



## DEVELOPMENT OF AN EXPERT SYSTEM FOR ANALYSIS OF PLUTONIUM PROCESSING OPERATIONS

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**Key words:** simulations, planning, exposure

### ABSTRACT

At Los Alamos National Laboratory (LANL) an expert system has been developed for the analysis and assessment of plutonium processing operations. This system is based upon an object-oriented simulation environment specifically developed for the needs of nuclear material processing. The simulation environment, called the "Process Modeling System" (ProMoS), contains a library of over 250 plutonium-based unit process operations ranging from analytical chemistry, oxide operations, recycle and recovery, waste management, and component fabrication.

Fundamentally, ProMoS views the world as the capability to move resources such as materials, tools, and people, through containers, such as locations (a room or facility), workcenters (such as a glovebox), and bulk containers. Central to the ProMoS construct is the concept of material-balance. Since it was designed for nuclear operations, ProMoS has strengths in both part-based and bulk-material-based operations. It also has a vast array of mechanisms that provide constraints to material movement and processes. Finally, ProMoS was designed to also solve pragmatic radiation exposure problems.

Given a schedule of operations, resource pools, initial inventories, detailed part "travelers", and restricted part movement and control associated with special nuclear material operations, a detailed time history of part movement can be constructed. However, as with models of other complex systems, visualization is a key component of debugging or understanding the operations in the model. To assist users in visualizing material flow through a subsystem such as a processing area, Los Alamos has coupled the simulation engine with a visualization post processor using the PROOF® tool. This tool is used to enhance material and component flow understanding and to provide a ready means of assessment communication.

The unit process database in the system can be coupled as necessary to simulate the full suite of activities in a nuclear material facility. In particular, we show the detailed characteristics of a potential mixed-oxide fuel fabrication line at the LANL plutonium facility. Estimates of total throughput, waste generation, personnel requirements, material inventories, and accumulated radiation dose are shown.

In the framework of GRETE, the ability of various NDT techniques to monitor the effect of aging (irradiation and fatigue) will be assessed through experimental measurements to be made on aged specimens representative of materials used in the nuclear industry.

The purpose of this paper is to present the project and its programme of work. GRETE is supported by the European Commission in the framework of the programme NUCLEAR ENERGY (contract n°FIKS-CT-2000-00086).

## METHODOLOGY

For each damage mechanism, series of specimens will be prepared by the partners. These specimens will be then tested using non destructive techniques based on ultrasonics, magnetics, thermoelectricity and dynamic indent. The NDT signals will be correlated to the microstructural changes observed in the material because of aging and to the mechanical characteristics of the material. At last, possible technology transfer towards the industry will be evaluated for practical use of these techniques by the utilities.

## PARTNERS OF GRETE

A consortium of fifteen partners has been formed to achieve the work programme of GRETE. These partners are either utilities or their representatives, research centres or universities. A list is given in Table I.

Table I : partners of GRETE

Partner	Country
Electricité de France	France
Netherlands Energy Research Foundation	The Netherland
VTT Manufacturing Technology	Finland
TECNATOM s.a.	Spain
Fraunhofer Institut für Zerstörungsfreie Prüfverfahren	Germany
Joint Research Centre of the European Commission	The Netherlands
AEA Technology National NDT Centre	United Kingdom
Centro de Investigaciones Energéticas Medioambientales y Tecnológicas	Spain
Austrian Research Centers	Austria
Atomic Energy Research Institute	Hungary
University of Hannover	Germany
Paul Scherrer Institute	Switzerland
National Research Institute	Czech Republic
Siempelkamp Prüf- und Gutachter- Gesellschaft mbH	Germany
Siemens Nuclear Power	Germany

## NON DESTRUCTIVE TECHNIQUES

The non destructive techniques that will be used by each partner in the framework of this project are reported in Table II (neutron irradiation damage) and Table III (fatigue damage). A brief description of each technique is proposed here below.

Table II : NDT techniques for the characterisation of neutron irradiation damage

NDT technique	Partners
Automated ball indenter	National Research Institute
Magnetic Barkhausen Noise	Fraunhofer Institute Centro de Investigaciones Energéticas Atomic Energy Institute
Micromagnetic measurements	Fraunhofer Institute
Non Linear Harmonic Analysis of Eddy Current signals	University of Hannover
Thermopower measurements	Electricité de France Joint Research Centre

Table III : NDT techniques for the characterisation of fatigue damage

NDT technique	Partners
Magnetic Barkhausen Noise	Fraunhofer Institute Atomic Energy Institute
Micromagnetic measurements	Fraunhofer Institute
Non Linear Harmonic Analysis of Eddy Current signals	University of Hannover
Fluxgate, Giant Magnetic Resistor, Superconducting Quantum Interference Device	Fraunhofer Institute Paul Scherrer Institute Siempelkamp
Ultrasonic Scattering or Backscattering	VTT Austrian Research Centres

### - Magnetic Barkhausen Noise

The magnetic Barkhausen effect is observed as transient pulses induced across a search coil placed near or around the ferromagnetic material undergoing a change in magnetisation. These pulses can either be observed individually by counting and amplitude sorting or as a rms signal as a function of the applied magnetic field. The BE signal arises from irreversible magnetic domain wall movements as domain walls become successively pinned and jump over obstacles in the material. These obstacles are typically dislocation defects, second phases or grain boundaries and consequently the technique is particularly sensitive to the microstructure and mechanical properties of the component. The technique is also sensitive to the internal stress state because of the partial domain alignment along the maximum principal stress axis. Thus, tensile and compressive stresses usually increases and decrease the BE signal respectively.

### - Non Linear Harmonic Analysis

This technique utilises the whole magnetic hysteresis loop and the way in which it is influenced by the microstructural changes due to degradation. An oscillating

sinusoidal magnetic field is applied to the material ; this field is modified by the material which acts as a transfer function, so that a detector coil picks up a distorted signal which is analysed for amplitude and phase of different harmonics of the original signal frequency. To calibrate it, the variation of these parameters is fitted using a 'multidimensional regression analysis' to provide the best correlation with material property. Some degree of selectivity to the different mechanical properties is achieved.

### - The 3MA Analyser System

The 3MA analyser system (**M**icromagnetic, **M**ultiparameter, **M**icrostructure and Stress **A**nalysis) has been developed by the Fraunhofer Institute for Non-Destructive Testing (IzFP) at Saarbrücken in Germany. As its name implies, the instrument measures a combination of different magnetic parameters, enabling some degree of separation between variations in the stress and microstructure states. The 3MA analyser employs the techniques of magnetic Barkhausen, Coercivity (derived from Barkhausen profiles) and magnetic field frequency harmonics. The instrument is designed for use in a wide range of applications including : detection of different heat treatments, residual stresses, hardness gradients and parameters loosely related to strength and toughness.

To achieve some quantitative measurement the 3MA analyser has to be calibrated against samples containing the variations of interest. Indeed a great deal of work has been done by the researchers in investigating a large range of materials and heat treatment conditions. Recently, new approaches have been developed which concentrate on using linear multiple regression or neural network algorithms to calibrate the system for limited well defined set of specimen or component conditions. These calibrations rely on detailed mathematical variational formalism which notably does not involve any empirical or fundamental understanding of the physical principles of the magnetic techniques.

### - Physics of the Superconducting Quantum Interference Device

The **S**uperconducting **Q**uantum **I**nterference **D**evice (SQUID) is the most sensitive magnetic field sensing element known. One version of the SQUID, the thin-film DC SQUID, has two Josephson weak links interrupting a superconducting loop. The maximum supercurrent that can be passed through such a loop before a voltage develops (the critical current) is periodic in the magnetic flux passing through the loop, with period  $F_0=2 \times 10^{-15} \text{ T} \times \text{m}^2$ . Typically SQUID electronics DC-bias the device close to the superconducting critical current, apply an AC modulation bias field to the loop, and feed back on a DC bias field to keep the voltage output at the modulation frequency constant. The DC feedback field is then directly proportional to the magnetic flux through the loop.

The SQUID sensor requires a cryogenic environment, since it must be superconducting to operate.

### - Physics of the Giant Magnetoresistance

The **G**iant **M**agnetoresistance (GMR) effect is a very large change in electrical resistance that is observed in a ferromagnet/paramagnet multilayer structure when the relative orientations of the magnetic moments in alternate ferromagnetic layers

change as a function of applied field. Changes in resistance with magnetic field of up to 70% (typically up to 20%) were observed. The basis of the GMR is the dependence of the electrical resistivity of electrons in a magnetic metal on the direction of the electron spin, either parallel or antiparallel to the magnetic moment of the films (layers). Electrons which have a parallel spin undergo less scattering and therefore have a lower resistance. When the moments of the magnetic layers (e.g. NiFe) are antiparallel at low field, there are no electrons which have a low scattering rate in both magnetic layers, causing an increased resistance. At applied magnetic fields where the moments of the magnetic layers are aligned, electrons with their spins parallel to these moments pass freely through the solid, decreasing the electrical resistance. The resistance of the structure is therefore proportional to the cosine of the angle between the magnetic moments in adjacent magnetic layers. GMR magnetic field sensors have recently been evaluated for use in many applications (e.g. geophysical exploration) and found to have a noise floor of 0.1 to 1.0 nT in an unshielded, unfiltered system.

#### **- Thermoelectric Power**

A system based on Seebeck's effect has been developed by the Technical University INSA de Lyon. Laboratory measurements are performed on a thin  $40 \times 5 \times 1 \text{ mm}^3$  plate of material applied on two small copper blocks held at two different temperatures. A thermoelectrical voltage is generated by Seebeck's effect due to the thermal gradient between the blocks. The thermopower is defined as the ratio of the difference of voltage by the difference of temperature between the two blocks. EdF and INSA have developed a portable TEP system, which can be used on large components after some surface preparation. This device is used to follow the thermal aging of cast duplex stainless steel elbows.

#### **- Ultrasonic Scattering or Back-Scattering**

Ultrasonic scattering or Back-scattering are techniques that are based on the measurement of ultrasonic waves scattering back to the transducer from the microstructure of the material. The measurement can be performed based on the reflection received from the back-wall of the sample or on the back-scattered signals coming from the interior of the sample. These basic techniques can be complemented by measurement of sound velocity and center frequency of the back-wall echo. The measurements can be performed also in industrial environment with reasonable accuracy. For measurement of sound velocity special techniques based on the use of electromagnetic acoustic transducers (EMAT) are available. Under laboratory conditions the velocity of leaky surface acoustic waves can be measured precisely by using the  $V(z)$ -technique.

### **CHARACTERISATION OF NEUTRON IRRADIATION DAMAGE**

Irradiated specimens are provided by EDF, TECNATOM, the University of Hannover and the National Research Institute (see Table IV). They are made of materials representative of reactor pressure vessel (A533 cl.1, A508 cl.3 and 12Kh2NMFA steels). Samples from EDF, TECNATOM and the National Research Institute belong to the surveillance programme of industrial reactors (PWR and VVER) whereas



samples from the University of Hannover were irradiated in an experimental reactor (VVER).

Table IV : Irradiated specimens for NDT testing

Partner	Steel grade	Irradiation conditions		Fluence ( $10^{19}$ n/cm <sup>2</sup> ) [E > 1 MeV]					
				none	1.7	3.7	5.6	7.6	
ELECTRICITE DE FRANCE	A533 cl.1	PWR	290°C	none	1.7	3.7	5.6	7.6	
		PWR	290°C	none	1.7	3.0	4.7	6.6	
UNIVERSITY OF HANNOVER	A533 cl.1	VVER-2	254°C	none	5.7	9.4	10.2	-	
	A508 cl.3	VVER-2	254°C	none	5.7	9.4	10.2	-	
TECNATOM	A533 cl.1	PWR	290°C	-	0.7	2.1	5.2	-	
NATIONAL RESEARCH INSTITUTE	12Kh2NMFA	VVER- 1000	to be specified						

Irradiated samples for NDT testing are Charpy-V notch test specimens broken in the brittle domain in order to avoid local deformations. Non irradiated specimens will also be available for NDT testing.

The mechanical characteristics of these materials have already been measured in previous studies. They will be made available for the project.

The NDT measurements on irradiated specimens will be performed in the hot cell facility of the Netherlands Energy Research Foundation.

## CHARACTERISATION OF FATIGUE DAMAGE

Two grades of austenitic stainless steel will be provided by EDF (AISI 304L) and by SIEMPELKAMP (AISI 321). These steels are widely used in power generating systems for piping and support structures.

Hour glass specimens designed according to ASTM E606 will be manufactured and tested (Low Cycle Fatigue) by the partners according to the experimental matrix in Table V.

The fatigue testing will be performed at room temperature and at 300°C (in service temperature) at a frequency of one Hertz. Three usage factor have been chosen : 0.6, 0.8 and 1.0.

Two series of specimens will be cold rolled in order to see the effect of the presence of martensite (about 30 %) on the fatigue behaviour of the material.

Table V : Experimental matrix for fatigue testing

Partner	Steel grade	Material conditions	Stress ratio	Total strain range (%)	Temperature of testing (°C)	Frequency (Hz)	Usage factor (N/Nf)			
							0.0	0.6	0.8	1.0
EDF	AISI304L	as received	-1	0,4	20	1	0.0	0.6	0.8	1.0
		as received	-1	0,4	300	1		0.6	0.8	1.0
		cold worked	-1	0,4	20	1	0.0	0.6	0.8	1.0
		cold worked	-1	0,4	300	1		0.6	0.8	1.0
PSI	AISI321	as received	-1	0,4	20	1	0.0	0.6	0.8	1.0

SPG	AISI321	as received	-1	0,2	20	1	-	0.8	1.0
		as received	-1	0,2	300	1	-	0.8	1.0
		as received	0	0,2	20	1	-	0.8	1.0

After fatigue testing, EDF and the Paul Scherrer Institute will observe the microstructural changes related to fatigue damage (i.e. dislocation network and martensitic phase). The Technical University of INSA of Lyon (France) will support EDF in its microstructural examination and analysis.

The dislocation network will be observed through Transmission Electron Microscopy, Neutron Diffraction and advanced X-Ray diffraction methods. The presence of martensitic phase will be detected and its amount measured using metallography, X-ray diffraction and neutron diffraction techniques. Martensitic constituents and their distribution will be observed through Scanning and Transmission Electron Microscopy, Neutron and X-ray diffraction methods.

Scanning Electron Microscopy will be also used to assess the fracture mode, to identify the location and the distribution of microcracks and to identify the microstructural components which have played a role in the microcrack initiation.

In the meantime, NDT measurements will be performed on fatigue specimens by each partner (as specified in Table III).

### **ANALYSIS OF NDT DATA AND MICROSTRUCTURAL EXAMINATION**

The data obtained through non destructive testing will be compared to the results of microstructural examination. The partners will try to establish a relationship between the microstructural changes, the NDT signals and the mechanical characteristics of the material.

Quantitative criteria (accuracy, reproducibility, dispersion of data) will be set and used to characterise the performance of each technique for monitoring materials damage.

Recommandations will be drawn to exploit the potential use of the most promising non destructive techniques. The industrial feasibility of non destructive measurements on real components has to be evaluated and on site practical problems have to be taken into account.