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Underwater Spectroscopic Techniques for In-situ Nuclear Waste Characterisation–22573

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ABSTRACT

Decommissioning legacy spent fuel ponds within nuclear facilities can be a complicated process, largely in part due to the unknown state of materials deposited into such storage ponds during the operational lifetime of the facility. Materials may have corroded, and their condition deteriorated. Due to the nature of the materials deposited in such storage sites, minimising disturbance is desirable, and as such non-destructive techniques such as optical analysis methods are preferred over destructive techniques. In this work, we demonstrate three such optical techniques (Raman spectroscopy, photogrammetry, and hyperspectral imaging) capable of ascertaining useful characteristics of objects such as material type, surface corrosion, degradation, and 3D structure. A pool environment was used to capture data and demonstrate the techniques suitability for use in nuclear waste characterization in active spent fuel ponds. The optical techniques used enabled material characteristics to be obtained.

INTRODUCTION

Spent nuclear fuel assemblies are removed from nuclear reactors but continue to generate decay heat for a long time after removal from the reactor. Fuel assemblies produce about 6% of the heat during steady-state critical reactor operation upon shutdown, which rapidly decreases to about 0.2% after a week. Despite the fast decay of short-lived fission products, heat generation of spent fuel assemblies typically still amounts to ca. 10W per kg after one year after removal from a reactor, and therefore must be cooled for at least that period. Water has a high heat capacity and absorbs ionising radiation, which makes it a good coolant for spent fuel assemblies. Spent fuel is usually stored in a spent fuel pond, a large body of water that serves two purposes: to cool the fuel, and to provide shielding from harmful ionizing radiation.

Spent fuel ponds are found at most nuclear facilities handling or storing spent nuclear fuel assemblies, with sites typically having two or more ponds. While the ponds are in active operation, they must be regularly maintained, and the stored materials monitored. Sites such as Sellafield’s legacy ponds, ponds used for long term storage of radioactive materials, provide a significant challenge from a decommissioning perspective as they date back to the start of the UK’s nuclear program. As such these ponds were not originally designed with decommissioning in mind, or to handle the throughput associated with servicing of the UK’s Magnox reactors. They present unique challenges as they were designed open to the elements and therefore over the years have accumulated debris settling in the ponds, producing a sludge at the bottom. This sludge has covered objects and fuel assembly skips, as well as increasing the overall water turbidity, making identifying and moving objects more difficult [1].

Optical techniques deployed in the marine environment are generally rapid non-destructive assessments, requirements which also suit the characterisation of nuclear materials in spent fuel ponds. However, underwater optical techniques are not without their drawbacks. Imaging under water presents challenges resulting from differences in light propagation properties [2] as well as pressure, temperature, and salinity [3].

In this study, spectroscopy techniques were utilised and applied to the characterisation of nuclear materials in spent fuel ponds. Photogrammetry and hyperspectral imaging were further used to obtain information about the condition of materials in these spent fuel ponds.

Understanding light attenuation in water

To conduct submarine spectroscopic studies, it is fundamental to account for the wavelength dependent interactions of light with water. For example, optical absorption is strongly dependent on the wavelength of light. In pure water, there is a 100x increase in optical absorption for light with a wavelength of 700nm compared to light with wavelengths of 450nm. Consequently, the longer wavelengths of the light spectrum—red, yellow, and orange, penetrate water relatively poorly. Respectively, under direct sunlight, these colors of light would only reach depths of approximately 15, 30, and 50 meters [4]. Conversely, the shorter wavelengths of the light spectrum violet, blue and green can penetrate much further [4]. Freshwater attenuates light in an equivalent manner to saltwater except that it is slightly less attenuating at shorter wavelengths [5]. Aside from water molecules, real-world aquatic environments contain many additional constituents such as dissolved salts, organic and inorganic substances, and particulates. All of these constitute additional absorbers should also be considered when determining the total optical aqueous attenuation [6].

Aside from optical absorption, optical scattering must be considered. Unlike absorption, scattering processes redirect photons instead of absorbing them. This has a similar net effect to absorption by decreasing the total amount of transmitted light, i.e. it attenuates the transmitted light. The degree of optical scattering often depends upon the wavelength-particulate size ratio. In marine settings this wavelength dependent scattering occurs via two possible mechanisms. Light can either scatter at the atmosphere-water interface (i.e. the surface) or it can scatter from particulates in the water [7]. As with optical absorption all scattering processes should be considered to determine the net attenuation and thus calibrate instrumentation correctly.

RGB imagers

The traditional RGB camera has become a vital characterisation tool in the marine environment for wildlife surveys and infrastructure inspections. It has become a staple of marine robotics, and every modern remotely operated underwater vehicle (ROV) is equipped with at least one RGB camera for piloting the robot and/or for recording data. This system can be used for a wide variety of data gathering techniques from simple stills to the observation of submerged objects, to more complex amalgamations of image sets to produce orthomosaic maps. These can be used to understand object distribution in spent fuel ponds. A drawback of RGB cameras and photography is the spectral range, largely limited to the spectrum visible to humans + infrared. This spectral range can be extended with other techniques to determine the molecular composition of an object (with Raman spectroscopy) or to observe NIR fluorescence (hyperspectral imaging).

Photogrammetry

A set of RGB images can be used to generate photomosaics and extract 3D information using photogrammetry techniques such as Structure from Motion (SfM). This technique requires images to be taken in a raster pattern with sufficient overlap [60-80%] between adjacent images [8] and cover the object of interest from all angles. From these images, the SfM algorithm can calculate correlation between images using points of interest, and then stitch the images together to generate the photomosaic. The algorithm can also generate 3D information using camera positions and the points of interest relative to each other [9]. Generating a mesh or Digital Elevation Model (DEM) can be used to extract information about the structure of a 3D object. In nuclear applications, capturing the 3D structure of an object can assist in the identification process, and repeated capturing over time enables continuous monitoring of corrosion and degradation processes.

Hyperspectral imaging

Underwater hyperspectral imaging is a relatively new field but has the potential to revolutionise the way marine environments are mapped. The technology has already been adopted for aerial survey applications using UAVs in the last decade, with hyperspectral imagers deployed on drones. It is used to survey objects of interest on land, and submerged objects down to a depth of about 20 m [10]–[15].

When mounted on underwater robots, hyperspectral imaging has the potential to be a time- and cost-efficient identification and mapping method covering large areas over a short time [16]. Hyperspectral imaging has proven useful on satellites and aircraft, however taking hyperspectral images from the air or from space has several limitations. Even in optimal circumstances, it is not possible to distinguish features on the sea floor or suspended matter beyond a depth of a few meters [17]. In water bodies with reduced visibility, the range is typically less than 1 m. Additional issues result from interference from the air between the water surface and the imager, for example due to clouds or Rayleigh scattering. It is also necessary to consider the angle of the sun in the sky. The spatial resolution of conventional remote sensing systems, such as a hyperspectral imager mounted in an airplane, is typically relatively low [17].

Potential applications of hyperspectral object identification are described by Johnsen [15]; these include mapping and monitoring of man-made structures such as seafloor pipelines for: material, crack, corrosion, and leakage detection; shipwrecks to determine the state of the structure, and artefacts of archaeological value. Environmental applications include monitoring the health of marine habitats such as coral reefs [18]. For nuclear applications, many of the techniques used in the monitoring of man-made structures under water could be adapted to the more stringent requirements of a nuclear environment.

Using emerging optical filter technology, hyperspectral imaging advanced to the forefront of low-cost technologies, which proves beneficial to applications where the imager is introduced to areas with higher risk such as nuclear environments where contamination with radioactive materials is a possibility. Reducing the cost of sensors to be deployed in these environments is of significant interest to the industry. Additionally, recently emerged hardware using linear variable filters and alternative hyperspectral processing techniques allows additional data to be collected with a single instrument. Whilst also reducing the cost of spectral imagers by up to 80% [19].

Underwater Raman

An alternate spectroscopic technique that could be utilized is Raman spectroscopy. Raman spectroscopy is a non-destructive technique that measures the molecular and crystallographic vibrational modes of materials [20]. The Raman effect arises due to the inelastic scattering of photons. As discussed above, when monochromatic radiation is incident upon a sample it is predominantly absorbed or elastically scattered (i.e. reflected). However, some inelastic scattering may occur if the photon is scattered with a different energy and consequently different wavelength. The most common cause for this is fluorescence, however, there is a small chance (approximately one in ten million) that photons may scatter due to interactions with molecular vibrational bonds in the material. It is this energy change that is measured in Raman spectroscopy.

A monochromatic light source, typically a laser, is used to illuminate the sample. When the spectrum of the scattered light is measured, not only is the incident radiation wavelength seen (Rayleigh scattering) but also, a small amount of radiation shifted to some different wavelength (Stokes and Anti-Stokes Raman scattering). These optical emissions arise due to interactions with the vibrational modes of the target molecules or crystals. The wavelength shift is a direct measure of the characteristic vibrational modes of the sample and thus provides valuable chemical and structural information [21]. Consequently, Raman spectroscopy can be used to glean information about material properties or as a tool for molecular and material identification[22].

Within aquatic environments, Raman spectroscopy has previously been limited in underwater applications due to difficulties of accurately and stably positioning of the optical focal spot on the sample by submersible vehicles for prolonged exposures needed to obtain meaningful measurements. Accordingly, many studies have been limited to transparent and translucent samples where stable positioning is less important [23]. A few studies have been conducted that used the Raman technique in deep marine environments. One such study [24] developed and successfully deployed a sea-going laser Raman spectrometer (DORISS). This system included a fully waterproof spectrometer that was used to conduct deep ocean (3600 m) geochemical studies. This included the in-situ identification of rocks and minerals on the sea floor and the chemical composition of pore water, gas seeps, and hydrothermal vents. However, an entire Raman system needed to be submerged because the extreme depths precluded a tethered solution. In shallow (<100m) marine settings, tethers are a practical possibility such that it is not necessary to submerge a whole spectrometer system.

As with any spectral technique the effect of the water must be accounted for when conducting Raman spectroscopy underwater. Primarily one must allow for additional Raman signals from the water and other constituents. However, these spectroscopic contributions are constant and can be simply determined by obtaining a reference background. An additional complication when conducting in-site Raman studies underwater is the generation of fluorescent contaminants. In particular, within many natural water systems there are organisms which contain chlorophyll and other fluorescent proteins which can produce intense fluorescent signals that mask the fainter Raman emissions.

METHOD

Raman and hyperspectral imaging experiments were conducted in outdoor swimming pools to demonstrate and assess functionality of the techniques. Selected objects were used in the pools for simulation purposes.

Photogrammetry and hyperspectral imaging experiments were conducted on a 1:5 scale spent fuel assembly skip. Data was collected by translating the camera over the skip in a raster pattern, ensuring adequate overlap between images in the sequence required for the photogrammetric reconstruction processes. Colour images for the photogrammetry models were captured with GoPro Hero 6 camera mounted in tandem with the hyperspectral camera. The hyperspectral imager used was a modified digital single lens reflex (DSLR) camera equipped with Linear Variable Filter (LVF) [19].

A robust low-cost (>£2000) standoff Raman probe was developed using off-the-shelf components and optics (Thorlabs, USA). The probe, connected to a laser source and spectrometer with an armoured optic fibre, was housed in an underwater housing (6” series, BlueRobotics, USA) with custom penetrators for the optical fibres as well as an uncoated broadband precision window (WG12012, Thorlabs, USA). The probe was then mounted at the bottom of an ROV (BlueROV2, BlueRobotics, USA), as shown in Figure 1.

Raman spectra were obtained for a range of different materials at depths of approximately 1m, with a laser power at sample of 150mW at 830 nm wavelength with an exposure time of 1s at a working distance of 3cm.

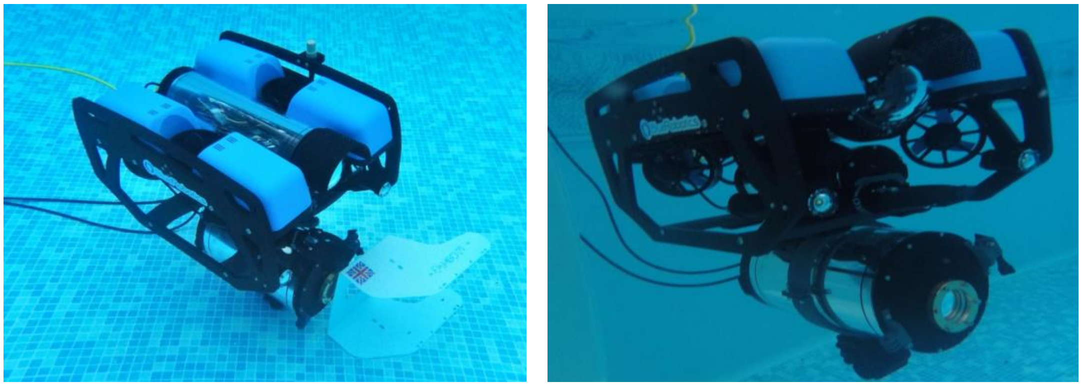

Fig. 1 - BlueROV with Raman cannon equipped, imaging a white powder coated aluminum panel.

DATA

Raman

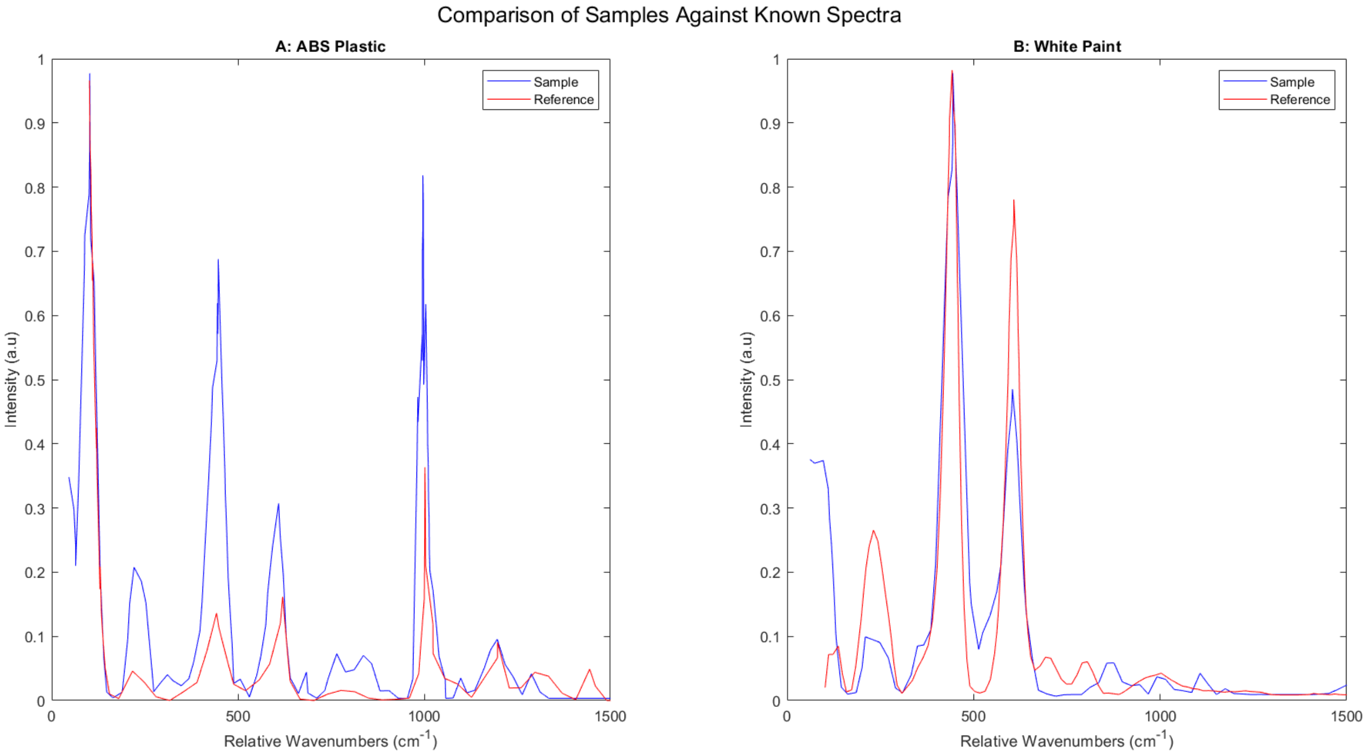

Fig. 2 - Raman spectra of samples A: Acrylonitrile butadiene styrene (ABS) panel compared against known spectra from Horiba Scientific (sample ID 1454). B: White powder coated aluminum panel compared against known spectra from RRUFF (sample ID 4641). Reference spectra obtained from Cowger et al, 2021[25].

Photogrammetry

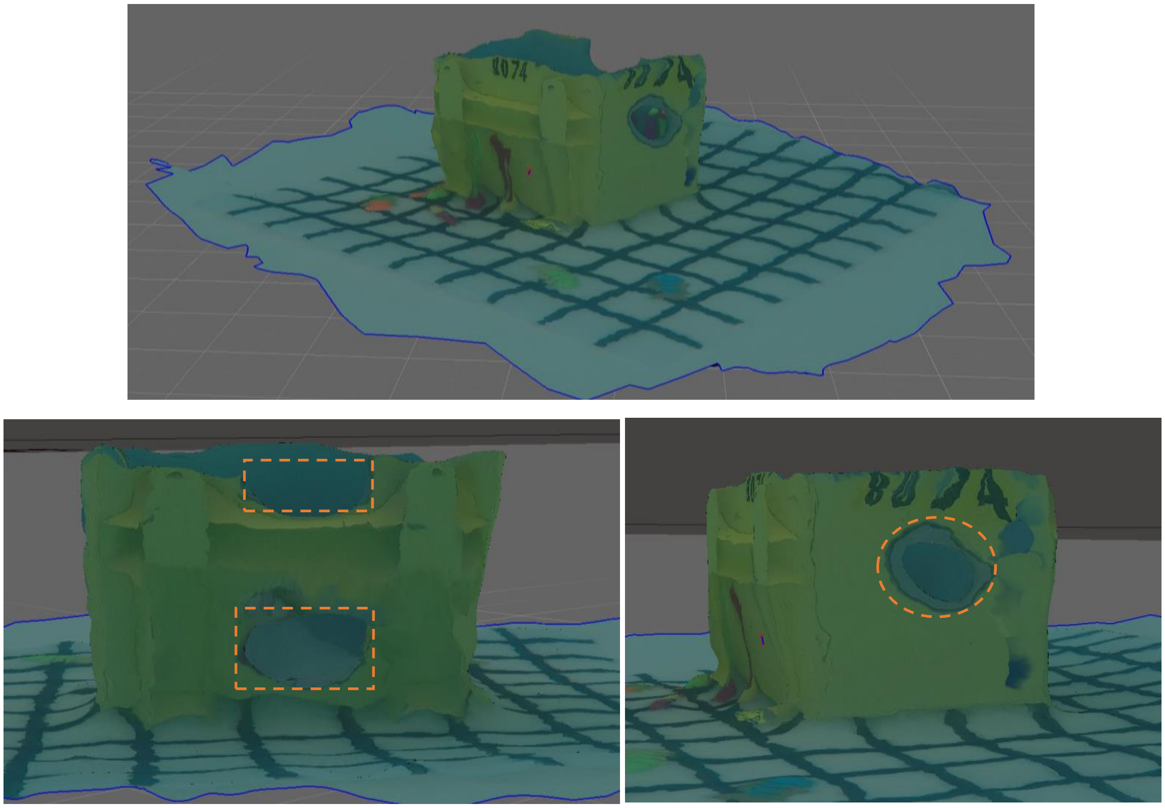

Fig. 3 - 3D reconstruction of the 1:5 scale nuclear fuel assembly skip outlining unique features such as holes cut out of the box clearly visible in the reconstruction.

Hyperspectral

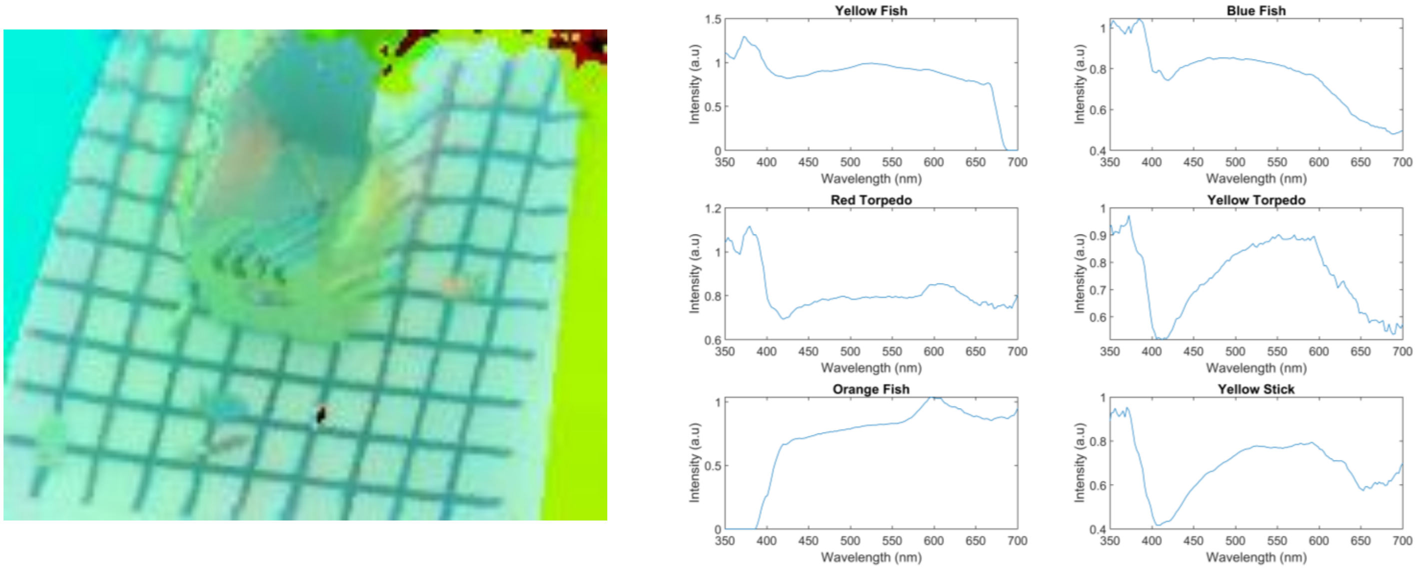

Fig. 3 - Left) hypercube image of the scene, Right) Raw spectra of objects identified from the hypercube scene.

DISCUSSION

The demonstrated techniques were able to acquire information about submerged objects in a simulated environment. The collected evidence suggests that the techniques could easily be transferred to nuclear fuel characterisation applications in spent fuel ponds. The demonstrated techniques are low-cost solutions, making the technology accessible for marine applications where such devices could not be previously applied because of prohibitive cost and/or risk of radioactive contamination.

Raman

Raman spectrums were successfully acquired with a ROV-mounted Raman probe. This spectral analysis technique reveals specific characteristics of the molecular composition of the material. A Pearson's correlation coefficient revealed an r value of 0.9 for the white paint compared against rutile (titanium dioxide) and an r value of 0.79 for the plastic sample compared against acrylonitrile butadiene styrene (ABS), indicating high confidence in the quality of spectra obtained.

Experimental observations revealed the optical attenuation of water at 830nm, and background light levels were darker than laboratory conditions despite operating outside in full daylight. This attenuation effect can be advantageous to achieve an improved signal-to-noise ratio.

Photogrammetry & Hyperspectral

Photogrammetry surveys can reveal defects in objects as shown in figure 4 where holes cut in the skip model are clearly visible and can be measured.

The generated hypercube, shown in Figure 4, outlines spectra of objects within the scene, which were successfully reconstructed using image stacks photographed by the LVF-DSLR. The data shown is raw, meaning it has not been processed to correct for the attenuation of light through water.

Due to the thin (3 mm thick) metal of the skip model, the reconstruction software (Agisoft Photoscan) struggled to recreate crisp edges of the skip. This problem would not be seen in real world applications where the metal sides are much thicker, which in turn improves reconstructions as more points of reference would be available to the image stitching process

CONCLUSION

The optical techniques presented all gathered data that would be advantageous for characterising materials in spent fuel ponds and assessing their state. In the case of disused spent fuel ponds, it has the potential to aid the planning phase of the decommissioning process. The advantage of the techniques field tested in this work is that they are all low-cost solutions, whereas the traditional cost associated with these techniques used to be prohibitively expensive for deployments in high-risk environments such as spent fuel ponds.

Mounting the optical hardware on ROVs allows for risk to be minimized by removing the human element in these environments. Traditionally, teams of divers are employed for these types of tasks, and although water attenuates the ionizing radiation emitted by spent fuel, it is still not without its inherent risks. The use of robots reduces human radiation exposure to zero.

It should be noted that optical imaging techniques are limited by the water conditions such as clarity/turbidity which have an adverse effect on the ability to deploy these techniques. The presented techniques are therefore not a solution to all spent fuel pond environments, and suitability assessments must be made on a case-by-case basis.

REFERENCES

[1] M. Wareing, “Cleaning up our nuclear past: faster, safer and sooner,” 2019. [Online]. Available: https://nda.blog.gov.uk/2019/06/07/turning-the-corner-to-exciting-times/. [Accessed: 11-Aug-2020].

[2] J. S. Jaffe, K. D. Moore, J. McLean, and M. P. Strand, “Underwater optical imaging: status and prospects,” *Oceanography*, vol. 14, no. 3, pp. 66–76, 2001.

[3] J. Teague, M. J. Allen, and T. B. Scott, “The potential of low-cost ROV for use in deep-sea mineral, ore prospecting and monitoring,” *Ocean Engineering*. 2018.

[4] R. A. Davis, *Oceanography: an introduction to the marine environment*. William C Brown Pub, 1991.

[5] N. G. Jerlov, “Chapter 7 Theory of Radiative Transfer in the Sea,” in *Marine Optics*, 2nd ed., vol. 14, Amsterdam: Elsevier, 1976, pp. 83–100.

[6] R. C. Smith and K. S. Baker, “Optical properties of the clearest natural waters (200--800 nm),” *Appl. Opt.*, vol. 20, no. 2, pp. 177–184, 1981.

[7] N. G. Jerlov, *Optical oceanography*, vol. 5. Elsevier, 2014.

[8] I. Colomina and P. Molina, “Unmanned aerial systems for photogrammetry and remote sensing: A review,” *ISPRS Journal of Photogrammetry and Remote Sensing*. 2014.

[9] J. Teague and T. B. Scott, “Underwater Photogrammetry and 3D Reconstruction of Submerged Objects in Shallow Environments by ROV and Underwater GPS,” *J. Mar. Sci. Res. Technol.*, vol. 1, no. 005, 2017.

[10] G. Johnsen, “Kelp forest mapping by use of airborne hyperspectral imager,” *J. Appl. Remote Sens.*, 2007.

[11] G. Johnsen *et al.*, “Underwater hyperspectral imagery to create biogeochemical maps of seafloor properties,” in *Subsea Optics and Imaging*, 2013.

[12] W. M. Klonowski, “Retrieving key benthic cover types and bathymetry from hyperspectral imagery,” *J. Appl. Remote Sens.*, 2007.

[13] H. M. Dierssen, “Overview of hyperspectral remote sensing for mapping marine benthic habitats from airborne and underwater sensors,” in *Imaging Spectrometry XVIII*, 2013.

[14] P. Mouroulis *et al.*, “Portable Remote Imaging Spectrometer coastal ocean sensor: design, characteristics, and first flight results,” *Appl. Opt.*, 2014.

[15] G. Johnsen, M. Ludvigsen, A. Sørensen, and L. M. Sandvik Aas, “The use of underwater hyperspectral imaging deployed on remotely operated vehicles - methods and applications,” *IFAC-PapersOnLine*, 2016.

[16] R. Pettersen, G. Johnsen, P. Bruheim, and T. Andreassen, “Development of hyperspectral imaging

as a bio-optical taxonomic tool for pigmented marine organisms," *Org. Divers. Evol.*, 2014.

[17] G. Johnsen, "Underwater hyperspectral imaging." Google Patents, 2013.

[18] J. Teague, J. Willans, M. Allen, T. Scott, and J. Day, "Hyperspectral imaging as a tool for assessing coral health utilising natural fluorescence," *J. Spectr. Imaging*, 2019.

[19] J. Teague, D. Megson-smith, M. J. Allen, J. C. C. Day, and T. B. Scott, "A review of optical techniques for coral monitoring & introducing low-cost hyperspectral imaging .," *Preprints*, no. May, pp. 1–19, 2021.

[20] H. J. Bowley, D. L. Gerrard, J. D. Louden, and G. Turrell, *Practical raman spectroscopy*. Springer Science & Business Media, 2012.

[21] M. S. Dresselhaus, G. Dresselhaus, R. Saito, and A. Jorio, "Raman spectroscopy of carbon nanotubes," *Physics Reports*. 2005.

[22] S. N. White, R. M. Dunk, E. T. Peltzer, J. J. Freeman, and P. G. Brewer, "In situ Raman analyses of deep-sea hydrothermal and cold seep systems (Gorda Ridge and Hydrate Ridge)," *Geochemistry, Geophys. Geosystems*, 2006.

[23] S. N. White *et al.*, "Development and deployment of a precision underwater positioning system for in situ laser Raman spectroscopy in the deep ocean," *Deep. Res. Part I Oceanogr. Res. Pap.*, 2005.

[24] P. G. Brewer *et al.*, "Development of a laser Raman spectrometer for deep-ocean science," *Deep. Res. Part I Oceanogr. Res. Pap.*, 2004.

[25] W. Cowger *et al.*, "Microplastic Spectral Classification Needs an Open Source Community: Open Specy to the Rescue!," *Anal. Chem.*, vol. 93, no. 21, pp. 7543–7548, Jun. 2021.

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