THIN LAYER ACTIVATION FOR MATERIALS RESEARCH & DEVELOPMENT AT IAM OF THE EUROPEAN COMMISSION

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Abstract

After a short description of the cyclotron of the European Commission Institute for Advanced Materials (IAM)-Ispra site (Italy) and its activities, an overview of the Thin Layer Activation (TLA) technique is given. The use of TLA for materials studies is illustrated with examples of ongoing research within the IAM. Notably this includes corrosion studies and laboratory and industrial wear testing. Due to its specific properties, including area selectivity, high sensitivity and speed in addition to the possibility for in-situ and on-line monitoring, TLA can contribute significantly to monitor surface degradation in both research and industrial applications.

1. Introduction

The cyclotron of the Institute for Advanced Materials (IAM)-Ispra site of the European commission is a Scanditronix variable energy MC-40 machine. It accelerates protons and alpha particles up to approximately 39 MeV, 3He2+ to 53 MeV and deuterons up to 19 MeV. Depending on the target and the accelerated particles, the external beam intensity can reach 60 μA. The cyclotron facilities include three irradiation halls with six beamlines fully equipped and several dedicated laboratories. The construction of a new building, directly besides the cyclotron is fully dedicated for engine testing using also cyclotron activation.

At present, the main activity of the IAM cyclotron is the application of the Thin Layer Activation (TLA) technique with all its methods for a wide range of wear and corrosion studies. Additionally, isotope production for bio-medical applications, especially I-123 and the production of radiotracers for environmental studies in collaboration with other institutes of the European Commission is undertaken.

In this paper, a description of the TLA technique is given. Due to its specific properties, TLA can contribute significantly to monitor surface degradation in both research and industrial applications. Besides using TLA for wear testing both for industrial purposes as well as for the development of wear resistant materials, the IAM is also active in improving its application and extending it to new fields. In this area, it benefits strongly from its combination of materials oriented research and the possession of a cyclotron. This is illustrated with some examples of ongoing research within the IAM.

2. Principle of TLA

The research in the material field is continuously improving and consequently the associated test methods are forced to be of increasing performance. Beside numerous applications of radioactivity in research and industry such as medical (treatment and diagnostic), chemical analysis, environment surveillance, radioactivity is since a long time welcome in material research. TLA is one of the promising techniques employing radiotracers for material performance studies.

The principle basis of TLA is the creation of an appropriate radionuclide in a given material to a well defined depth of a selected area [1,2]. This activation is realised by exposure of the component to a charged particle beam using e.g. a cyclotron facility. If the material is subjected to mechanical or chemical degradation such as wear or corrosion, any loss of the activated material will result in a loss in radioactivity of the activated component. This reduction in signal can be directly related to mass loss or thickness reduction; the removed material debris, collected in a filter or fluid sample, also gives a measure of material degradation. The specific properties of TLA [2] include area selectivity, high sensitivity, speed and applicability as a non-contact in-situ

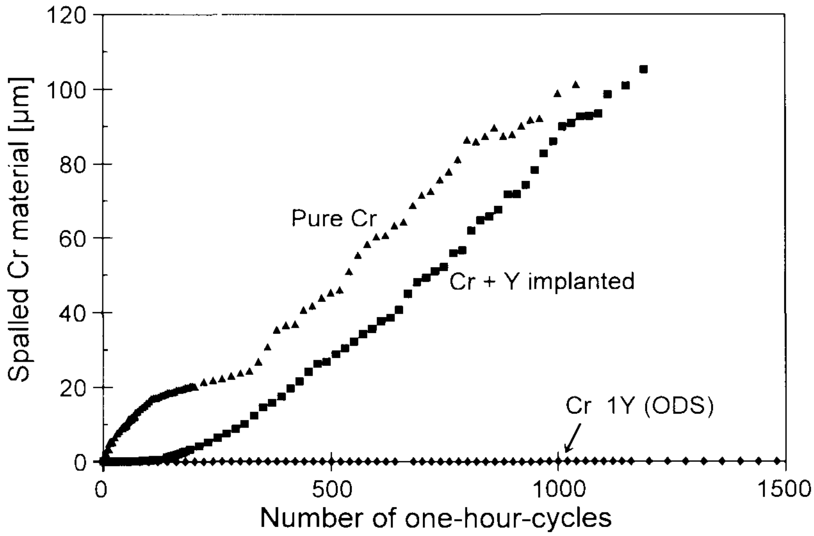

Fig. 1 Spallation behaviour measured using TLA of pure Cr, Cr implanted with 10^{17} Y-ions/cm^2, and Cr-1%Y_2O_3 alloy upon thermal cycling at 1000°C [7].

method. Due to its specific properties, TLA can contribute significantly to monitor surface degradation in both research and industrial applications. The technique itself has been used for some time, especially for wear testing related to automotive applications [e.g. 3,4]. In first instance, the cost of TLA might look high since it needs a charged particle accelerator and nuclear instrumentation such as gamma spectrometer for data acquisition and processing. Nevertheless, the profitability of the method is strongly based on its advantages outlined above and related to this the time gain. Psychological aspects related to the use of radioactivity hampers the application of TLA. This despite the fact that the activity used in TLA studies is in the range of 10 µCi (370 kBq). Additionally, the radionuclides used in TLA are normally gamma emitters (no neutrons, no alphas), and the normal half life times are in general only few months, therefore the risk of contamination is minimal. In addition only small thicknesses of the material are activated, due to the high stopping power of charged particles.

3. Examples of TLA applications at the IAM

3.1. Corrosion at high temperatures

The protection of alloys and coatings against high temperature corrosion is generally related to the formation of a dense, adherent and protective oxide scale of, e.g. Cr_2O_3, SiO_2 or Al_2O_3 [5]. Besides the chemical properties, the mechanical properties of these scales are also of considerable importance. Service stresses, as generally encountered under operating conditions, may undermine the scale stability causing cracking and spallation, leading e.g. to accelerated corrosive attack of the underlying material. One of the commonest sources of stress is due to thermal cycling as a consequence of the difference in thermal expansion coefficients of the oxide scale and base material. Spallation of protective surface layers due to thermal cycling is a significant problem in a wide range of technological applications, for example coatings on gas turbine blades, heat exchanger applications and oxide fuel cells. The resistance of materials to thermal cycling is in general determined from cyclic tests, in which a sample is exposed at a test temperature which is changing with time in a periodic way, often simulating the actual conditions. A typical thermal cycle might comprise heating of a sample at 1000°C for 1 h, cooling the sample to room temperature and maintaining this lower temperature for 10 min.

The application of TLA to cyclic oxidation testing was developed at the IAM and described in detail elsewhere [6,7]. The principle consists of measuring the activity of the spalled material relative to the original activity of the sample. The contribution of the method in this application is especially important due to its high sensitivity, and area selectivity. As an example Fig. 1 shows the influence of yttrium additions on the cyclic oxidation behaviour of chromium at 1000 °C [7]. The yttrium was added by alloying and ion implantation. Clearly, the addition of yttrium improves the performance of the Cr. Using conventional techniques, the quantification of this effect could only be reached with great difficulty.

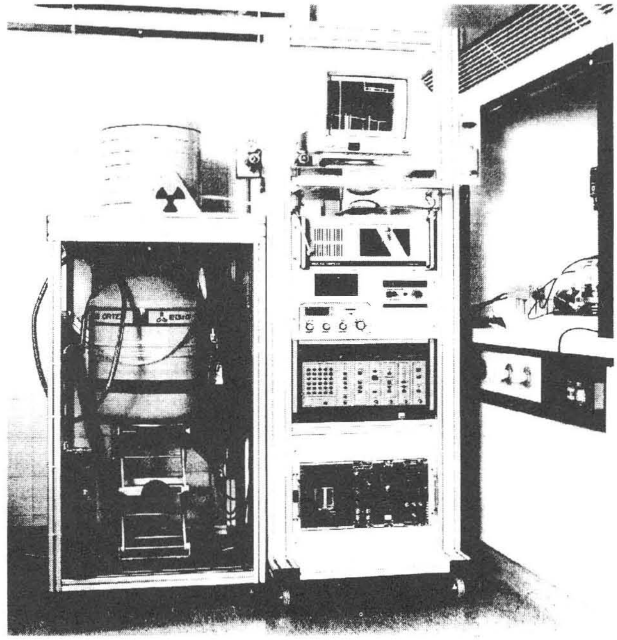

Fig. 2 : Test facility for electrochemical testing using conventional methods and TLA.

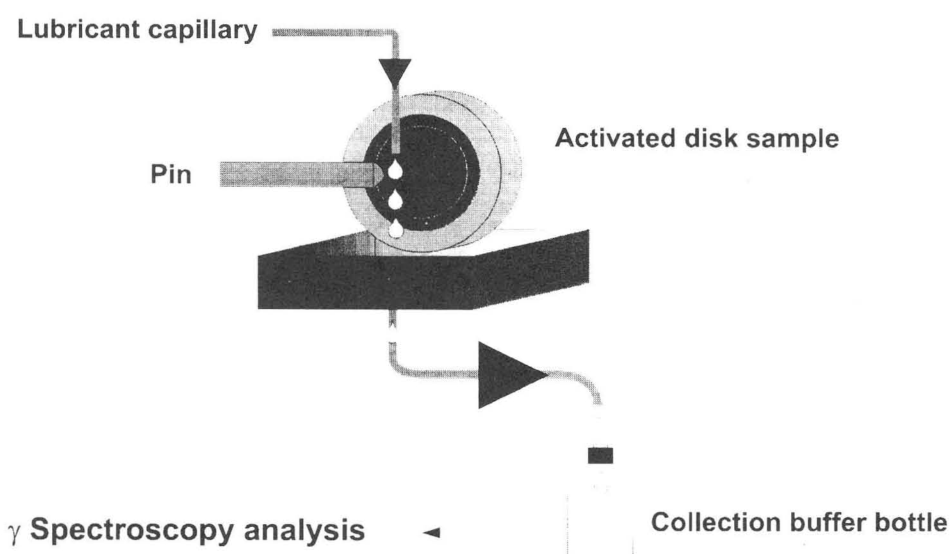

Fig. 3 : Schematic diagram of the test facility for lubricated pin on disk wear testing using TLA.

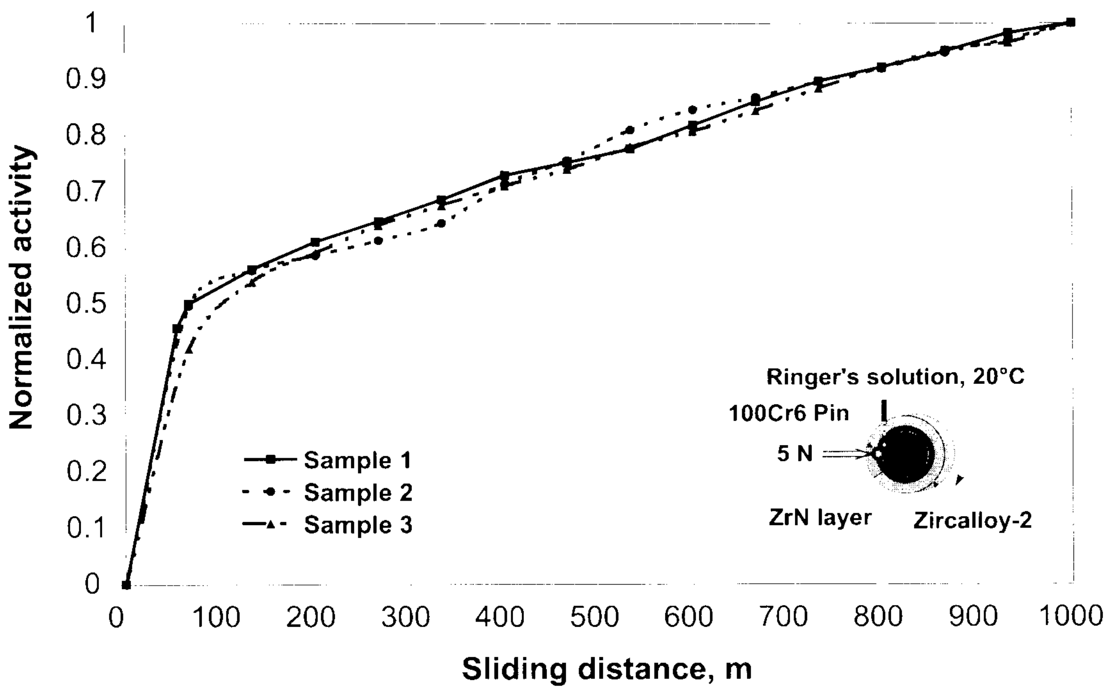

Fig. 4 : Normalised summed activity in collection buffer bottle versus sliding distance during wear testing of 3 different samples of nitrided Zircalloy-2.

3.2. Electrochemical testing and industrial plant reliability monitoring

Also in the field of electrochemical corrosion, TLA might significantly contribute to a better scientific understanding of the underlying processes. The technique is complementary to conventional methods especially due in addition to its high sensitivity, area selectivity, to the testing speed and the possibility to follow on-line the release of individual elements. Fig. 2 shows an experimental facility integrating TLA with advanced conventional techniques. This facility is used among others at IAM to study a wide range of corrosion phenomena of practical importance.

Efficient monitoring of corrosion in industrial equipment is essential for an efficient and safe operation, to allow for optimal maintenance and prevent unscheduled shutdowns. The costs of unscheduled shutdown of large chemical processing facilities, nuclear power plants and nuclear reprocessing plants might be several millions ECU per day. TLA might be a suitable technique for the reliable assessment of technical components in several applications.

One of the ongoing research projects at the IAM within the European Coal and Steel Commission (ECSC) programme of the European Commission, is the Assessment of Plant and Structure Corrosion Reliability by TLA. Several European partners are involved in the project with the following objectives:

- to develop the TLA technique for use in various industrial applications with particular corrosion conditions in which the use of conventional methods is difficult;
- to improve the quality of monitoring as compared to current techniques, by obtaining more reliable data concerning specific parts of a structure which will allow for better corrosion control;
- to improve the reliability of steel structures by a continuous monitoring technique;
- dissemination of results.

Fig. 5 : Artistic drawing of the TLA/engine test facility.

3.3. Laboratory wear testing

Using the TLA technique in wear studies can lead to a significant reduction in test time. Among others, it can significantly contribute to studies for the development of very wear resistant materials, such as bio-materials [8,9]. Fig. 3 represents the use of TLA in pin-on-disk wear testing. The tribometer uses a pin which runs with a controlled speed and normal force over the test sample, which has previously been activated. The lubricant which is flowing continuously over the vertically placed sample, removes the material debris into a collection buffer bottle. After a pre-determined sliding distance, the collection buffer bottle is removed to determine its radiation intensity as a function of the γ-energy. The wear test is continued with a new collection buffer bottle. As an example a study of the potential use as a biomaterial for surgical implants of ZrN coatings was undertaken [9]. Zirconium has an excellent biocompatibility and could be used for biomedical applications. However, its tribological properties are poor and in order to overcome this disadvantage, a 5 μm thick ZrN coating was prepared by thermal nitridation of a Zr-based alloy. The wear was simulated with a pin spherically ended with 100Cr6 hardened steel ball applying 5 N normal force with 0.1 m/s sliding speed. Coated Zr-based alloy disks with a diameter of 40 mm and thickness of 8 mm were activated with deuterons on a specific circular area of the sample. For this study the Nb-92m nuclide activity was used as an indication for the wear losses. In order to assess the reproducibility of the technique, the experiment was carried out on three samples up to a sliding distance of 1000 m. The variations of the activity of the collection buffer bottles versus the sliding distance are shown in Fig. 4. This indicated on one hand that the reproducibility of the data is very high and on the other that the TLA measurements are indeed orders of magnitude more sensitive than those obtained by conventional gravimetry [9].

3.4. Industrial wear testing

At present, the major use of TLA is in engine wear testing, including for testing of new lubricants and fuels. The majority of the studies employing TLA have close practical relation to the development of new products and for this reason are not published. Concerning engine wear testing, the performance of most components subject to wear such as pistons, folders, and camshafts can be monitored. TLA in engine testing benefits from its area selectivity, high sensitivity, high speed response time and applicability as a non-contact in-situ method. The method can significantly reduce the number and duration of the tests and thereby save considerable amounts of time and money on repetitive studies and inspection procedures. Fig. 5 represents an artistic view of the dedicated building for engine testing, directly next to the cyclotron buildings. It is recently equipped with a new engine test bench. The facility will be operational in the near future.

4. Concluding remarks

TLA is a very efficient method to detect and measure material degradation caused by such processes as wear and corrosion, in research as well as in industrial plant monitoring. Due to its specific properties, TLA can contribute significantly to a better scientific understanding of surface degradation processes. Additionally, its use as an on-line monitoring technique can contribute to reliable assess the service life of technical components and thus contribute to an efficient and safe operation of a wide range of industrial facilities.

The use of TLA is mainly in the field of wear testing related to applications in the automotive industry. The technique, however, shows an enormous potential for diversification and further development, as shown by some examples of ongoing work at the IAM of the European Commission. Its use is limited due to the restricted number of dedicated facilities available and unfamiliarity with the technique. Synergy between materials research and nuclear applications of accelerators is a necessity for a more extensive exploitation of the possibilities of TLA in materials technology.

References

[1] - T.W. Conlon, Contemp. Phys. 26 (1985) 521.

[2] - M.F. Stroosnijder, in "Application of Particle and Laser Beams in Materials Technology", P. Misaelides (ed.), Kluwer Press, 1995, p 399.

[3] - T. Kosako and K. Nishimura, Nucl. Instr. And Meth. B56/57 (1991) 900.

[4] - Fehsenfeld, A. Kleinrahm and H. Schweickert, J. Radianal. Nucl. Chem. Art. 160 (1992) 141.

[5] - M.F. Stroosnijder, R. Mevrel and M.J. Bennett, Materials at High Temperatures 12 (1994) 53.

[6] - M.F. Stroosnijder, V.A.C. Haanappel and I. Murris, Materials Science Forum Vols, 251-254 (1997) 275.

[7] - M.F. Stroosnijder and G. Macchi, Nucl. Instr. and Meth. B100 (1995) 155.

[8] - W. Neumann, W. Woelfli, P. Heimgartner and R.M. Streicher, Nucl. Instr. And Meth. B50 (1990) 57.

[9] - C. Brugnoni, F. Lanza, G. Macchi, R. Müller, E. Parnisari, M.F. Stroosnijder and J. Vinhas, Surf. Coating Technology in press.