Collaborative Research Programme in: Decommissioning, Immobilisation and Storage Solutions for Nuclear Waste Inventories (DISTINCTIVE)

Description of the Proposed Research:

Background

The UK Government remains committed to Nuclear Power as an important part of the energy generation capacity over the next 30 – 50 years, and possibly beyond. [1] The recently published 'Nuclear Industrial Strategy' describes the Governments aspirations for both 'New Build' power stations and life extension of the existing fleet. [2] A central component of public acceptance for this new build programme, however, remains a demonstrated ability to safely manage and dispose of high- and intermediate-level wastes from legacy nuclear operations. This links to on-going studies of public attitudes to nuclear power which repeatedly show that acceptance is directly linked to having scientifically robust routes for the safe clean-up and disposal of any wastes arising. [3]

Within the 2013 'Nuclear Industrial Strategy', clear milestones associated with the needs of "waste management & decommissioning" are provided and these rely upon further research and technical developments being delivered over the next 10 – 20 years. Indeed, central objectives for the strategy are "To have a joined up approach to nuclear R&D across government, industry and academia which serves to benefit the UK economy and ensures the security of supply" and "To establish the UK industry as a global leader in waste management and decommissioning …". It is clear, therefore, that there is a substantial, current and real need for further research in the broad area of nuclear wastes supporting this strategy. UK government policy associated with nuclear waste management was updated and extended through the 2008 White Paper "Managing Radioactive Waste Safely (MWRS)" published by DEFRA [4] which was based on the 2006 recommendations of the Committee for Radioactive Waste Management (CoWRM). [5] The Nuclear Decommissioning Authority (NDA) through its strategy is responsible for the delivery of the policy aims with respect to the legacy wastes. [6] Through both the policy documents and the associated strategy for delivery, it is understood that the ultimate goal within the UK is for the management of nuclear waste through a geological disposal facility (GDF). This must be underpinned by a credible programme of safe interim storage options, at various sites across the UK. The waste management and disposal strategy is also relevant to wastes arising from new build with any disposal facility requiring the capability to house wastes from these sources. [2]

Despite a programme of consultation over the last 5+ years, public attitudes to hosting a GDF remain sceptical and there is, as yet, no defined host site. [7] This sceptical attitude is undoubtedly linked to a lack of confidence in the proposed waste management approaches and highlights a continuing need to improve fundamental understanding and technology in this area. Within the scope of this call and taking account of the current UK position, we aim to provide a world-class programme of relevant research into nuclear waste management and decommissioning with special focus on UK needs. Through our Pathways to Impact Strategy we aim to bridge the credibility gap in public perception of radioactive waste management, through improved public understanding of the supporting scientific evidence and genuine two-way dialogue concerning socio-economic and ethical impacts of waste management practice. The consortium does not directly address geological disposal or radionuclide leakage/mobility, although many of the research packages will link directly to this, as this is currently covered within complementary research activity supported by NERC.

We bring together leading researchers in this field from a diverse array of backgrounds and experience with a track-record of innovation and problem solving of relevance in the nuclear field. This consortium builds upon and consolidates the work of a previous EPSRC funded programme in the same field, known as Diamond (Decommissioning, immobilisation and management of nuclear wastes for disposal, EP/F055412/1). Importantly, this new proposal draws in researchers from a larger group of universities and increases the multi-disciplinary nature of the group.

The leadership group of the consortium are all distinguished researchers with a significant track-record of working on topics of direct relevance to the major aims of this consortium funded by both industry and research councils. The research programme outlined here builds upon the collected expertise of this core academic group. Importantly, the detailed scope of the programme, and its organisation into key themes, was co-created with our industrial partners who were intimately involved during the development of this proposal; participating in various consortium building meetings and advising on the selection of projects for the research programme. This is a major strength of the consortium and ensures the relevance of the research to the medium- and long-term needs of UK industry in this area.

The strategic aims of the consortium are:

- To carry out internationally leading science and engineering research in the area of decommissioning and nuclear waste management.
- To support research that provides routes to innovative technology developments that can be applied to decommissioning and nuclear waste management.

- To foster and develop new multi-disciplinary research partnerships between academic and industry researchers.
- To train the next generation of UK researchers, equipping them with skills and experiences relevant to nuclear waste management and decommissioning issues.
- To provide a focal point for government, industry and academics through which current and future R&D issues associated with nuclear waste and decommissioning can be discussed.
- To provide a route for public understanding of the underlying research and development needs, opportunities and solutions to nuclear waste and decommissioning.

National Importance

The UK energy requirements in a "low-carbon" economy have led to a renewed interest in nuclear power in the UK. [2] There is a clear imperative, given the needs of both new build reactors and legacy wastes, to develop innovative approaches to waste management and decommissioning for safe and cost effective answers to storage and disposal issues. The need for a strong and vibrant R&D community focussed on the issues of safe storage for spent nuclear fuels and other wastes has been recently highlighted. The government response to these reports is found in the 2013 'UK Nuclear Industrial Strategy' document where the need for a strong R&D community is unequivocally recognised. [2] Indeed the report states that a key aim is for "…the UK to have a clear competitive edge in waste management and decommissioning technologies … through innovation and experience …". The work of this consortium is part of the response to this need, providing support to a strong and internationally leading group of researchers in this key technology area. It also sends a powerful signal to the international community about the UK's commitment to the field.

The consortium widens and deepens the pool of talent within academia having direct experience of nuclear related R&D. This, when allied to strong links with the industry R&D community provides an excellent route to build a greater capacity for R&D within the UK, underpinning the strategic needs of the country. Finally, we note that the consortium will train a significant number of PDRA researchers (10) with high level skills of direct relevance to issues in nuclear waste and decommissioning. This cohort will be complemented by up to 20 PhD researchers funded jointly by the university and industry partners who will be aligned to the grant. This large cohort of research staff and students directly addresses the skills agenda, a major issue for this industry. The consortium members have a proven track record in providing employees to site licensees, supply chain companies, the NDA and NNL.

The proposed research addresses key strategic priorities in EPSRC's portfolio. The research proposed directly addresses the Energy theme and the associated sub-theme of Nuclear Power. The construction of the consortium as a close partnership between academics from across the UK and key industry partners is completely aligned with the stated EPSRC strategy for this sub-theme of "… support (for) a balanced portfolio of research, to support the supply of skilled people and to maximise impact through close links with users and international partners."

Research Plan

The current UK waste inventory is large and complex: It presents multiple challenges that have not been fully understood, with a view towards finding viable technology for its ultimate treatment, storage and disposal. New knowledge and technology is still needed in areas such as waste recovery from storage facilities, conditions for interim safe storage, and stable wasteforms for different wastes over geological timescales. These needs cut across all stages of waste management and disposal and require a range of skills and knowledge to be brought to bear.

Our approach for clustering the research uses the strategic R&D themes in the 2011 NDA Strategy [6]: "Site Restoration", "Spent Fuels", "Nuclear Materials" and "Integrated Waste Management", mapping directly on our themes of "Structural Integrity", "AGR, Magnox and Exotic Spent Fuels", "PuO_2 and Fuel Residues", and "Legacy Ponds and Silo Wastes", respectively. This ensures excellent strategic alignment with the needs of UK industry.

The consortium involves staff from a range of science and engineering disciplines, including radiochemists, radiation chemists, physicists, environmental scientists, process engineers, materials scientists and civil engineers. Each research project will have a team of supervisors and we will encourage and foster interactions across the themes.

Research Plans by Theme[1]

1. AGR, Magnox and Exotic Spent Fuels (Theme Leads: Scott & Evans)

1.1 The Technical Challenge

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[1] Note that the research theme descriptions include both the directly requested EPSRC PDRA resource and industry/university supported PDRAs and PhD students attached to the consortium.

The current declared lifetime for the AGR power stations from EDF Energy will result in the generation of approximately 8800t of AGR fuel across the whole fleet. Of this inventory over 2,300t has been reprocessed to date, meaning there is estimated to be about 6,600t which still needs to be managed [8]. NDA has reported that their preferred option for AGR spent fuel, excluding current reprocessing contracts in THORP, is to keep the fuel in interim storage prior to packaging for disposal in the UK GDF in *ca.* 2075 [9]. Risks exist with longer-term wet storage of AGR spent fuel and so a transition to dry storage may be a preferred option. A similar argument has also been made for Magnox uranium spent fuel stored in the Sellafield ponds, where retrieval and repackaging is needed. For both types of spent fuels this transition, as well as the dry store environment, may carry unknown risks, e.g. to AGR cladding integrity or hydride formation on U metal, and so a better understanding is required before this can be implemented.

The goal of this spent nuclear fuels (SNF) theme is to increase knowledge and mechanistic understanding of the processes involved, and quantify the physiochemical evolution of SNF and wasteforms of different types in storage environments, with specific emphasis on aqueous (pond) as well as dry storage. The concurrent aim of the theme is to develop an internationally renowned and integrated team of experimentalists, theoreticians and modellers with significant expertise and capability in tackling waste management problems for SNF of different types in numerous different storage scenarios.

1.2 Aims and Objectives

Aim. To provide technical underpinning to the options for the management of the UK's AGR, Magnox and Exotic Spent Fuels

Objectives.

1. To understand the evolution of Magnox and exotic SNF during recovery from aqueous storage, drying and repackaging.
2. To develop spectroscopic methods for improved determination of SNF dissolution and corrosion rates in water.
3. To determine the optimum drying conditions for AGR fuels and the subsequent surface reactivity and alteration of unclad UO_2 in dry storage.
4. To determine the consequences of radiation damage in SNF, cladding and other wasteforms for safe long term storage.
5. To determine suitable waste management options for spent carbide fuels.

1.3 Work Packages

WP 1.3.1 Wet Fuel Storage Issues

Evans & Read (Loughborough), Boxall (Lancaster)

Time Resolved Laser Fluorescence Spectroscopy (TRLFS) is a very useful technique and is becoming a widely used method for fundamental actinide & lanthanide studies, particularly in Germany. In this project, the corrosion and dissolution rates of SNF in water will be investigated using TRLFS by a PDRA working at Loughborough. Using a recently acquired state-of-the-art instrument, the system will be optimised to investigate the dissolution and corrosion rates of uranium fuels (U, UC and UO_2) will be investigated, with particular focus on the solid-solution interface. TRLFS is unique in being able to determine in-situ metal speciation at environmentally relevant (pM) concentrations, which is essential when dealing with incipient corrosion of speciality metals or the alteration of ceramic and other materials used in the nuclear fuel cycle. The development of "ultra-fast" cryogenic systems offers the possibility of obtaining speciation information on ions with decay lifetimes of picoseconds. TRLFS is a very sensitive technique thatprovides information on oxidation state; U(VI) phases emit characteristic fluorescence signals and can be distinguished from an unaltered U(IV) subsurface. The final part of this project will be to extend the above investigations to mixed U/Pu and SYNROC materials in conjunction with Sellafield Ltd. This project, and UK university-based facility, will lead to many such collaborative projects in the future, both within and without the UK. Active working in NNL’s Central Laboratory will be a key part of the project where there is a state-of-the-art Am-active TRLFS.

WP 1.3.2 Transitions to Dry Fuel Storage

Scott & Hallam (Bristol), Hanson (Leeds)

A PDRA project at Bristol will utilise an array of surface analysis techniques to investigate the physiochemical changes occurring in SNF - specifically uranium metal and exotics – during recovery from aqueous storage, forced drying and repackaging in an ‘open’ but nominally dry engineered containment system. Samples will be corroded under different aqueous storage conditions and then dried using a specialist gas- rig capability to control both the drying temperature and composition of the cover gases. A key aspect will be the determination and evaluation of the transformative reactions occurring at the material surface. The project links directly to planned recovery and repackaging of spent U in the Sellafield ponds but also relates to the study of both non-irradiated Magnox U and UC (regarded as exotic fuel). The ultimate aim is to provide sufficient understanding to underpin the safety case for repackaging Sellafield's oldest wastes.

A PhD project at Bristol will explore issues associated with UO_2 surface reactivity in dry storage. The surface reactivity of UO_2 will be explored in simulated dry and wet environments, and whilst under irradiation. Photocatalytic phenomena have recently been observed in preparatory experiments that may alter the composition of storage environment gases. Under irradiation significantly increased rates of oxide dissolution and surface-mediated splitting of water are observed and these physical and chemical behaviours are of direct relevance to the safe disposal of SNF in both dry and wet environments. The project will combine both surface analysis and modelling approaches to study oxidation, hydrogenation, hydrolysis and photocatalysis on uranium oxide surfaces. Linked to this work, a PhD at Leeds will explore the optimum drying conditions for AGR fuels using a series of small scale tests on a simulant AGR fuel element. The test element will consist of cladding that is representative of that stored in a wet environment with a sealed simulant pellet inside. The testing will be carried out in a bespoke drying rig that will be capable of investigating the effect of temperature and pressure with a range of gases, followed by post-mortem materials analysis. A key aspect of this project will be a high degree of monitoring of the sample and rig to ensure that a full mass balance can be constructed and the physical and chemical processes present can be identified. An important output will be an experimentally validated process model able to predict optimum conditions for AGR drying.

WP 1.3.3 Long-term Storage Effects and Exotic Fuels

Lee (Imperial), Jimenez-Malero, Pimblott & Sherry (Manchester)

Longer term stability issues associated with the storage of SNF will be explored by 2 linked PhD projects. The consequences of radiation sensitisation for AGR fuel cladding will be investigated by a PhD student based at Manchester, who will utilise the new irradiation capabilities at the Dalton Cumbrian facility (DCF). The key aim is to elicit the role of irradiation sensitisation on the stability and performance of AGR fuel cladding through a systematic series of radiation damage experiments under relevant environmental conditions for the long term storage of AGR fuel. Data from this project will underpin a fundamental understanding of the mechanisms and rates by which a cladding may corrode in both storage and disposal environments.

Finally, a PhD student at Imperial College will explore options for the storage of exotic spent fuels. The UKs inventory of exotic fuels includes certain carbides for which no waste management decision has yet been made. This project (supported by Bristol) will examine treatment options such as thermal conversion to UO_2, direct disposal, encapsulation and immobilisation. We will employ a variety of analytical approaches, underpinned by modelling, to assess the rates and mechanisms of product corrosion in each scenario. Ultimately the project will provide invaluable data to inform the waste management strategy for these unusual wastes.

2. PuO_2 and Fuel Residues (Theme Leads: Boxall, Hyatt & Kaltsoyannis)

2.1 The Technical Challenge

The safe and secure management of Pu is a matter of serious international concern, with ~250 tonnes of separated Pu currently stockpiled worldwide. [10] Nearly 50% of this material is in long-term storage in the UK whilst the British Government develops its options for final treatment and disposition. Options for this are: [11] (i) Reuse as fuel in modern reactors (e.g. as MOX fuel or within a fast reactor (e.g. PRISM)) prior to disposal; (ii) Prompt immobilisation for disposal as soon as it is practicable; or, (iii) Continued long term storage (prior to disposition).

Option 1 is best considered in the context of Pu fuel cycle research. Thus, the focus of this theme is on options 2 & 3. R&D needs of both are now pressing: in option 3 due to it being the current default; in option 2 because of a comparative lack of R&D on Pu conditioning & packaging due to policy uncertainty as to whether it would be disposed in a GDF. Addressing these needs is complicated by Pu's high radioactivity, decay heat & radiotoxicity, nuclear safeguards requirements and, for some UK Pu contaminated materials (PCMs) targeted for disposal, poor inventory. Thus, there is also a critical requirement for research on characterisation methods for Pu bearing materials.

2.2 Aims and Objectives

Aim. To provide technical underpinning to the options for the UK's civil Plutonium inventory

Objectives.

- To understand how the structure and properties of PuO_2 change with time in the presence of H_2O.
- To understand the roles these processes play in gaseous product evolution from PuO_2 in storage.

- To understand radiation induced amorphisation and dissolution kinetics of Pu wasteforms.
- To develop novel, fast neutron based radiometric methods for the quantification, isotopic composition assessment and remote imaging of Pu bearing materials.

2.3 Work Packages

WP 2.3.1: The Behaviour of PuO_2 during Interim Storage

Boxall (Lancaster), Pimblott (Manchester), Kaltsoyannis & Kerridge (UCL), Read (Birmingham)

Of the world's 250 tonnes of separated Pu, >100 tonnes are stored at Sellafield as PuO_2 powder in sealed steel cans. Under certain circumstances, gas generation may occur in these cans, with consequent pressurisation. This is one of the most serious fault scenarios to be considered in the safety cases for PuO_2 storage and must be avoided in practice. 5 routes to gas production have been suggested: (i) Helium accumulation from α decay; (ii) Decomposition of polymeric packing material; (iii) Steam produced by H_2O desorption from hygroscopic PuO_2 due to self-heating; (iv) Radiolysis of adsorbed water; and, (v) Generation of H_2 by chemical reaction of PuO_2 with H_2O, producing a postulated PuO_{2+x} phase. The last 3 mechanisms, all involving PuO_2/H_2O interactions, are complex, inter-connected and poorly understood.

A further challenge to safe storage and eventual disposition is that a substantial portion of the Pu inventory is "out-of-specification" due to impurities (Cl or C) or unfavourable powder properties (moisture content, unusually high/low surface area). These must be treated to stabilise them for storage in welded cans in new stores. Thus, in support of option 3, this WP will seek to: (a) Understand how the structure and properties of PuO_2 change with time in the presence of H_2O; (b) Attribute these changes to fundamental chemical, physical, radiation driven processes at the PuO_2 surface; (c) Understand the generation, retention and release of helium (alpha) from within the PuO_2 matrix; (d) Understand the roles these processes play in gaseous product evolution at Pu oxide surfaces; and, (e) Understand how these processes are affected by Pu ageing, including variations in Pu isotopics. Additionally, we will study the surface adsorption mechanisms of chloride on PuO_2 and how effects such as radiation, temperature and adsorbed water affect the surface speciation and consequently desorption of chloride species under conditions to be employed in likely treatment processes.

WP 2.3.1 is organised into 3 major tasks, as follows:

Task 1 Physico-chemical interactions between PuO_2 and H_2O.
Task 2 Hydrogen generation mechanisms at the PuO_2 surface.
Task 3 Modelling bulk and surface PuO_2 structure, molecular-level chemistry and radiation damage.

Each task will be led by a PDRA. Due to Pu's unique nature (the coupling of, inter alia, challenging f block physics, radiation, anomalous materials and radiation chemistry), results from non-active simulants cannot be extrapolated to Pu and so the largely experimental Tasks 1 & 2 will focus on real PuO_2 samples. Task 1 will first establish the baseline behaviour of H_2O adsorption at PuO_2 surfaces by direct gravimetric measurement of H_2O uptake using a novel quartz crystal nanobalance technique pioneered at Lancaster. This will be followed by hydrothermal and electrochemical studies of the chemical interactions between adsorbed H_2O and PuO_2 and their possible roles in H_2 evolution at the PuO_2 surface with accompanying production of the postulated PuO_{2+x} phase. Hydrothermal studies will focus on hot-stage based experiments with state-of-the-art fuel cell based H_2 sensors to assess H_2 evolution rates as a function of T, humidity, pO_2 and Pu isotopics, the latter to discriminate between chemical and radiolytic routes to H_2. By analogy to work on UO_2, coupled electrochemical (cyclic voltammetry, impedance spectroscopy)/nanogravimetric measurements will be conducted on PuO_2 coated electrodes in aqueous electrolytes as functions of T/[O_2]/pH to determine the thermodynamic/kinetic susceptibility of PuO_2 to oxidation and especially any evidence of the PuO_2/PuO_{2+x} transition.

Task 2 will use radiation chemical experiments and stochastic simulations to understand the behaviour of the adsorbed surface water on PuO_2 and its role in radiolytic H_2 generation. Experiments will again be conducted as a function of Pu isotopics and in contact with H_2/air and $H_2/O_2/Ar$ gas mixtures to interrogate putative H_2/O_2 recombination at the PuO_2 surface. These studies will deploy the same oxides and contaminants used in Task 1 to allow development of a synergistic data set. Hydrothermal, electrochemical (Task 1) and radiochemical (Task 2) experiments will be conducted in the presence and absence of Cl impurities and supplemented by surface and bulk phase characterisation using Raman, FT-IR, P-XRD, SEM, BET at NNL and XPS at JRC-ITU, the latter via an access application through TALISMAN, affording both international collaboration and resource match to the project.

Complementing Tasks 1 & 2, the UCL-led modelling in Task 3 will be supported by work at the Universities of Birmingham and Manchester through 2 PhD students. At UCL, Density Functional Theory will be employed with (i) periodic boundary condition techniques to study bulk and surface PuO_2 and (ii) periodic electrostatic embedded cluster method (PEECM) to model interactions of PuO_2 surfaces with water and their radiolysis products. H_2 generation mechanisms will be explored, as will the surface reactivity of chloride species. Atomistic modelling techniques will be employed at Birmingham to model bulk and surface PuO_2 and its defect chemistry, and molecular dynamics will be used to model radiation damage and helium accommodation. In Manchester, a PhD student (funded by a President's Scholarship) will support Task 2 by focusing on stochastic modelling of water on PuO_2 surfaces

WP 2.3.2 The Behaviour of Pu Bearing Wasteforms and Encapsulants

Hyatt, Provis, Whittle & Corkhill (Sheffield)

Approximately 5% of the 100 tHM Pu stockpile is considered unsuitable for reuse as fuel, due to contamination by Cl (from PVC packaging) or ^{241}Am ingrowth, and will require immobilisation (i.e. Option 2, above). This WP will address fundamental aspects of Pu wasteform processing, radiation damage behaviour and disposal, in three distinct projects.

A key concern for the long term storage and ultimate disposal of actinide ceramics and glass-ceramics is amorphisation resulting from cumulative α-recoil damage, leading to swelling and cracking of the wasteform. Building on previous research, we hypothesise that radiation amorphised structures are stabilised by the formation of oxometallate polyhedra *with reduced co-ordination number* in damage cascades. The amorphous structure is thus *topologically stabilised* by acquisition of surplus degrees of structural freedom. A PDRA project will address this novel hypothesis in three ways. First, we will utilise *ex situ* ion beam irradiation (100 keV - 1 MeV) to produce thin (0.1 - 1 μm) surface amorphised layers on polycrystalline ceramic or thin film specimens of perovskite, garnet and pyrochlore host phases. Utilising XAS, Mossbauer and other spectroscopic methods with surface sensitivity (e.g. in grazing angle or electron yield mode), we will unravel the element specific averaged local co-ordination environments. By careful control of chemical composition we will investigate the influence of ion size, crystal field, and electronic structure effects on the stability of oxometallate polyhedra with reduced co-ordination number. Second, we will utilise energetic (GeV) ion beams to fully amorphise synthetic host phases, yielding macroscopic samples (ca. 100 mg) for neutron and X-ray total scattering studies. Combining this novel approach with the study of metamict mineral specimens, we will use Reverse Monte Carlo simulations to develop the first comprehensive structural description of radiation amorphised materials, providing insight into the role of small oxometallate polyhedra in stabilising amorphous structures formed across different dose rate and energy transfer regimes. Third, we plan to investigate 30y old ^{238}Pu doped Synroc samples within NNL active laboratories, using TEM methods to characterise the crystalline/amorphous nature of the component phases and accumulation of radiogenic helium; EELS speciation will provide a link to our study of co-ordination environments using bulk techniques. Overall, this work will link to an existing PDRA project at Sheffield, using molecular dynamics simulations and topological analysis to understand radiation damage structures. Analysis of this unique suite of specimens will provide invaluable insight on the evolution of these materials during long term storage, to inform the safety case supporting ceramic disposition of Pu.

Current glass-ceramic formulations Pu disposition incorporate ca. 2- 5 mol% CaF_2 which is believed to act as a "mineraliser", aiding digestion of PuO_2 (or CeO_2 / UO_2 surrogates). However, α-decay of Pu induces (α,nγ) and (α,pγ) reactions on ^{19}F (100% nat. abundance) leading to prohibitive dose uptake by glove box operators. The project will develop a mechanistic understanding of the "mineralising" action of CaF_2, as a first step to developing an improved glass-ceramic formulation in which CaF_2 addition is minimised, supplemented or replaced by an alternative. Building on this approach, we will develop a mechanistic understanding of Ce, Th, U surrogate partitioning between ceramic / glass phases, using thermodynamic calculations to predict the behaviour of Pu.

A PhD project will investigate the incorporation of Pu, Np and Am within cements relevant to application in waste encapsulation and engineered barriers in a GDF. This requires a mechanistic understanding of Pu/Np/Am interactions with the main sorbing components of the cement, including CSH and CASH phases, as a function of C:S ratio, redox conditions and extent of carbonation. Standard and advanced (e.g. NMR) cement characterisation techniques and aqueous and solid radioanalytical techniques (LSC, autoradiography, α -spectroscopy) will be complemented by synchrotron μ-XRF/XRD/XAS to determine the spatial distribution, speciation and chemical environment of the sorbed radionuclides, leading to a robust model of sorption mechanisms under relevant storage / GDF conditions.

WP 2.3.3 Methods for the Characterisation of Stored Pu, PCM and Pu Contaminated Facilities

Joyce (Lancaster)

This WP will involve the work of 2 PhD students at Lancaster. The first will explore how the isotopic composition of Pu in storage changes with time as a result of radioactive decay such as ^{241}Am in-growth and presence of ^{244}Cm. The latter's tendency to spontaneous fission and stimulus of intense (n,α) reactions complicates isotopic composition assessment by fast neutron multiplicity, so requiring use of lab based methods. These are complicated by current radiometrics being dependent on ^{3}He gas which is no longer available. Based on significant prior art at Lancaster, we will research use of non-^{3}He constrained fast-neutron multiplicity analysis to infer isotopic composition, in-situ. Focus will be on discrimin-ation of $^{240}Pu_{eff}$, ^{252}Cf, ^{241}Am and ^{244}Cm, with applications including Pu accountancy in storage (supporting WP1), criticality assurance and proliferation prevention during Pu management. We shall draw on the use of facilities at NPL, IAEA, NNL for the associated experimental campaign, complemented by prior simulations with Geant4, FISPIN, MCNP.

A second PhD project will explore an imaging system for *in situ* characterisation of Pu distribution in heavily contaminated α facilities. Based on the collimation & detection of fast neutrons, Pu content will be identified separately from fission fragment radioactivity without need for swabbing / manual intervention, so informing the decontamination plans for such facilities. As above, it will be relevant to the security and integrity assessment of Pu storage environments.

3. Legacy Ponds and Silo Wastes (Theme Leads: Hriljac & Lee)

3.1 The Technical Challenge

The legacy ponds and silos (LP&S) at Sellafield are currently the biggest safety and security threat facing the UK, costing £70M/annum just to maintain their basic condition and prevent leakages. [6] They represent 22% of all Sellafield site programmes, 35% of total site costs and 77% of major project costs during next 4 years and >90% of nuclear hazard potential. LP&S decommissioning and clean up has to be the UKs top priority engineering programme. Objectives of the NDA's LP&S Strategy include: Acceleration of high hazard/high risk reduction; Reduction or mitigation of the impact of the risk of a loss of containment of nuclear materials; Preparation of the facilities for retrieval operations; Retrieval of the waste; and Immobilisation of the waste. The projects in this theme will provide fundamental understanding of key aspects of the Sellafield programme including Characterisation, Mobilisation and Transport, Multi-phase sludge transport, immobilisation and conditioning, and effluent clean-up and monitoring of the effluent prior to discharge.

3.2 Aims and Objectives

Aim. To develop innovative technical approaches to clean up UK legacy wastes.

Objectives.

- To understand durability of heterogeneous ILW glass/ceramic wasteforms from LP&S wastestreams.
- To develop improved ways to remove radionuclides (RNs) from solution, both novel inorganic ion exchange solids and tailored binding superparamagnetic nanoparticles, to treat complex and variable effluents.
- To develop new micro- and ultra-filtration methods for use with sludges.
- To provide three-dimensional modelling and simulation for sludge disturbance, mobilisation and transport, with supportive experimental studies, and manipulation planning for removing corroding nuclear materials.
- To develop a better understanding of gas hold-up in sludges.
- To develop improved techniques for remote monitoring of sludges and heterogeneous wastes.

3.3 Work Packages

WP 3.3.1 Wasteform Durability

Lee & Grimes (Imperial), Hyatt (Sheffield)

The safety cases associated with the long-term stability of wasteforms used for the storage and disposal of Sellafield's LP&S wastes require improved understanding of their corrosion mechanisms. An issue with such wasteforms is likely to be their heterogeneity arising from incomplete characterisation of the wastes from which they are derived. Through a PDRA project, linking to existing projects at Imperial & Sheffield, we will build on previous surrogate trials using thermal methods such as plasma vitrification and Joule heated in-can vitrification. The project will use a combination of durability testing and modelling across length scales to examine the impact of local equilibria on corrosion at different locations in the microstructure. Key features to be examined include impact of differential thermal expansion of phases and modelling interfaces at high temperature, subsequent lowering of temperature to analyse how the residual strain is accommodated, variation of corrosion down cracks compared to outer surfaces, and transport along interfaces since they are likely to be an important route for loss of RNs. This will be linked to durability testing of active samples. This will be the first time that such a combination of hierarchical modelling and corrosion testing of multiphase materials has been attempted and will give the UK a world-lead in this field as well as providing crucial support for the safety case to dispose of these complex wasteforms. This project will be supported by an allied PhD project at the University of Sheffield, which aims to develop thermal treatment processes for UK legacy ILW wastes (e.g. PCM, ion exchange materials, sludges). The focus of this project will be a programme of bench scale experiments using realistic simulant mock ups, aimed at understanding the interactions between waste and glass forming additives during thermal processing. This will be achieved by time resolved investigation of the melting process through *in situ* characterisation of off gas chemistry and particle entrainment, and *post mortem* characterisation of the product phase assemblage, microstructure, and composition, supported by thermodynamic calculations where possible.

WP 3.3.2 Effluent Treatment and Analysis

Hriljac & Read (Birmingham), Evans, Platt & Holdich (Loughborough) Ryan & Vandeperre (Imperial)

Efficient and reliable removal of RNs from effluent will become increasingly challenging in the UK nuclear industry as decommissioning and post-operational clean-up will produce a variety of waste streams in widely differing physical conditions. Working through a PDRA and 3 linked PhD student projects, this work package will develop new effluent treatment materials and processes. Standard ion exchange materials for removing cations such as Cs and Sr from solution are porous inorganic solids such as zeolites that work well in properly conditioned effluent in plants such as SIXEP, but have limitations in other waste streams. A SL SWOT analysis as part of effluents strategy development has identified a knowledge gap for ion exchange materials to use in acidic environments, there are also concerns with high concentrations of competing ions and the presence of decontamination agents/complexants. At Birmingham, a PDRA will use atomistic computational modelling as a means to sift through libraries of potential materials based on known mineral structures and then synthesise, characterise and test them under realistic conditions (based on SL/NNL input) for RN removal. Calculations will focus on comparisons of thermodynamic stability of different cation forms and ion exchanger compositions including those likely found in acidic environments, this is a novel approach to the development of new ion exchangers. Parallel research from 2 linked PhD students will focus on a different strategy to simultaneously quantify and remove RNs from waste streams using superparamagnetic bead and nanopore technologies. Nanoparticles will be synthetically modified with ligands capable of binding specific RNs from solution. A PhD student at Loughborough will focus on the detection and quantification of species bound to the particles, whilst another at Imperial will focus on developing the separation technology for the RN material. The ultimate aim is to develop a multiplexed platform technology allowing detection and separation of different species by using targeted ligands on the nanoparticles.

Ultra- and micro-filtration approaches for effluent stream treatment using rapid recycle pumping of membranes can lead to significant shear of the suspended material leading to a variety of problems. An alternative approach is to move the membrane using vertical vibrating tubes oscillated by a resonant force. A PhD project at Loughborough will investigate the use of different filtration media in this resonant fashion with a number of simulant materials to deliver a theoretical understanding of a compact system that can be used for a variety of different filtration applications.

WP 3.3.3 Pond and Silo Sludges

Day & Scott (Bristol), Biggs, Fairweather & Peakall (Leeds), Kerridge (UCL).

Our current understanding of the behaviour of relevant nuclear waste sludge is poor due both to the complex nature of the particle phases and to a limited availability of useful data for these materials. Further development of our understanding of these sludge materials, and the development of quantitative predictive procedures is needed if we are to design more efficient and safer treatment processes. The work to be undertaken here by two PDRAs (linked 2 year appointments) and 3 PhD students will improve our understanding of operational problems associated with sludges, and will lead to reductions in the costs associated with sludge separation and increases in process efficiency (and hence reductions in operation timelines). At Leeds, one PDRA will focus on the use of realistic sludge simulants (using recipes from NNL/Sellafield), with a range of particle shapes, sizes and densities, and their flow in horizontal and vertical pipes. Characterisation of the segregation of the particle materials within these different pipe flow arrangements will be undertaken using novel ultrasonic approaches recently developed at Leeds in partnership with Sellafield Ltd. Predictions will be carried out by a second PDRA using a coupled large eddy simulation/Lagrangian particle tracking technique. The influence of levels of turbulence, and the direction of gravity, on particle agglomeration and settling behaviour, and the shear break-up of particle aggregates will be explored. For the first time, measurements and modelling of non-spherical particles in turbulent flows will be explored. Ultimately our goal is a validated predictive model for complex particulate laden flows in closed pipes with geometries relevant to nuclear operations.

Hydrogen gas retention in nuclear waste sludge is also of critical interest for LP&S wastes. The risk of hydrogen hold-up in either man-made voids or within fracture zones inside a sludge bed and a sudden release of gas following bed disturbance is clear. Despite this, much remains to be understood about the likelihood of large gas pockets forming within sludge and the features of the sludge that may increase or decrease the risk. The focus of this PhD project at Leeds will be to develop an understanding of how gas is retained and released from nuclear waste sludge in order to (i) address current storage issues, and (ii) to design future sludge storage regimes in order to avoid sudden gas releases.

In the immediate aftermath of sludge disturbance, sequestered RNs including Pu, Am, Cs, and Sr are likely to be released into the immediate aqueous environment, along with other particulate matter. Atomistic modelling simulations through a PhD project at UCL will explore the dominant interactions in this environment, including RN absorption on brucite, artinite and other hydrous magnesium carbonate particulates. Simulations will reveal how such interactions are affected by the presence and concentration of carbonate species, contaminants, and other ions. This work links strongly to new approaches for the immobilisation of RNs through the introduction of strongly sorbing materials.

Finally, within this work package, a PhD project on developing fibre optic probes for remote collection of Raman and fluorescence spectra in nuclear storage facilities will be undertaken at Bristol. The development of novel probes for characterisation of materials in storage facilities where human-entry is difficult or impossible is clearly of value in a nuclear context. Current research for medical designs are based on disposable probes, these may also have application where radiation damage is severe. This project will field-test prototype probes by the end of the project, incorporating the use of fluorescence imaging and Raman Spectroscopy in a single instrument.

4. Structural Integrity (Theme Lead: Lunn)

4.1 The Technical Challenge

Monitoring, preserving and improving the integrity of on-site structures is a significant issue for site decommissioning and management. This theme focuses on complex technical challenges ranging from site-scale infrastructure preservation and restoration, to characterisation and handling of individual waste packages. Legacy plants contain buildings, chimneys, storage ponds, silos and waste packages that have deteriorated over several decades. The nature of site operations has also led, in some areas, to the creation of extreme radiation environments. Access to infrastructure, for characterisation and preservation, may be limited by high radiological hazard and/or by the proximity of other fragile or hazardous structures. These uniquely challenging conditions can result in an increased financial cost and a severe reduction in the availability of engineering solutions.

4.2 Aims and Objectives

Aim. To develop reliable systems for infrastructure characterisation, restoration and preservation, that minimise current, and future, radiation exposure to the workforce whilst providing economically viable technological solutions.

Objectives.

- To develop in-situ ground barriers that could act as a ‘second skin’ surrounding on-site structures, such as silos and ponds, for prevention of subsurface radionuclide migration.
- To develop smart solutions for remote crack detection, infrastructure health prediction and building preservation that can be retrofitted to existing sites.
- To develop autonomous systems with increased functionality and to coordinate them through a CAD-based real-time management system, to facilitate planning and execution of decommissioning works.

4.3 Work Packages

WP 4.3.1 Physical Ground Barriers for In Situ Contaminant Containment

Lunn, Hamilton, El Mountassir, Saafi, Tarantino, Yang & Lord (Strathclyde)

This work package will focus on the development of low viscosity colloidal silica based grouts for reducing the hydraulic conductivity of near surface soils to inhibit the migration of radionuclides in groundwater. Previous research at Strathclyde has developed proof of concept data for novel grouting technologies. These include microbially induced calcite precipitation for sealing fractured rock; colloidal silica for grouting rock fractures and reducing the permeability of porous media; and, novel cementitious grouts that enable remote detection of grout penetration.

Low viscosity grouts have greater penetration distances than cements and as such can be injected at greater distances from the final barrier location and will require fewer injection points, thereby reducing the potential for radiation exposure to workers and the volume of contaminated soil for disposal. This project will combine the colloidal silica based grouts with novel magnetic technology for remotely monitoring the grout penetration front from the ground surface to enable monitoring of barrier construction and integrity.

This project, involving both a PDRA and PhD student, will involve experimental testing from batch experiments up to full-scale field trials. Initial laboratory batch experiments will investigate the gelling behaviour of colloidal silica under a range of different environmental conditions, including groundwater pH, salinity & composition and near surface soils with varying compositions of sand, silt and clay. Column experiments in a variety of soils with varying grain size distributions, including sediments similar to those sampled at the Sellafield site, will determine the reduction in hydraulic conductivity that is achievable (desirable $\leq 10^{-9}$ m/s). Additional column experiments will be carried out with the University of Loughborough to investigate how the grout interacts with radionuclides (Tc, Sr).

A second phase of laboratory investigation will involve the construction of a large-scale 3D laboratory testing facility (>8m^3). Here, a purpose-built, watertight tank will be constructed with multiple drainage, injection and monitoring points, to design and test the grout injection strategy. The experiments will involve grout injection via multiple boreholes to enable formation of a hydraulic barrier within the tank. The efficacy of the hydraulic barrier will then be determined via cross-barrier hydraulic testing. The accuracy of the surface monitoring technology to detect grout penetration distances will also be explored. Experiments will be conducted in both layered and non-layered sedimentary environments. Specifically, in the layered sediments, the potential will be determined for higher permeability sedimentary layers to be actively deployed for creation of horizontal hydraulic barriers. Finally, an experiment will be designed to investigate the potential for hydraulic drainage of the soil volume between a legacy containment vessel (pond or vault) and an injected barrier, applying a similar concept to draining the gap between the primary and secondary concrete skins on double-skinned storage ponds.

The results of the large-scale field experiments will be used to design the field injection strategy in consultation with BAM Ritchies; the leading experts in contaminated ground isolation in the UK. BAM Ritchies will provide the operational component of the field trial (see letter of support) including borehole drilling, grout injection and post-grout hydraulic testing. The field trial will be conducted at an appropriate location, determined through consultation with industry; NNL have already offered the Workington site. In the field trial(s) a hydraulic barrier will be created via multiple injection points. A particular target in barrier design will be to develop strategies that minimise the waste arising from drilling and the time that workers are required on site. The efficacy of the injected barrier will be determined via monitoring of grout penetration at the surface (using novel magnetic technology developed at Strathclyde to determine the location of the penetrating grout front) and via pump tests conducted in-situ to determine the reduction in hydraulic conductivity..

WP 4.3.2 Remote Crack Detection, Infrastructure Health Prediction and Building Preservation

Hamilton, El Mountassir, Yang & Saafi (Strathclyde)

Intelligent monitoring and preservation systems for management of concrete structures on nuclear sites could bring a number of key advantages: exposure of workers for maintenance checks could be reduced by remote monitoring devices; early intervention and preservation schemes could reduce the amount of restoration required, thus reducing both radiological exposure and cost. To develop structural preservation systems, concrete degradation in building infrastructure needs to be understood and halted. This project will involve 3 PhD students and focus on 4 main areas: (1) development of microscale embedded Miniature Micro-Electro-Mechanical Systems (MEMS) based sensors and smart surface paint for detection of water, chloride ion content and surface cracking; (2) Modeling water flow paths through the building façade and reactive transport; assessing damage patterns, including allowing for climate change effects; (3) Multi-scale modeling to predict crack nucleation and growth; and, (4) crack healing technologies via novel nanoparticle sealants. Research will build on current expertise at Strathclyde and will involve laboratory investigation and field trials through retrofitting of monitoring devices to an existing site.

WP 4.3.3 Development and Real-time Management of Autonomous Systems for Decommissioning

Stolkin (Birmingham), Lennox (Manchester)

A significant part of projected decomissioning work, especially at Sellafield, will need to be carried out using robotic tools and equipment. There is a need to improve current robotic capability and operational robotic control. Through two linked PhD projects, at Birmingham and Manchester, the work package will focus on (1) novel remote material characterization technologies in extreme radiation environments and (2) the real-time on-site management of autonomous systems with improved control systems. Before robotic actions can be properly planned and executed, 3D characterisation of the scene is needed. Research will develop methods for instant real-time 3D as-built CAD modelling of nuclear plant. A linked PhD at Manchester will develop tomographic methods to autonomously characterize radioactive sludge in legacy storage ponds and silos. An invasive method will be developed for imaging through thick concrete for future use on mobile robots. A key deliverable will be the construction of a demonstrator system that will operate in a 3.6m x 3.6m x 2.4m tank located at the National Nuclear Laboratory's rig-hall in Workington. WP 4.3.3 will be complemented by integration with a recently awarded industry-funded PhD project (beginning October 2013) to develop vision-based trajectory control of non-sensored robots to increase functionality, without robot hardware modification.

Cross-Cutting Activities

Active Research Projects

A key component of the research programme outlined here is the use of world-class active research facilities at the National Nuclear Laboratory (NNL), and elsewhere. By its very nature, active research is costly, time consuming and needs careful planning. Two major projects, through WP2.3.1 of theme 2 in this proposal, are already planned: These require the embedding of 2 PDRA staff within the NNL for the duration of the research programme. Costs for this and the associated leverage are defined and built into our plan. A number of other projects across the consortium have also indicated a potential need for active research facilities, although as yet the detailed understanding of their facility needs and the duration of the work has not been fully defined. Simon Pimblott will act for the consortium as the Cross-Cutting Champion in this area with a responsibility to promote the use of active facilities and to ensure that the consortium receives excellent advice and support in the area, especially relating to technical needs and duration of work.

Outreach & Public Engagement

The outreach activities will be led by Hyatt, who will act as Cross-Cutting Champion in this area. Full details are provided within the 'Pathways to Impact' statement and JeS form. The cross-cutting theme will have a responsibility for promoting the work of the consortium through delivery of this plan. Central to this is the development of outreach materials and activities that underpin a core message about the importance to the economy and society of our research programme. The cross-cutting theme leader will also be responsible for supporting the promotion of the consortium to stakeholders, policy makers and the general public, engaging other researchers in the consortium, as needed, to deliver the plans.

Industry Support

The consortium has 3 key partners from the UK based industry, viz.: Sellafield Ltd., the National Nuclear Laboratory (NNL), and the Nuclear Decommissioning Authority (NDA). These partners have pledged a variety of support to the consortium. This includes 10 PhD scholarships, supervisor time for engagement in projects, attendance of staff at key research meetings and support for the consortium leadership the International Advisory Group (IAG). Additional support from a wider-range of industry partners is expected. We have had preliminary discussions with partners who worked with us on the 'Diamond' consortium such as AMEC, Nuvia and MMI. We have also had contacts with other interested possible partners such as EDF and Rolls-Royce. Given the limited time available for the development and submission of this proposal, this wider package of support to be offered is not yet fully defined. However, the collaborating academic partners have an extensive range of contacts across the nuclear industry supply chain that will have an interest in the research associated with this consortium. We fully expect, therefore, to receive further support both cash and in-kind for work that will complement that described in the proposal.

Over the last 5 years, consortium members have received > £10M of support from various industry partners including Amec, British Energy, NDA, NNL, Sellafield, Magnox, Nuvia, Rolls-Royce and AWE. Given this track record and the advantages of linking to an internationally-leading research consortium, we realistically expect significant additional support for this program. A key goal of the consortium during the first 6 months will be to deliver additional support for the activities of the group from industry; the management team intend to undertake a targeted 'roadshow' of potential partners to showcase the consortium and benefits of interacting. This forms part of our 'Pathways to Impact' plan.

International Partners

Members of the consortium have a wide range of important international partnerships in the nuclear research field. Examples of these links include ANSTO, CEA, FZK, LANL, INL, PNNL and SRNL. Research groups in the consortium have regular interactions with these centres of excellence and researcher exchanges are commonplace. We have received offers from partners such as ANSTO, PNNL and SRNL to participate in the consortium through the hosting of researchers for short placements. We also will look to maximise the value of these links for access to specialist facilities, for example at FZK. We will actively seek routes to allow these links to be further developed for the benefit of the consortium. We are especially interested in developing opportunities for training with these partners and informal discussions have already begun.

Links to On-Going Research

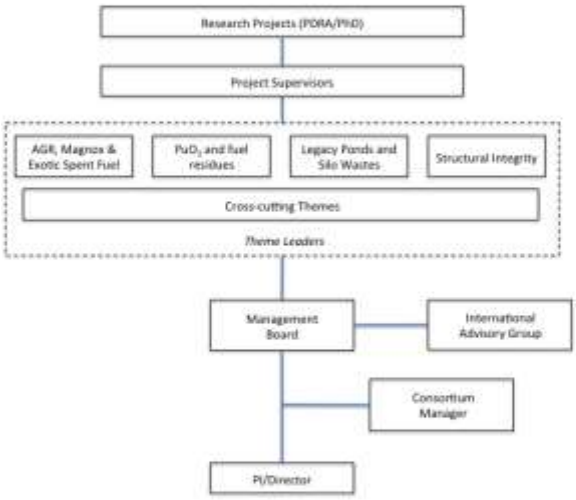
This consortium involves key members of the previous EPSRC consortium in this area (Biggs, Hyatt, Lee, Evans, Kaltsoyannis) known as Diamond (Decommissioning, immobilisation and management of nuclear wastes for disposal, EP/F055412/1). These partners have ensured that the research proposed here builds upon and significantly extends the outcomes from this earlier project. Members of this consortium are also key members of a number of EU FP 7 consortia such as ASGARD, CINCH, SACCESS etc. There are also links between consortium members (Grimes, Lee, Boxall) and the NERC consortium on Spent AGR fuels as well as with the UK-India EPSRC programme (Lee, Hyatt & Fairweather). Lee is also part of an IAEA Coordinated Research Project on HLW forms and Spent Fuels involving 10 countries.

There are also strong existing links between the academic group and our key industry partners. Scott and Biggs lead the Sellafield funded Centres of Expertise on Reactive Metals and Sludges, respectively. The NNL also has strategic research partnerships with Imperial College, Lancaster, Leeds, Sheffield and Manchester. A key objective for the management team during the first 6 months will be to maximise the value of all these links and explore routes for collaborative work between these groups and the consortium.

Leadership & Management

The Principal Investigator and Director of the Consortium will be Professor Simon Biggs. He will be supported by a consortium manager (0.5FTE) who will provide general support to the Director and consortium members across all of the specified activities within this proposal. Biggs has extensive experience of leading large research projects and is, or has been, a PI or Co-I on 6 major consortium grants (EPSRC, ERDF or EU) in the last 5 years. Biggs will devote 0.2FTE to leading the consortium. He will be supported by an academic leadership team of Boxall, Evans, Hriljac, Hyatt, Kaltsoyannis, Lee, Lunn, Pimblott and Scott as members of the Management Board (MB). All are leading academics with extensive records of research leadership, collaborative projects in the nuclear industry, and training of PDRA staff.

Strategic Leadership: The consortium will seek regular advice from its International Advisory Group (IAG) which will include representatives from partner companies involved in supporting the research objectives as well as leading academics from complementary international research activities. The IAG will meet annually and advise the Director and MB on the scientific program to ensure continued relevance and focus within the consortium. The MB will meet formally biannually and its members include the Director, the academic leadership team and the consortium manager. It will have oversight of the budget and allocation of resources, and will review progress against targets. Importantly, we have a strategic research fund to facilitate active research, international research secondments, and small equipment purchases. These need to be allocated against bids from individual projects and taking account of the strategic aims of the consortium. The MB will establish guidelines for projects to bid and will be responsible for deciding how and when to use this research fund for the achievement of the consortium research goals. A representative from EPSRC has an *ex officio* position on all management and leadership groups.



Functional Management: Responsibility of the Director and Manager, who will: ensure contracts are up-to-date, funds flow as appropriate, maintain website and newsletter, facilitate and promote cooperation between members, ensure opportunities for collaboration are realised, collect and organise data (financial and academic) for reporting, ensure international links are promoted, keep industry updated with consortium activities and successes, organise annual meetings, and prepare annual reports.

Research Themes: Theme leaders will organise meetings (annual), ensure effective communication between theme members, liaise with Director/manager to supply information for newsletters, annual reports, website, etc., and organise aspects of annual meetings as required.

Cross-cutting activities: Two cross-cutting activities are planned, “Active Research” and “Outreach & Public Engagement”. The cross-cutting theme leaders (Pimblott & Hyatt) will serve as members of the leadership team on the management board. They have responsibility for delivering against the plans of the consortium in these areas ensuring effective communication of the issues associated with these themes to other members of the management board.

Projects: Project leaders will be responsible for the delivery of the research aims of consortium, writing of journal articles and participation at conferences and will ensure involvement of all supervisors in projects through regular meetings and e-mail updates/consultations.

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