank to estimate system reliability (fracture probability).
- Acoustic emission testing to identify defects in pressurised components to increase reliability of the evaluation of primary circuit components' inspections.
- Developing scientific basis for ultrasonic testing of primary circuit pressurised systems.
- Selecting numerical and experimental methods to determine stress-strain conditions of pressurised components, taking into account operating and transient loading and defects with various shape and size.
- Investigating forms and extension of cracks due to corrosion degradations, stress corrosion and corrosion fatigue for RPV pressure retaining and cladding materials, nozzles, weld materials, and welded joints.
- Comparing the conditions of stress corrosion and corrosion fatigue crack initiation and extension using fracture mechanics testing and numerical models.

The sub-program contributed to the development of a broad, local expertise in the subject, which helped to handle any material related problems during construction of the units. At the same time, the knowledge acquired could be well applied in the operation period. To the best of the author's knowledge, there is no existing or planned coordinated research program dedicated to materials' integrity issues of the new reactors. The solution to this question does not depend on the materials science community only, but rather on political decision makers.

3.2. Justification of the research program's necessity

Since the coordinated nuclear materials science research program described above was established, a new generation of materials scientists and engineers have grown up. Their individual knowledge is unquestionable, but it is also undoubted that an integrated knowledge base established by a properly coordinated program worth much more than the simple sum of knowledge owned by individual experts.

According to the author's opinion, the main reason for the need of establishing a coordinated, domestic, material related research program is that the construction of the new reactors differs in some – not negligible and not properly investigated – solutions from the operating ones. One difference is the RPV material: the manufacturer uses nickel as an alloying element. The effect of nickel is known from the literature [4, 5], but neither data for LTO is yet available, nor domestic practical experience and experimental database exist. Nickel as an alloying element – in certain circumstances – can reduce the resistance of steel against irradiation embrittlement. Another, more general difference is the method of the RPV structural integrity assessment. Currently, there are two engineering methods, namely, the internationally accepted Master Curve [6] and the Russian Unified Curve [7] that allow the construction of the time-dependence curve of fracture toughness for various RPV steels. The Unified Curve differs from the Master Curve in that it takes into account the transformation of the shape of the curve as a function of the embrittlement of a material.

Another fundamental difference between the new and old reactors is related partly to material selection and partly to manufacturing. The new reactor coolant pipeline is made of low-alloy steel equipped with stainless steel cladding in its internal surface. This solution is new considering both the corrosion resistance behaviour and the in-service inspection, i.e. periodic non-destructive examinations. Dissimilar metal welds (DMWs), like cladding and base metal interface, may have an increased corrosion sensitivity. The production of welded joint is more than a routine procedure because of the difficulties resulting from different chemical composition, microstructure, and thermal-physical characteristics of the three materials. These also make the DMW non-destructive examination more difficult. Figure 2. shows an ultrasonic testing simulation on a DMW illustrating some anomalies in sound beam paths. Regarding crack initiation sensitivity and in-service inspection problems of the cladding and especially the underclad area, the operator of the old reactors gained some experience on RPVs.

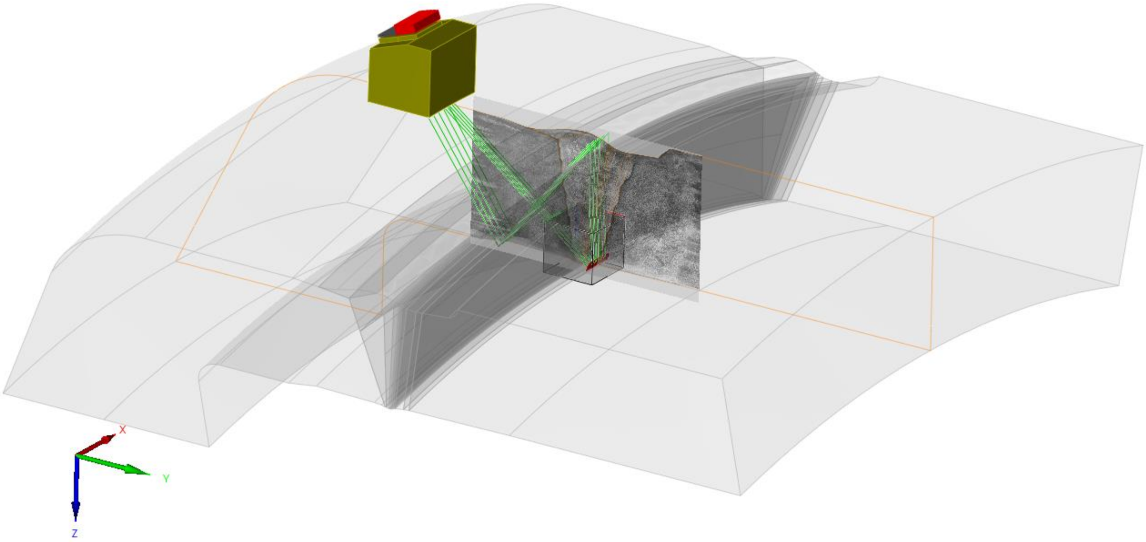

Figure 2. Simulation of dissimilar metal weld examination [8]

If the primary coolant pipelines (except for the reactor coolant) is made of the same austenitic steel as in the old units, it is advisable to apply the practical experiences gained by plant operators during the third decade of operation [9]. Experience showed that there is still not enough knowledge on special corrosion phenomena; such process is the microbiologically influenced pitting corrosion.

As mentioned above, according to the design intent, the new reactors will operate for 60 years. This gives another strong argument for the coordinated program, i.e. creation of an up-to-date database meeting Industry 4.0 requirements must be started during or preferentially before the construction period. The lack of pre-service condition experience of mechanical components in the "old" units' operation often led to serious difficulties.

3.3. *Objectives of the research program*

To be prepared for material related issues of the new reactors, the author recommends to consider the establishment of a state-sponsored, coordinated research program involving competent institutes and experts of Hungary. Ideally, duration of the research program should last for at least 5 years. The minimum objectives should be the following:

3.3.1. Establishing knowledge base. It is necessary to be familiar with the properties of the structural materials and production technologies of the new reactors, and it is necessary to understand the physical basis of ensuring and maintaining necessary properties. The best way to achieve this objective is to collect results of our own research within the coordinated program.

3.3.2. Creating foundation of the ageing management program. Today, one if not the most important criterion of safe and reliable NPP operation is to establish and maintain a systematic and integrated ageing management program. Besides knowing the fundamental characteristics of structural materials, dominant, service induced degradation processes have to be identified and understood from the point of view of substantiate the components' ageing management program. Since the ageing management program has to be available from the start-up of the new reactors, the coordinated research program provides a unique opportunity to lay down its foundations.

3.3.3. Preparing the domestic institutes for solving material issues. During the construction and in the first decade of operation of the old reactors, the law required notification of a leading institute on materials science. The licensee had to ask for the institute's expert opinion in subjects like material selection, material procedures, especially welding, material testing, failure assessment; the nuclear regulator has always considered this expert opinion as a gold standard when taking decisions. This mechanism is still present in the Russian normative technical documents but no longer legally binding in Hungary. There is no intention, of course, to re-establish this mechanism, but without any doubt, the task itself – with its complexity and diversity – will permanently come up and has to be solved.

3.3.4. Contributing to long-term, safe and reliable plant operation. All materials science related issues, without exception, contribute to the assurance of the mechanical components' structural integrity. Structural integrity as an important pillar of the "defence in depth" concept [10] is one of the most important conditions of plant operational safety as well as economic efficiency.

3.4. *Organisation of the research program*

The author of this paper is strongly convinced that the establishment of a coordinated research program with the aforementioned objectives is reasonable and necessary to be able to solve this complex and large-scale task sufficiently. Currently no single institute of Hungary accumulates such a knowledge base that could cover all relevant scientific areas presented here. Therefore, it seems advisable to delegate the elaboration and then the coordination of the coordinated research program to the Committee for Materials Science and Technology of HAS. The HAS Committee must be capable to gather and coordinate experts internationally recognized. The best form of the program's operative implementation is still to be discussed and agreed upon. One possible option could be the

IOP Conf. Series: Materials Science and Engineering **903** (2020) 012044 doi:10.1088/1757-899X/903/1/012044

establishment of a so-called 'virtual' institute composed of assigned experts of relevant scientific areas.

4. Materials challenges of the operating reactors

As mentioned earlier, ageing phenomena and related issues get into the focus of LTO. NPPs one after the other reach a stage of their operating life, that no single NPP in the world has ever reached yet. It is not a coincidence that worldwide there is ongoing systematic research activity to better understand the operation induced degradation effects of reactor structural materials and to enhance the effectiveness of their monitoring.

On the basis of research results and possessing plentiful operation experience, material scientists agree that structural materials of the reactors in operation or under construction face the following significant ageing challenges [11, 12]:

- Neutron irradiation embrittlement and loss of toughness.
- Corrosion, especially its localised forms, e.g. stress corrosion cracking (SCC) and irradiation-assisted SCC.
- Environmentally assisted fatigue.

The ductile-to-brittle transition temperature shift and decrease in toughness (usually known as embrittlement) are typical means of degradation of RPV pressure retaining material. The origin of this issue stems from the impact of the increased fast neutron fluence on the ageing process due to extended service life. Figure 3 shows the scanning electron-microscopic (SEM) image of the broken surface of a half Charpy specimen; brittle fracture is caused by loss of grain boundary cohesion due to phosphorus segregation on grain boundaries (fluence: $1.07 \cdot 10^{25}$ $n \cdot m^{-2}$, E>0.5 MeV). In this regard, it is still necessary to investigate the influence of impurities of the RPV steels on microstructure changes (e.g. phosphorus), or the influence of nickel on formation of Ni-rich, so called late blooming phases and the impact of these phases on the embrittlement process. The RPV structural integrity assessment requires further research: The Master Curve concept enabling direct measure of fracture toughness still needs quantitative validation (see earlier). LTO also requires to keep the thermal embrittlement investigation on the research agenda. Increased attention, and consequently, research is necessary in the area of welded joints integrity with special regard to the behaviour of DMWs, joining alloys with different chemical compositions, microstructures and thermal-physical characteristics. In the case of DMWs, corrosion can play an important role.

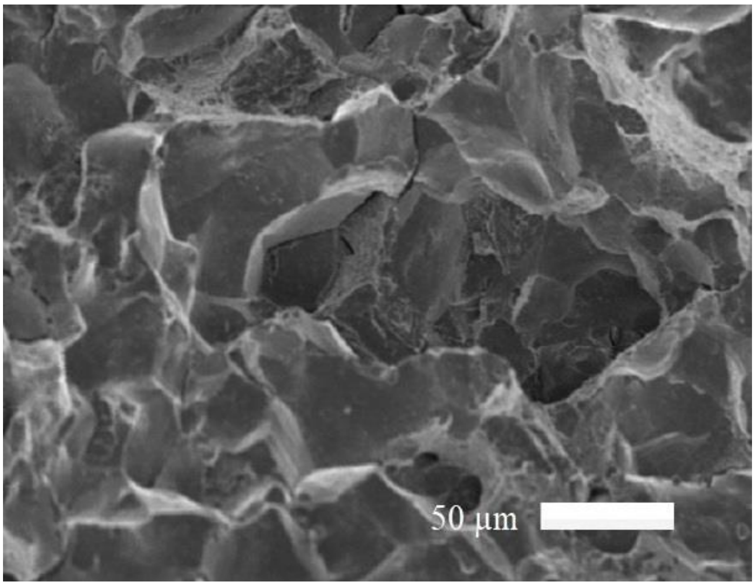

Figure 3. Cleavage fracture on irradiated Russian RPV steel (SEM)

Corrosion attacks the components and pipelines made of stainless steel, e.g. RPV cladding, reactor internal structures that control hydraulic flow, support fuel assemblies, and control rods. SCC degradation mainly develops in the steam generator heat exchanger tubing made of nickel-based alloy. SCC can appear in stainless steel components, too (e.g. steam generator tubing). Serious consequences of SCC can occur in reactor internals: in the case of the baffle bolts in core basket, as a result of a highly complex degradation effects, the irradiation assisted SCC leads – in extreme case – to the fracture of the cracked bolt head. Figure 4 shows the relations of loading situation and degradation effects.

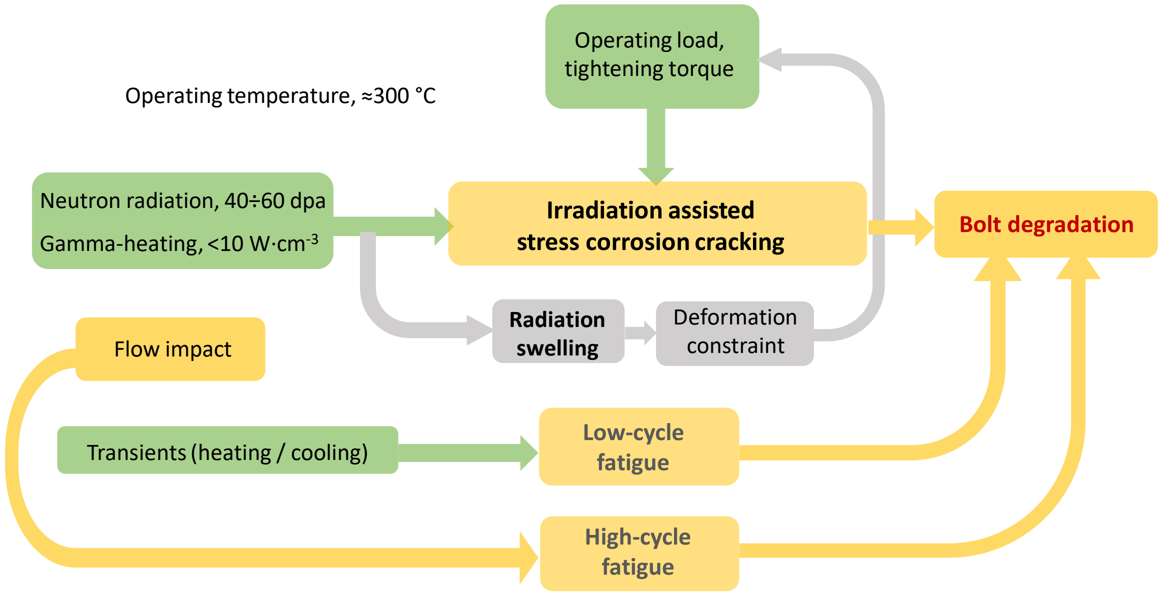

Figure 4. Loading of and their impact on baffle bolts [13]

In LTO, the austenitic stainless-steel pipes of the reactor cooling system dwell longer on the operation temperature, than they were originally expected by the designer. During the longer dwell-time, thermal activation process leads to precipitation of grain boundary carbides and thus to sensibilisation of the grain boundary area against intergranular attack (IGA). Even though the operation temperature is relatively low, depending on other factors such as time, C-content, Ni-content the austenitic steels may be sensitive to IGA. Figure 5 shows an example: dwell time = 225 000 hours, temperature = 300°C, C = 0.08%, Ni = 10%). The grain boundary carbide precipitation is clearly visible [14].

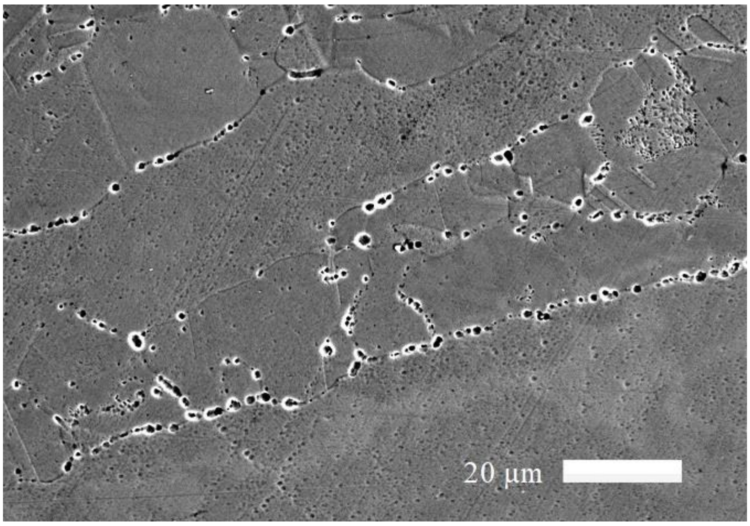

Figure 5. Grain boundary segregation in stainless steel (etching: ASTM A262 Practice A, SEM)

Environmentally assisted fatigue (EAF), sometimes called as corrosion fatigue, concerns the reduction in fatigue life that is observed in a reactor water environment compared to room temperature air. EAF involves two primary elements: 1) the effects of reactor water on the overall fatigue life of reactor components (as represented by either multiplying the fatigue usage factor by an 'environmental factor' to account for aqueous corrosion effects or the use of an environment-adjusted fatigue design curve), and 2) the potential accelerated growth of an identified defect caused by exposure to reactor water environments. Fatigue crack initiation and growth resistance is governed by a number of materials, structural and environmental factors, such as stress range, temperature, dissolved oxygen or hydrogen concentration, mean stress, loading frequency (strain rate), surface roughness, and number of cycles [12].

5. Conclusions

It is obvious that both building of the two new reactors and long-term operation of the four old units define numerous issues for the Hungarian materials science community in the coming decades. The author of this paper is strongly convinced that establishment of a coordinated research program is reasonable and necessary to be able to solve this complex and large-scale task sufficiently. The major objectives of the research program were presented here. It seems advisable to delegate the elaboration and coordination of the program to the Committee for Materials Science and Technology of HAS. Currently no single institute of the country accumulates such an expert system that could cover all relevant scientific areas presented here. The HAS, however, is capable to gather and coordinate experts recognized internationally and provide them with appropriate financial support. The form of the program's operative implementation is still to be discussed and agreed upon. One possible scenario includes the establishment of a so-called 'virtual' institute composed of assigned experts of relevant scientific areas.

Acknowledgment

The author would like to express his gratitude to professors L. Tóth and B. Verő for their invaluable suggestions to the research program, and to F. Oszvald for performing the electron-microscopic investigations.

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