

SAFIR2018 Project plan

COMRADE

Condition Monitoring, Thermal and Radiation Degradation of Polymers Inside NPP Con- tainments

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1. Research theme and motivation

A joint Nordic project on Condition Monitoring, thermal and Radiation Degradation of polymers inside NPP containments (COMRADE) has been developed, following an initiative from the Nordic NPPs through Energiforsk. The project development has been made by SP Technical Research Institute of Sweden and at VTT Technical Research Center of Finland.

COMRADE is developed based on input from a feasibility study from Energiforsk AB [2015:157], an ongoing study ordered by STUK [Penttilä, 2016] and through discussions between VTT Technical Research Center of Finland, SP and the Nordic NPPs through Energiforsk. When developing COMRADE it was understood that there are gaps in knowledge for setting functional based acceptance criteria at the nuclear power plants. Furthermore a need in gaining a better understanding on how a polymeric component reacts to different levels of low dose radiation and synergistic effects between thermo-oxidative and irradiation degradation was identified. The plan is to divide the work into different steps, all with the aim of providing the power plant operators as well as regulators and polymer manufactures with a deeper knowledge of the degradation of polymers and to develop methods for setting acceptance criteria of polymeric materials. This is done through three work packages:

- WP 1 focusing on method development of condition monitoring and implementation at NPPs
- WP 2 is a pre study to map how the closed down plant Barsebäck can be used to verify the method developed in WP1
- WP3 focusing on polymer ageing mechanism and effects inside the NPP containment

By completing these work packages the nuclear power plants and radiation safety authorities will have the possibility to use functional based acceptance criteria, see how Barsebäck can be used to verify models for acceptance criterias, gain better understanding in synergistics effects between heat and irradiation and see how different levels of low dose irradiation will affect the polymeric component.

The work will be done in cooperation between VTT, SP Technical Research Institute of Sweden, the Swedish and Finnish Nuclear Industry through Energiforsk and a manufacturer of nuclear grade elastomers. Polymer experts from the Nordic NPPs will have an active role in WP1. The work packages are all managed by one of the institutes, but the work done within the packages is shared between the participants.

1.1 Background and state-of-the-art

According to IAEA, operating experience has shown that ineffective control of ageing degradation of the major Nuclear Power Plant (NPP) components can jeopardize plant safety and also plant life. Ageing in NPPs must therefore be effectively managed to ensure the availability of design functions throughout the plant service life [IAEA, 2000]. As discussed at the Energiforsk (ELFORSK) Seminar on aging of polymeric materials in NPPs held in 2014 the importance of defining a method to determine the acceptance criteria for polymeric components was pointed out. The acceptance criteria should be based on functional demands since it is the first step in the process of lifetime estimations of existing or new materials. As a result of the seminar a feasibility study featuring interviews with polymer experts at the NPPs in Sweden and Finland was launched to investigate the preliminary viability of defining a method to determine acceptance criteria for polymeric materials. The feasibility study was carried out by polymer experts at SP during March through August. The study started with a set of interviews with the 5 Nordic nuclear power plants Loviisa, TVO, Forsmark, OKG and Ringhals. Through the interviews a chosen set of components were identified to be included in the feasibility study. This includes but is not limited to type of polymer and environmental aspects such as heat and radiation. Based on the components, and the type of poly-

mers used for it, a literature study was done in order to identify existing knowledge on aging mechanisms. From the knowledge gained through the literature study and existing know-how at SP, tables with suitable methods was proposed, if possible, acceptance criteria was concluded for each component. However, it was difficult to find already defined acceptance criteria for the proposed polymeric components.

As concluded in the feasibility study [2015:157] there are issues with identifying already available acceptance criterias based on functional demands in existing research. There are tests done when material properties have been monitored during accelerated ageing through both radiation and heat. There is one recent doctoral thesis done at KTH Polymeric materials in nuclear power plants – Lifetime prediction, condition monitoring and simulation of ageing [Linde, 2015] which tests how properties changes over time in accelerated tests using both radiation and heat. The test was made on insulation for electrical cables (PVC, EPR, XLPE and NBR). Even though the tests showed interesting results for evaluating ageing properties tests done are not verified by the actual function of the polymeric component but focuses instead at the degradation of a material property. To be able to set an acceptance criteria based on the function there is a need to correlate the material property changes to a change in the function of the polymeric component.

1.1.1 Ageing

Based on the interviews in the Energiforsk report 2015:157 different components were chosen by the 5 Nordic NPPs. These components are estimated to cover a large portion of the polymeric materials in system component at a NPP and a more detailed percentage is provided in WP1. By studying a function based acceptance criteria for these components and correlates it to a material property the NPPs will be able to better understand the lifetime of their polymeric components. It will also help the regulatory authority to better understand how the function of the polymeric components changes over time and what causes the degradation.

Polymeric materials used in NPPs are subjected to different temperatures, dose rate and total doses of radiation. The polymers expected life time depends on its resistance to degradation and the environment it is subjected to. During its life time it will receive a total dose of radiation at a specific temperature. These are factors that need to be taken into account when developing the method for ageing. As written in Prediction of service lifetimes of elastomeric seals during radiation ageing [Burnay, 1984] historically radiation tests on polymeric material, samples have been irradiated at high dose rate ~10 kGy/h (1 Gy = 100 rad) to obtain “lifetime dose”. However, it has been well established that many polymers exhibit dose rate effects or synergism between temperature and radiation making high dose rate predictions of limited use. An exposure at high irradiation also concentrates the ageing to the surface, which can generate insufficient correlation to real use. For accelerated ageing using temperature the relationship between temperature and heat may follow the Arrhenius equation which is commonly used for accelerated tests for polymers not subjected to radiation. It is important for both heat and radiation to consider the potential risk in changing the degrading reactions when using too high temperature or too high dose rate.

The dose rate for the components, during normal operation, in [2015:157] ranges from none, low (mGy/h) to a highest range of (0,1 – 0,5 Gy/h). These dose rates are far below what has been used in experimental trials identified in the literature study. Values are often in the magnitude of 200 times higher than what is estimated as the higher dose rate range for the components in [2015:157]. Therefore this study will be done using lower dose rates for the work done when studying the functional based acceptance criterias. There will also be a work package studying the dose rate effect in order to determine the detrimental effects of this phenomenon to different polymer components during accident situation and normal usage of NPP.

1.1.2 Synergy of radiation and heat and DLO effects

The synergism in degradation of a polymer when exposed to heat and radiation is further supported in Degradation of elastomer by heat and/or radiation [Masayaki, 2007] for EPDM and at 140/70 C and 5 kGy/h / 3 kGy/h. The test showed synergism for the degradation with an increase in rate when the material was subject to both factors. According to Kuriyama et al [Kuriyama, 1979], the value of E_a (activation energy) for nonirradiated samples is typical of thermo-oxidative degradation process, however for irradiated samples the values of E_a are more typical of oxygen diffusion in polymers. In the referred tests the oxygen penetration of the cross-linked polyethylene was limited to the surface (~0.6 mm at dose rate of 0.1 kGy/h). Synergy effects during aging treatments are not included in regulatory instructors [Häkkä-Rönholm, 2004] and evaluation of this effect is further clarified with

literature survey and experimental work. Whether these synergy effects should be noticed when creating acceptance criteria for different components is determined.

Due to the morphology of the material, distribution of antioxidants, oxygen availability etc. ageing is a heterogeneous process. Diffusion of oxygen is often a rate determining step and surface degradation is observed in several studies. Decrease in mechanical properties such as elongation at break is often observed as a consequence of surface degradation [Wüdrich, 1985]. Most incidents caused by failure of polymeric materials subject to radiation are related to elastomeric seals and electric cables. Even though many studies show a synergy between heat and radiation most of the failures are caused by thermo oxidative ageing rather than radiation ageing [Kotthoff, 1994]. However, it is still unclear how the diffusion of oxygen effect to the mechanical properties of polymers in NPP environments. There is not clear opinion how much the surface defects induced during heterogeneous oxidation decrease mechanical properties compared to homogeneously oxidated material.

1.2 Objectives and expected results

This project consists of three main objectives which include improving condition monitoring of polymeric components used inside containments, providing ageing data which is used in evaluation of acceptance criteria and providing tools for robust lifetime management for these components. To achieve these objectives three different work packages have been created which study polymer ageing in different perspectives.

- WP 1 focusing on method development of condition monitoring and implementation at NPPs
- WP 2 is a pre study to map how the closed down plant Barsebäck can be used to verify the method developed in WP1
- WP3 focusing on polymer ageing mechanism and effects inside the NPP containment

These work packages are based on issues recognized during feasibility studies on acceptance criteria polymer components conducted in Finland and Sweden [2015:157].

The objective of work package 1 is to identify the acceptance criteria for the function of the polymeric component. This includes

- Development of test methods
- Performing experimental tests to validate the method
- Development of a theoretical model that can be used to calculate different geometries
- Deployment of the results into the daily operations at the NPPs.

The polymeric component that will be tested is an o-ring and there will be 3 different materials tested EPDM, Nitrile and Viton. The implementation will be done in close collaboration with the interest group.

The objective in work package 2 is to fulfil a pre-study for polymeric materials from Barsebäck that have undergone ageing for many years. This will include: Identify the polymeric components that can be available to study, the amount of data that exists on these today and the access of these at Barsebäck. The pre study will also include a work shop to present and discuss the results.

Studying the combined effects of radiation and heat in work package 3 is performed in the form of i) a literature study, and ii) experimental work. In the literature study, the atomistic scale mechanisms governing the combined effects are identified in order to provide relevant scenarios for atomistic scale simulations, e.g. reactive molecular dynamics simulations. It is envisaged that such simulations could ultimately be used in lifetime predictions of polymeric components. In the experimental section, the amount of degradation caused by the synergy is determined with a test matrix that compares the synergy induced degradation to radiation and thermal degradation. The effect of oxidation depth is analysed with different methods in order to determine detrimental effects of surface oxidation and bulk oxidation. This section provides also data of material behaviour during a service failure and the same data is needed when the lifetime prediction model is developed.

Dose rate effect studies aim to clarify whether lower dose rates tend to cause significant amounts of degradation to polymer components compared to high dose rates during severe accident, service failure and normal service life scenarios. Since irradiation treatments with low dose rates are time consuming and costly, more theoretical approach is adopted as different extrapolation methods are studied. Extrapolation methods are evaluated by their applicability and based on the evaluation one suitable method is used in evaluation of dose rate effect. During

future work (2017) experimental data can be produced and used in evaluation of dose rate effect in relatively high dose rates (ca. >0.1 kGy/h) and same results can be used as input data for the chosen extrapolation method in order to provide data on lower dose rates.

1.3 Exploitation of the results

Challenges in the polymeric materials, such as cable materials, O-rings, joint sealants and linings are recognized in the current power plants as well as in the new plants. In this project, strong international co-operation will be carried out for increasing new information and expertise in the Nordic countries. The ultimate end users of the project results are the power plant operators as well as regulators and polymer manufactures, which are involved with assessing the integrity of the components that may be subjected to ageing degradation by irradiation and heat.

Understanding the combined effects of radiation and heat is needed when evaluating the lifetime of polymeric components during normal service life or Design Based Accident (DBA) situations. In order to evaluate acceptance criteria for new components, it is vital to understand the effects of these environmental stressors. Atomic scale simulation methods can be used to complement experimental observations. Understanding these atomistic scale degradation processes provides also a possibility to create a model that can be used when simulating ageing phenomena induced by radiation and heat. This would provide a very useful tool in the lifetime management of polymeric components. However, developing such modelling tool can be thought as a two-stage process which is implemented during this project. The first stage would be assessing the feasibility of atomistic scale modelling of thermal & radiation ageing. Whether the first stage is successful, the second stage would include development of such a modelling framework.

Regarding the dose rate effect, it has been recognized to be detrimental for some polymer materials. In order to recognize the detrimental effects of the dose rate effect during normal service life and DBA a series of experimental data and a proper modelling tool is provided as a result of this project. Thus extrapolation of the degradation caused by the dose rate effect during normal service life and DBA would be possible. This would clarify the risks involved to this phenomenon and provide information during lifetime management of these kinds of components.

Furthermore the results gained from the project will allow regulators, power plant operators and polymer manufacturers to work with polymeric materials with greater knowledge concerning ageing phenomena and acceptance criterias. This will allow better monitoring of polymeric materials, life time prediction and to make sure the component is replaced at the correct time. This will also help estimating the status of a component before and during accident conditions. The project contains tasks for implementation in the work packages which means that after the implementation phase it is estimated that the knowledge, test method or other result can be used by the regulator, power plant operator and polymer manufacturer.

In order to ensure industry/authority relevance, an interest group will be formed and representatives from NPPs and authorities will be invited to take an active part in the project. The group will participate with their knowledge on the needs of the end users and they will also be an important communication channel to the industry and the authorities. As a for instance during the deployment phase in WP1 the group will act as the end users of the developed method.

The results gained from the project will also strengthen the regulatory authorities competence concerning ageing phenomena and acceptance criteria's of polymeric materials in their role as a supervisory authority. It will allow improved monitoring of polymeric materials, life time prediction and to make sure that components are replaced at the correct time. This will also help estimating the status of a component before and during accident conditions.

The direction of the proposed method development is relevant for ageing control which is essential for long term operation of nuclear power plants. It strengthens the competence within the regulatory authorities activity area. Openly published scientific-based results will increase the knowledge base with regards to ageing of polymeric materials in environments with ionizing radiation. Given the active involvement of the nuclear power plants, the results will also be implemented in the daily operation of the nuclear power plants.

Table 1 Summary of overall project objectives and results

Objective	Procedure	Result	End users	Finish date
Condition Monitoring method development based on functional demands	Study ageing effects on functional demands of o-rings	Acceptance criteria for o-rings	NPP operators, regulator	2018
Development of FE-model for o-rings	Use of experimental data in model parameter calculations	Modelling tool for o-ring performance	NPP operators	2018
Study materials available from Barsebäck	Identify available components for study and gather detailed data on these components	Data set that can be further used when ageing during normal service life is evaluated	NPP operators, regulator, component manufacturers	2016
Evaluate the most important stressors and ageing mechanism governing the ageing during normal use and different accident scenarios	Gather data from Barsebäck and provide experimental data to evaluate dose rate effect and synergistic ageing effects	Data on importance of different ageing stressors	NPP operators, regulator	2018
Development of reactive molecular dynamics model	Study atomistic scale processes induced by combined effects of radiation and heat	Modelling tool that can be used in lifetime management and prediction of polymer components	NPP operators, regulator, component manufacturers	2018

1.4 Appropriateness of the project to SAFIR2018 programme

The SAFIR2018 programme emphasises new openings improving knowledge on NPP component ageing and lifetime management. In particular the framework plan cites irradiation resistance of cable materials as a new important field of research, and irradiation-induced ageing of organic materials is mentioned in the framework plan. The project studies the influences of irradiation and heat on a number of different polymer materials in the cases of design basis accidents and normal service together with research institutes and polymer manufacturers, making it a cross-cutting project benefitting from the specific backgrounds of the experts in the Nordic countries. The project enables access to important research areas in the nuclear field that are less known in Finland e.g. study polymer components and ageing environment at Barsebäck nuclear power plant under decommissioning.

The ageing of polymeric materials has been studied in some content but the simultaneous effect of radiation and heat is a less commonly studied topic. Typically many polymeric materials are used in locations / components where they can be replaced. However, according to radiation safety principle ALARA, all reasonable methods must be employed for minimizing radiation doses to NPP personnel. Thus this safety principle supports strongly

for using ageing resistant materials in order to avoid unnecessary radiation doses on personnel. Their role in the improvement of reliability and safety must be taken into account by research since the importance of defining the acceptance criteria for polymeric components has been pointed out lately [2015:157]. The acceptance criteria should be based on functional demands since it is the first step in the process of lifetime estimations of existing or new materials.

Strong international cooperation between VTT, SP Technical Research Institute of Sweden, the NPPs and polymer manufacturers are foreseen within this project concerning the combined effects of irradiation and thermal ageing on polymeric materials. In addition, cooperation with another SAFIR-project Fire Risk Evaluation and Defence-in-depth (FIRED) is established. This cooperation will first include common ageing treatments of samples. This project supports the strategy of SAFIR2018 and brings improved capabilities for testing of different types of polymeric material groups under irradiation. The work will start with o-rings in the first year but in the future for instance joint sealants and lining material are of interest. This project also features the adaptation of diverse techniques in testing and characterisation for use with irradiated polymeric materials, including the compression set, hardness, stress relaxation, Differential Scanning Calorimeter (DSC), Fourier Transform Infrared Spectroscopy (FTIR) and tensile testing as well as different modelling tools. Throughout the work packages synergism between experimental work and modelling can be seen. This will enhance the quality of scientific publications and provide better solutions to practice.

1.5 Education of experts

Staff from the NPPs will take an active role in the project, both junior and senior personnel will be invited to participate. The knowledge gained from the work packages will give the NPP experts better knowledge in the degradation of polymeric components. The knowledge concerning degradation of system components includes components used in safety classified equipments/functions. The possibility in setting acceptance criteria for safety classified components gives the plant a better possibility of having better control of the equipment through the intended lifetime including setting the requirements for new components.

The intention of the increased knowledge in regards to acceptance criterias is to be able to better estimate when a component needs to be replaced, know what functional/material requirements to set when purchasing, and to avoid the component to fail earlier than expected. Furthermore a better understanding on how low to high dose rate will effect polymers will be gained.

A master thesis is planned to be done together with Chalmers University of Technology Nuclear engineering and polymer technology department. The thesis work will be included in work package 1 lead by SP.

Two work shops are planned together with the nuclear power plants and safety authority, one at SP and one at VTT. The purpose of the work shops is to present the most recent results from the project including presentation for next steps. This will allow the end users to give their view of the ongoing work and the possibility to affect the way forward. The work shops are scheduled to be held in autumn 2016 and in spring 2017.

2. Work plan

Table 2. The overall plan is shown in the below Gantt chart.

Task	2016				2017				2018				Comments
	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	
T1.1													1 component, 3 polymers
T1.2													
T1.3													
T2.1													
T3.1													
T3.2													
T3.3													
T3.4													

2.1 Work package 1 (WP1) Development of condition monitoring methods for polymeric components including low dose rate radiation exposure.

The aim of this work package is to identify the acceptance criteria for the function of the polymeric component. This includes:

- Develop robust test methods that can be used by the power plants for condition monitoring through a material property. The material property will be correlated to the function of the component.
- Performing experimental tests to validate the method
- Development of a theoretical model that can be used to calculate acceptance criteria for components with different geometries
- Deployment of the results into the daily operations at the NPPs.

During a feasibility study [2015:157] acceptance criteria for functional properties for different polymers in system components was studied. The components were selected based on interviews with the five Nordic nuclear power plants. Furthermore, the need to study degradation using low dose rates was identified since previous work described in literature mainly focuses on using a high dose rate to achieve the life time dose during a short period of time. This may cause a different degradation, compared to that obtained with a long exposure at a low dose rate.

The study will focus on accelerated ageing through heat and radiation to selected components starting with o-rings. Both Teflon seal and reinforced EPDM are of interest and are valid components to be tested in the future but left out of this work package. The test will be done using one or several specific properties, compression set being one of them, and correlate this to a functional test for tightness of an o-ring. By doing so a correlation between compression set and tightness can be achieved. To be able to better compare the effect the radiation has on degradation, a parallel test in heat will be done.

The o-rings will be mounted in a compression set test rig as well as in tube connectors during exposure. Compression set after certain exposure times will be measured and the pipe connections will be tested by mounting them in the SP hose testing equipment. With this test the sealing performance will be measured as water pressure without leakage.

By testing the correlation between compression set and tightness of an o-ring an understanding of the function (tightness) based on a material property (compression set) can be made. The aim is to be able to use this to set acceptance criteria for an o-ring using compression set as a property.

Other evaluation methods are tensile testing, thermal analysis (DSC, Thermo Gravimetric Analysis (TGA)) and if necessary chemical analyses such as Gas-Chromatography Mass-Spectrometry (GC-MS) and elemental analysis. DSC has been identified as a valuable method since antioxidant content is indirectly measured and indicates the residual service life of a product. Another benefit is that only small samples (5 mg) are needed. Since the knowledge about the material composition is often limited it is valuable to analyse composition (amount of plasticizer and filler), type of antioxidant and vulcanization system.

The following materials are to be included in WP1:

- O-rings (EPDM) Temperature* 100, 120, 140 °C vulcanized and subject to low radiation.
- O-rings (Nitrile) Temperature* 60, 80, 100 °C, vulcanized and subject to low radiation.
- O-rings (Silicone or Viton), Temperature* to be decided depending on the chosen polymer. 3 different temperatures to be used, vulcanized and subject to low radiation.

*Temperature may vary slightly due to the formulation of the polymer. The temperature for the test will be set once the polymeric component has been provided.

The dose rate radiation exposure is set to 21 Gy/h during a total of 28 days. This gives a total dose of 14 000 Gy to be compared to a component during normal operation subjected to a high radiation environment of 0,1 Gy/h for a little bit over 16 years. In future work (2017 and onwards) ageing parameters can be chosen in a way that functional properties of these components can be evaluated in situations like service failures and severe accidents in order to provide acceptance criteria if the component has designed function during these situations.

To get an idea of how much of the polymeric materials used in the NPPs that would be covered using the results from this project, actual numbers from Ringhals o-rings (EPDM, Nitrile, Viton) indicate a coverage in order of 85-90%. If choosing silicone instead of Viton the coverage will decrease but still is approximately 65-70% at Ringhals.

The time line below followed by a test matrix, show the sequence of tests to be completed for one component including radiation. During the radiation treatment the heat in the chamber will be approximately 50 °C which is close to the user temperature. The test matrix shows 12 samples running at three different temperatures, with and without radiation and at two different geometries (cord diameter of the ring). Only the EPDM o-ring will be tested using two cord diameters. This is estimated to be enough for the modelling but more tests could be proposed as future work if determined to be of interest. There are 5 points for evaluation including starting point. The time between evaluation is decreased at the later stage of the test since it is the region where the acceptance criteria or end of life will be found. It is estimated that a minimum of 80% compression set is needed before the function will fail. If 80% is not met during the 6,5 month test the evaluation will help guide in how much longer heat treatment is needed to reach this area.

Figure 1: Time line for 1 test including radiation and heat treatment. The temperature and radiation rate can be found in table 3. Dates are exemplified with start January 1st 2016, actual start see Deliverable chapter.

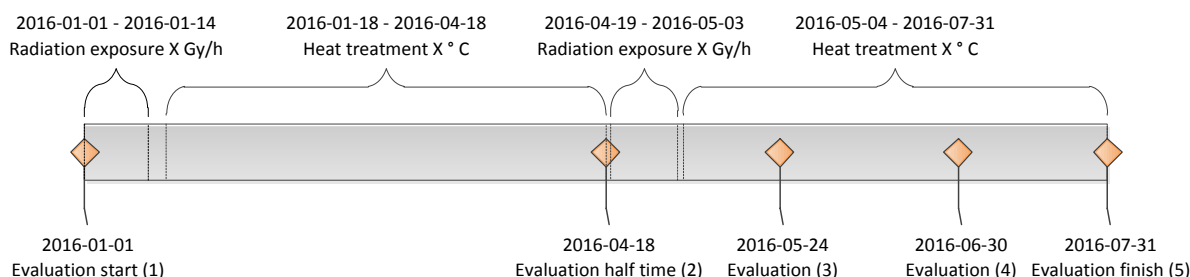


Table 3. Test matrix for WP1 exemplified for EPDM. Geometry stands for diameter of o-ring.

Sample	Temperature (C)	Radiation (Gy/h)	Cord diameter(mm)
1	100	0	3
2	100	0	6
3	100	21	3
4	100	21	6
5	120	0	3
6	120	0	6
7	120	21	3
8	120	21	6
9	140	0	3
10	140	0	6
11	140	21	3
12	140	21	6

Since there are many different sizes and shapes of o-rings used at a nuclear power plant, a theoretical model using the data identified through tests in WP1 will be created and verified. The model can be used to estimate acceptance criteria for a larger number of components with different geometries during a shorter time, than actual tests.

The result may be possible to use in technical documents setting requirements for polymeric components for the nuclear power plants. This can be used for existing components in the NPP or when purchasing new components (A fingerprint through for instance FTIR or DCS should be added). Depending on the components identified in WP2 Barsebäck a comparison can be made to the accelerated test in WP1.

The results will be presented at a seminar and in a report. Depending on the findings, a scientific article will be written and presented.

Partners and person months allocated to WP1 to be given in the table.

Table 4. Partners in WP1.

Partners in WP1	Person months
SP	2,7
VTT	0,55
James Walker	0,1
Nordic NPPs*	0,2

*in-kind contribution

Results from the joint SSM/Swedish NPP-funded project "Long term performance of polymeric materials in Nuclear power plants" will be taken into account in the work package.

2.1.1 Task 1 (T1.1) Ageing and functional testing

The goal of Task 1 is to develop and run the test method to be used when exposing the sample to ionized radiation and heat. This includes treatment in a chamber for low dose radiation, decision on what material properties to test with and what type of equipment's to use. The test rig will need a suitable tube connector manufactured for the functional test. Depending on the polymer reaction products after irradiation and heat treatment can be used as well to study the changes in the material.

The test will run in a repetitive cycle with functional and property tests, exposure to radiation and exposure to heat. See figure 1 with the timeline showing where the tests are done and the time of each cycle. An estimation of the acceptance criteria for when the function of the component is not satisfactory will be made and correlated to the compression set (if needed more material properties will be used).

2.1.2 Task 2 (T1.2) Implementation for the industry

The goal of Task 2 is to investigate the implementation of suitable methods in the daily operation at any of the nuclear power plants in the Nordic countries. The implementation phase will study the possibility for the plants to perform the needed tests on their own or if a separate lab needs to be consulted. Assistance in how a plant can set material properties for new components will also be discussed and presented. This task will be performed in close collaboration with the interest group and aims to improve lifetime management of polymer composites.

The goal of Task 1.2 is to investigate the implementation of suitable methods (including compression set and hardness measurement) in detecting defective o-rings at working site and thus create a procedure that can be used in detection of faulty o-rings before their installation. The method should be simple and reliable so it can be adapted in everyday use at NPPs. This task will be conducted in close collaboration with T1.1 as well as TVO material experts and during year 2016 it consists of the following steps:

1. Determine the working conditions and o-ring properties
2. Evaluate whether hardness and compression set can be correlated to proper functionality of the o-ring
3. Evaluate how hardness and compression set measurements can be implemented at the plant

As a result the use of these methods in detecting material defects between different patches during new o-ring installations at NPPs can be evaluated.

In future work (2017) the chosen method is further developed and tested in laboratory scale tests in order to provide a procedure that is simple and reliable as well as is able to be introduced to be used at NPPs. Also an acceptance criterion for tested component is determined.

2.1.3 Task 3 (T1.3) Modelling

The goal of Task 3 is to develop finite element (FE) models of the o-ring seals, which can be used to predict the leak tightness of different sizes and shapes of o-rings that have been exposed to heat and radiation. The model will use the compression set data acquired in Task 1 to tune the parameters of the material models used in the calculations. Additional testing of unaged material will provide the remainder of the needed parameters for the calculation model. Tests of the unaged material will consist of uniaxial tension and compression, and also a pure shear strain test. Validation simulations will be performed using the two different geometries that have been leak tested in Task 1. The calculation models will give the plants a broader knowledge about the functionality of the o-rings and tools which can be used to compare the performance of a wide range of components.

SP Structural and Solid Mechanics has performed a research project on leak tightness in large polyethylene pipe joints with rubber gaskets [Jacobsson, 2011 Part 1 and 2]. The project included modelling leakage with finite elements using creep models in the polyethylene and hyperelastic modelling of the rubber, followed by full scale leak tests on the joint. The project is now followed by a new ongoing research project. This current project features material testing to calibrate elastic-plastic creep modelling of the polyethylene and hyperelastic nonlinear viscoelastic modelling of the rubber used in steel reinforced o-rings. The material models are used in a full model of the joint with fluid penetration contacts to simulate leakage and accompanied by full scale leak tests.

Previous research also includes evaluation of the fatigue life of rubber components, where FE-modelling in combination with material testing was compared with other fatigue life evaluation methods (fracture mechanical approach and cracking energy density approach). Fatigue testing was then performed on the actual rubber components compared with the results of the fatigue life evaluation methods. Other, previous or ongoing, research includes determination of mechanical properties of rubber and aging of seals in water pipes.

When determining mechanical properties of rubber, a set of tests has been performed in the laboratory at the assumed future operating temperature. The set consists of uniaxial tension and compression, and also pure shear strain test and volumetric compression.

A general investigation was done on water tightness of rubber seals for the Swedish Water and Waste Water industry [Thörnblom, 2014]. It was concluded that the required ageing tests in the applicable standards are far from sufficient. The tests performed cannot be useful for forecasting a life of about 100 years, which lies in the area of the networks administrators need. An extensive study on ageing of seals (EPDM, NBR, TPE) is proposed to the Swedish Water & Wastewater industry.

2.1.4 Proposed future work

There are four other identified components from the feasibility study that was concluded to be of interest for the development of acceptance criteria. These are Teflon seal, reinforced EPDM, joint sealants and lining materials. These components were highlighted during the interviews with the Nordic nuclear power plant representatives. However, it was decided to exclude these from the first work package and perhaps include them in a future follow up project, based on the results provided in WP1.

2.2 Work package 2 (WP2) Barsebäck

The aim of this work package is to study materials from Barsebäck that have undergone ageing during operation for many years. This includes a pre-study to identify the polymeric components that can be available to study, analysis of the degradation of the selected materials and a workshop to present and discuss the results.

It would be very valuable to compare artificially aged materials studied in WP1 with actual aged materials that have been in operation in a nuclear power plant. Many materials that are of interest can be obtained from the Nordic NPPs that are in operation, but this is not possible for some of the safety related polymeric components. An alternative option could be to obtain materials and components from the closed down NPP Barsebäck.

In this WP a pre-study will be performed in order to identify polymeric components that can be available from Barsebäck, the amount of data that can be found on these components and information on if it is possible to gain access to these and possibly to take them off site.

The selection of components will be based on polymeric components included in WP1 to be able to correlate them to the accelerated tests. For the selection of materials at Barsebäck, it will be important to consider for example if the type of material could be available at sites in operation today, and/or if the materials have been exposed to both high temperature and radiation. The latter is significant to minimize the effect from storage after closure of the plant. It is also needed to take into consideration that since the reactor containment in a BWR is nitrogen filled during operation, i.e. that the 16 and 10 years period after termination of power operation, have exposed materials inside the containment to a much longer air period than during 30 years of operation.

The pre-study will also include finding the amount of data available on the components, and existing data for specific components will be used to show how a material has degraded. It is however likely that the possibility to find information will be limited. Chemical analysis will probably be needed to identify and characterize the materials further.

The access to materials will rather be limited to the radiological clearance than to availability of the material at the sites. Therefore the pre-study will also include the evaluation of the process of taking the materials off from the site, and the option to perform any of the analysis on-site.

The identified components in the pre-study will be used to correlate the results from other proposed WPs with materials that have undergone ageing during operation for many years; this will yield valuable information on the ageing phenomena. They can also be used to investigate if the degradation is on the surface or in the bulk of the material. Presentation and discussion of the results is included in the work package.

Partners and person months allocated to WP2 to be given in the table.

Table 5. Partners in WP2.

Partners in WP2	Person months
SP	0,3
Barsebäck Kraft*	0,2

*in-kind contribution

2.2.1 Task 1 (T2.1) Pre-study

The goal of task 1 is to fulfil the pre-study. This will include: Identify the polymeric components that can be available to study, the amount of data that exists on these today and the access of these at Barsebäck. The pre study will also include a work shop to present and discuss the results.

2.3 Work package 3 (WP3) Polymer ageing mechanisms and effects inside NPP containments

The ageing environment for polymers within the containments of NPPs is rather complicated. During normal service of NPP the temperature within the containment can be tens of degrees beyond room temperature. Also radiation levels can vary from less than mGy/h to Gy/h. [Penttilä, 2016] So called hot spots with elevated temperature and higher dose rates are located in the vicinity of steam generator tubing or in process valves. Thus polymer components are subjected to various stressors such as heat, radiation and moisture. In the both cases of thermal and radiation ageing, oxidation is considered to be the most common and dominating degradation mechanism. [Bartonicek & al] Oxidation of polymers is due to polymer radicals that are formed by thermal or radiation energy. These radicals react with oxygen forming peroxy radical which further reacts with the polymer chain forming hydroperoxide and polymer radicals. Hydroperoxide thermally decomposes to species that cause chain scission. Under radiation oxygen diffusion can be thus detrimental for polymers in room temperature and it is accelerated by increased temperature. The further complicating factor is diffusion-limited oxidation which has an effect to the oxidation behaviour of polymers. [Celina & al]

In Work Package 3 the effects of radiation and heat on polymer degradation are evaluated. There is on going work [Penttilä, 2016] that defines relevant materials to be studied. It is known that most of the polymer degradation during normal usage of an NPP is due to thermo oxidative ageing but the effect of radiation ageing cannot be neglected. Especially during Design Based Accident (DBA) and severe accident scenarios the effect of radiation becomes more significant. A typical testing procedure for normal service conditions and polymeric materials includes separate irradiation and heat treatments. During irradiation conservative dose rates and accumulated doses are used. Such practice does not reflect the actual service situation because it does not take into account the synergistic effects of radiation and heat. To evaluate the testing methods for polymers used inside containments and acceptance criteria for these components, a fundamental understanding of the oxidation processes induced by radiation and thermal energies is needed. To achieve such knowledge wider literature survey is necessary on the synergistic effects of radiation and heat. The literature survey includes also a survey on modelling methods that could possibly be used in predictive modelling of radiation and thermal degradation of polymeric materials. Especially the modelling of thermo oxidative ageing would create a basis for future work creating a model based on reactive molecular dynamics simulations. This kind of model is needed in order to evaluate the synergic effects of thermal and radiation ageing.

The synergy effects of radiation and heat are also determined experimentally. One of the key materials used in Finnish NPPs are studied (key materials are to be identified within [Penttilä 2016]) in order determine the amount of degradation caused by thermo oxidative ageing, radiation ageing and their combined effect during a service failure. Samples are exposed to thermal and radiation ageing separately and simultaneously and samples are analysed by tensile testing, hardness measurements and DSC. Thus it can be evaluated whether the thermal ageing is clearly dominant compared to radiation ageing of the component and should the effects of radiation ageing be taken into account when components are tested and acceptance criteria is evaluated.

It is well known that oxidation of a polymeric material occurs at the material surface and the heterogeneous oxidation is affected both material thickness and temperature. The oxygen diffusion follows Ficks law and also the Arrhenius equation i.e the diffusion increases upon temperature increase. Degradation initiated by radiation may

influence oxygen diffusion deeper into the bulk material and hence accelerate oxidation. Also the role of surface defects induced by surface oxidation in degradation of material properties needs to be clarified.

The dose rate effect has been recongized to be detrimental to some polymeric materials [Gillen 1981, Placek 2003] and as a part of this WP, the goal is to determine the significance of dose rate effect on polymers within the containment of NPP. The dose rate effect is related to the phenomenon where lower dose rates cause more degradation in the polymer properties than higher dose rates with the same total absorbed dose [Reynolds, IAEA-TECDOC551]. The diffusion of oxygen is closely related to this process since the diffusion of oxygen defines the depth of degradation within the polymer. With high dose rates all oxygen is consumed in the vicinity of the polymer surface and hence the damaged polymer structure is located near the surface and not in the bulk. With low dose rates oxygen has more time to diffuse in to the bulk and thus cause material degradation throughout the polymer.

Since the radiation levels during normal use in NPP are relative low (less than ~ 1 Gy/h) compared to the dose rates (10 kGy/h) defined in regulator instructors [Häkkä-Rönnhom, 2004], a predictive model that extrapolates the effects on lower dose rates is required. These kinds of models are power law extrapolation method and superposition methods. A material, that is used within containments and is suitable (that is susceptible to radiation in order to minimize the radiation times) for experimental procedure, is chosen as test material. To find a suitable model for this case, different models are evaluated and most suitable is chosen to be used in extrapolation. Since these models require experimental data on radiation and/or thermal ageing, such data is produced within this WP during 2017.

Partners and person months allocated to WP3 to be given in the table.

Table 6. Partners in WP3.

Partners in WP3	Person months
VTT	3,95
SP	1,0
Nordic NPPs*	0,2

*in-kind contribution

2.3.1 Task 1 (T3.1) Modelling tools for the synergistic effects of radiation and heat

The goal is to complete a literature survey on the synergistic effects of thermal and radiation ageing in polymeric materials. The survey will look for proposed mechanisms behind the synergism, compile an overview of related experimental work and identify modelling methods that can be used for studying the phenomenon. Both of the mentioned ageing mechanisms are based on atomistic scale processes, and it is already known that radiation ageing can be modelled at the atomistic scale using reactive molecular dynamics simulations. However, concurrent modelling that would also consider thermo-oxidative ageing seems less common. The literature study will form a basis for future modelling efforts in identifying the relevant physical and chemical phenomena, as well as other fundamental input parameters for a proposed reactive molecular dynamics model.

2.3.2 Task 2 (T3.2) Polymer ageing during service failure

The goal is to experimentally determine which ageing mechanism dominates during a service failure and what kind of synergy effects can be seen during them. For this purpose EPDM and Lipalon cables are tested in conditions similar to LOCA in order to perform a series of laboratory tests to clarify the effects of radiation, heat and combination of these two on the testing materials. Artificially aged samples are tested with tensile testing, hardness measurements and DSC. This will provide fundamental data when evaluating acceptance criteria for polymeric materials used within the containment.

2.3.3 Task 3 (T3.3) Synergy effects between radiation and heat and oxidation depth

This task focuses on the surface oxidation in comparison to the bulk oxidation and the effect on overall material properties. Samples exposed to the combined and separate exposure programmes in WP 1 and 3 will be analysed in this task in order to evaluate any synergy effects caused by surface oxidation. In total of three different test methods for determining oxidation profiles on samples are used. The plan is to analyse the surface as well as bulk material at different distances from the surface to follow the oxidation depths in materials exposed to the heat and radiation schedule described above. The levels of degradation products such as carbonyls and ketones are of interest. Two differently doped EPDM materials, representing good and poor radiation resistance properties are examined. Analyses will be performed after radiation, radiation followed by ageing and ageing only.

FTIR (Fourier Transform Infrared spectroscopy) equipped with an ATR (Attenuated Total Reflection) device allow analyses of the oxidation products in a polymer on the surface with depth of nanometer scale. This technique has previously been used on polymeric medical prosthesis in order to study degradation depths caused by irradiation sterilization. Most rubber materials contain high loads of carbon black filler and this causes total absorption of the infrared light. Therefore non-filled model rubber materials need to be used for this study. ToF-SIMS (Time of Flight Secondary Ion mass spectrometry) is a mass spectrometry method allowing very surface sensitive measurements, actually only one surface atom layer. This technique would allow analysis also on black rubber materials actually used in the power plants. DSC (Differential Scanning Calorimetry) measurements are performed on polymers that are induced to radiation by only one surface. Polymers are cut after radiation to obtain samples from different distances from the surface and analysed by DSC. Thus an oxidation profile is created. The proposed analyses only need small samples, a few milligrams.

2.3.4 Task 4 (T3.4) Evaluation of damage caused by dose rate effect to polymer components used within containments

Since the experimental studying of dose rate effect is costly due to long radiation treatment times, a theoretical approach to estimate magnitude of this phenomenon needs to be found. There is known methods that are designed for this purpose in the literature [IEC 61244-1, IEC 61244-2]. Typically these methods have limitations related to polymer type or experimental data. During this task these methods are gathered and evaluated in order to identify the most robust method that can be used in evaluation of dose rate effect. Suitable materials for dose rate effect studies are decided based on [Penttilä, 2016]. As proper method and relevant material has been recognized, during 2017 experimental data can be produced that is needed for the method to predict the behaviour of the material under low dose rates.

3. Deliverables 2016

Table 7. List of all deliverables planned for the project year 2016.

Deliverable number	Deliverable name Short description of the deliverable if necessary.	Indicative person months	Deadline date
D1.1	Development of condition monitoring technique for O-rings used in NPP applications (SP-research report)	2, 355 ¹ +0, 553 ²	01/2017
D2.1	Identification of available polymers and their data from Barsebäck (including one work shop and SP-research report)	0,7 ¹	09/2016
D3.1	Synergistic effects of radiation and heat and fundamental aspects on their modelling (VTT-research report)	1,0 ²	10/2016
D3.2	Synergistic effects of radiation and heat on EPDM and CSPE (VTT-research report)	1,4 ²	12/2016
D3.3	Oxidation of EPDM: oxidation depth measurements and effects on material properties (VTT-research report)	1,0 ¹ +0,85 ²	12/2016
D3.4	Methods used in evaluating severity of dose rate effect on low dose rates (VTT-research report)	0,7 ²	12/2016
Total pm		8,5	¹ SP person months ² VTT person months

4. Project organisation

The project is implemented as cooperation of VTT and SP. Konsta Sipilä (VTT) will act as the responsible person from VTT and for the overall project while Marcus Granlund (SP) will act as the responsible person from SP. Together with the team from VTT and SP there will be an interest group from the NPPs and other interested parties. The group will help to ensure industry/authority relevance as they are invited to take an active part in the project.

The project will report to a reference group appointed by the SAFIR2018 Management board.

Table 8. The project organisation at VTT and SP.

Name	Title	Organisation	Participates in tasks	Estimated person months (2016)
Anna Jansson	Senior scientist	SP	T1.1, T1.2, T1.3, T3.3	1,9
Jessica Eriksson	Research scientist	SP	T2.1	0,6
Marcus Granlund	Research scientist	SP (Responsible SP)	T1.1, T1.2, T1.3, T2.1, T3	1,5
Daniel Vennetti	Research scientist	SP	T1.3	0
Jan Henrik Sällström	Research scientist	SP	T1.3	0
Harri Joki	Senior scientist	VTT	T3.2, T3.3, T3.4	1,2
Antti Paajanen	Research scientist	VTT	T3.1	0,9
Tiina Lavonen	Research scientist	VTT	T1.1	0,3
Sami Penttilä	Research scientist	VTT	T3.2, T3.3, T3.4	0,5
Konsta Sipilä	Research scientist	VTT (Responsible VTT and overall project)	T1, T2, T3	1,6
Total				8,5

An interest group will work with the project and be invited to participate during the planned work shops. The group consists of staff from different part of the industry, see table below.

Table 9. The Interest group.

Name	Organisation
Monika Adtsen	Energiforsk AB
Eric Jansson	EON, OKG
Lauri Rintala	Fennovoima
Juha Rinta-Seppälä	Fennovoima
Ritva Korhonen	Fortum
Kristiina Söderholm	Fortum
Jukka Sovijärvi	STUK
Pekka Välikangas	STUK
Liisa Heikinheimo	TVO
Timo Kukkola	TVO
Henrik Widestrand	Vattenfall
Anneli Jansson	Vattenfall, Forsmark
Stejpan Jagunic	Vattenfall, Ringhals
TBD	Barsebäck
John Rogers	James Walker

5. Risk management

Table 10. Risk management plan. To be updated during the project on a periodic basis.

Risk	Probability of occurrence	Potential impact on project success	Mitigation Plan
Significant changes in the research plan.	Low	Medium	<ul style="list-style-type: none"> - Study the impact of the changes on schedules and results - Implement changes, if the impact is high
Poor data quality	Low	High	<ul style="list-style-type: none"> - Only use records which have good quality basic data sets - Only use qualified staff for testing
Costs could rise significantly during the time of the project	Low	High	<ul style="list-style-type: none"> - Monitor costs on a periodic basis - Using other funds.
Loss of key researcher (unable to complete key tasks)	Low	High	<ul style="list-style-type: none"> - Identify alternative resources in case of unexpected absence. - Ensure complete records of work are available at any point
New cooperation between SP and VTT	Low	Medium	<ul style="list-style-type: none"> - Arrange a project start up meeting in the beginning of the project. - Include periodic project meetings to follow progress and cost

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