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# In situ mechanical, temperature and gas exposure treatments of materials combined with variable energy positron beam techniques

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**Abstract**

An overview is given of the extension of the Delft variable energy positron (VEP) beam facility with equipment for in situ heating, cooling, 4-point bending, hydrogen permeation and gas ad- and absorption of bulk materials, surfaces and interfaces.


*Keywords:* Positron beam; Heat treatment; Permeation; 4-Point bending

## 1. Introduction

The manufacturing of new materials specifically tailored for industrial and research applications requires a thorough knowledge of and control over the different processing steps involved. A few examples of such materials are protective and wear resistance polymer and ceramic coatings, nonlinear optical materials, nano-structured materials and polymers for photovoltaics. Typical fabrication steps are material deposition and sputtering, and modification by ion-implantation, sintering and thermal treatments. On the other hand, the successful use of these materials may be affected by the environmental conditions such as heat, light, gases and mechanical stresses, under which they must retain their properties.

In order to assess the effects of some of the above mentioned processing steps and conditions with low energy positrons, the Delft variable energy positron (VEP) beam has been extended with equipment for a number of in situ treatments in combination with 1D and 2D Doppler Broadening studies. These include a high temperature oven, a closed loop cryostat, a 4-point bending device, a permeation cell, and gas handling system. The design details and performance characteristics as well as a short description of the existing positron beam setup will be given in the following paragraphs.

## 2. The VEP beam

The Delft VEP beam [1], in operation for more than 10 years now, utilizes $^{22}Na$ as positron source (Amersham, X1055), with a present activity of about 1 GBq. The emitted positrons are transmission moderated in a 2 μm thick poly-crystalline tungsten foil with an

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efficiency of approximately $2 \times 10^{-4}$. Transport of the moderated positrons from the source area to the accelerator and target is achieved by a $10^{-2}$ T axial magnetic field. An over 45° bent section with magnetic drift compensation is used as energy filter. At the target position the beam has an intensity of $2 \times 10^5$ e$^+$ s$^{-1}$ and a diameter of about 8 mm. The positron energy can be varied from several eV up to 30 keV. The facility is equipped with two Ge detectors for 1D or 2D Doppler Broadening studies and Positronium (Ps) fraction measurements. An $E \times B$ filter and retarding field grids placed in front of the target are used for energy selective detection of remitted positrons.

## 3. High temperature studies

In order to in situ study the generation of thermally induced defects and the formation of nano-cavities and metal nano-clusters in ceramic materials such as MgO and spinel, a high temperature oven has been constructed (see Fig. 1). It consists of a 1 mm diameter tungsten wire wrapped around a hollow, 60 mm long boron nitride (BN) cylinder with an inner diameter of 20 mm. Samples are pre-mounted in a ring shaped holder which is inserted at the center of the cylinder. Reduction of radiative heat loss is achieved by three thin (0.5 mm) cylindrical tungsten radiation shields positioned around the central BN cylinder. The front and back sides of the radiation shields contain circular holes with a diameter of 20 mm. The positron beam enters through the front side opening and in case of samples smaller than the beam diameter the nonintercepted beam leaves the oven through the back side opening and is dumped at the end-flange of the beam line at a distance of about 40 cm from the Ge detectors. The oven is supported by two 6 mm in diameter, 25 cm long Mo rods which are connected to two high current feedthroughs welded to the 6 in. top flange of the target chamber. The outer dimensions of the oven, including the shields, are such that it can be inserted into the target chamber through a 10 cm inner diameter vacuum tube. The temperature is measured by a W/Re thermocouple located at the center of the oven or by an optical pyrometer. First tests have shown that

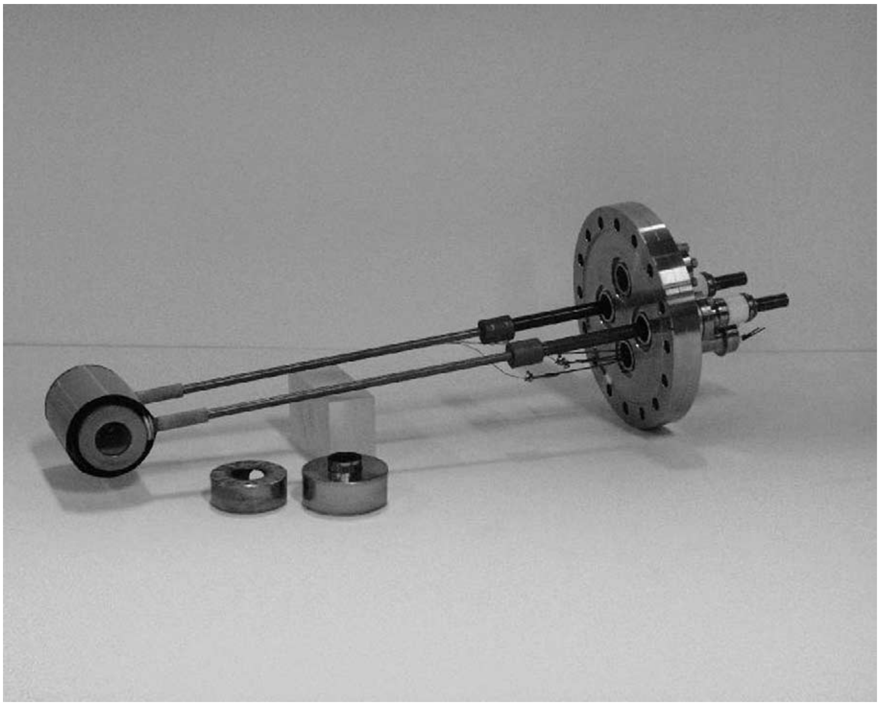

Fig. 1. A photograph of the high temperature oven. For clarity two of the front radiation shields are removed.

stable temperature of 1800 K can be reached at a DC current of 20 A (800 W). The magnetic field induced by the heating coil at the position of the target amounts to $5 \times 10^{-4}$ T/A and is parallel to the main axial field. Therefore, at the maximum temperature reached so far, no disturbance of the incoming positron beam is observed. This oven has been successfully applied to observe the formation of thermally induced defects in a Cu single crystal. A recently obtained Varian 350, 180 keV ion implanter is in use for ex situ implantation of He, Ar, Li etc. ions as a first process step for the formation of nano-cavities and metal clusters.

## 4. Low temperature studies

In order to perform positron beam experiments at low temperatures we have modified an APD Displex DE-20 closed loop cryostat. The modification concerns the extension of the cold finger and the radiation shield and insertion of a heating stage at the target position for rapid heating of the samples. The temperature of the sample can be measured by an additional chromel/alumel thermocouple. Samples can be mounted electrically insulated from the cold finger in order to study for example electric field and temperature dependent Ps formation in e.g. SOI (silicon on insulator) samples. The modifications of the cold finger of the cryostat are shown in Fig. 2. In case of samples smaller than the beam diameter the radiation shield can be removed in order to dump the not intercepted positrons at the end of the beam line. With this system stable target temperatures in the range from 15 to 600 K have been achieved.

## 5. In situ bending studies

A 4-point bending device (Kamrath and Weiss, GmbH), to be used at the future electrostatic positron beam [2], is adapted for use in the VEP. The device is shown in Fig. 3. It is used to study the effects of both tensile and compressive stresses on the adhesion and defect formation in thin coatings by Doppler Broadening experiments. The maximum load and displacement of this system is 200 N and 5 mm, respectively. An example of such a study is a 500 nm thin poly-(methyl methacrylate) (PMMA) layer spin coated on a

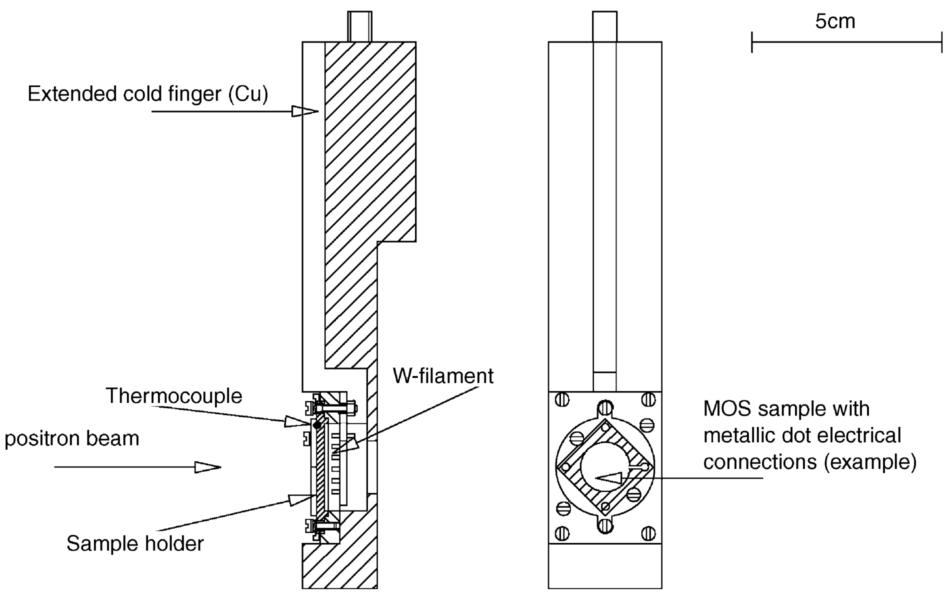


Fig. 2. Schematic drawing of the extended cold finger of the APD Displex DE-20 closed loop cryostat. Left: side view which indicated the sample holder, the position of the chromel/alumel thermocouple and tungsten filament for rapid heating from 15 to 600 K. Right: front view showing as an example an Al–$SiO_2$–Si (MOS) sample with electrical connections for studying temperature and electric field dependent Ps formation in $SiO_2$.

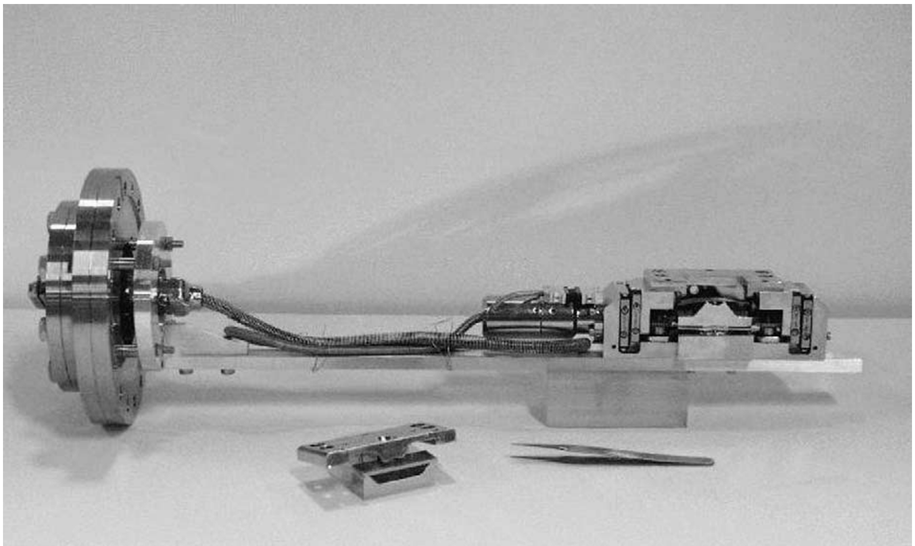

Fig. 3. A photograph of the adapted 4-point bending device (Kamrath and Weiss, GmbH). In the foreground the auxiliary parts for bending under compressive stress is shown.

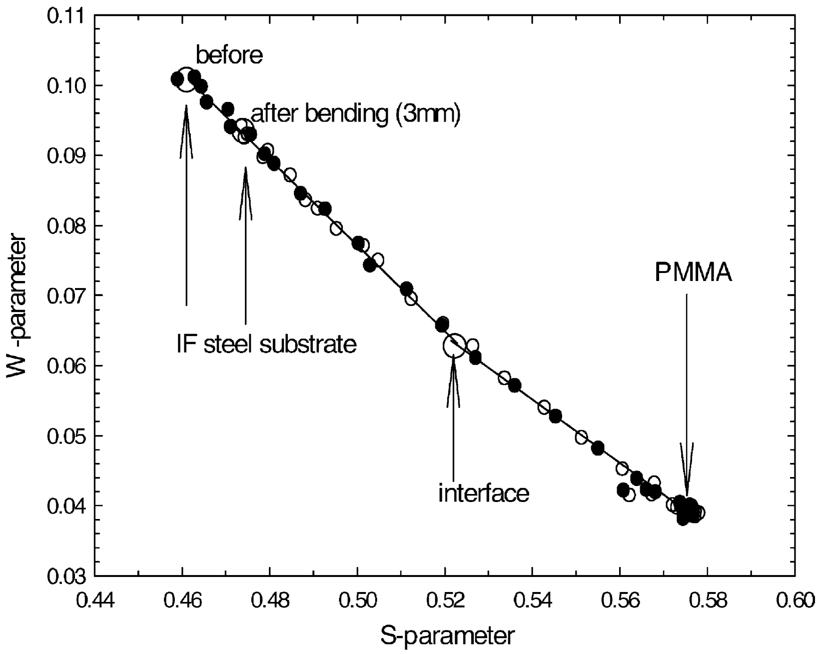


Fig. 4. The *S*–*W* map for a PMMA coating on steel for the as spin coated layer (•) and after 3 mm bending in tensile mode (○). The positron energy is the running parameter and increases in the direction from the PMMA layer towards the steel substrate. The characteristic *S*, *W* coordinates are indicated by the large open symbols (◯).

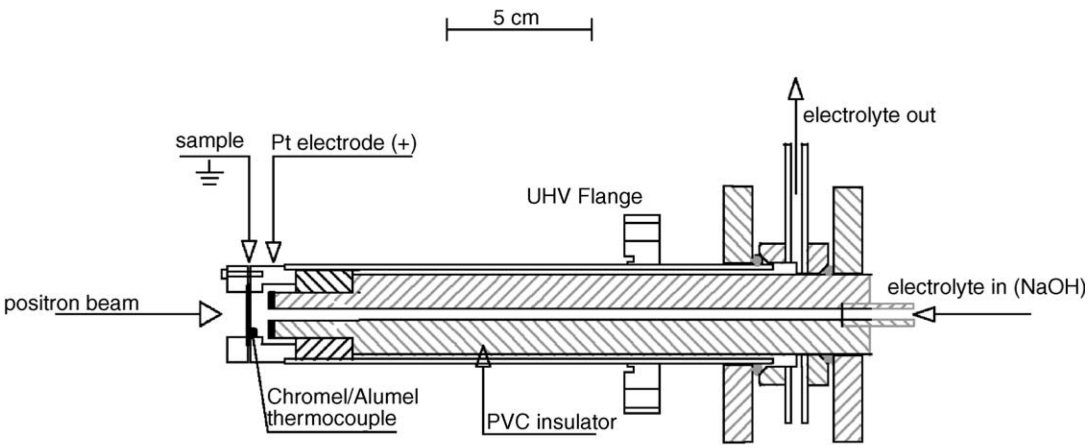


Fig. 5. A schematic drawing of the electrochemical permeation cell.

low carbon content interstitial free (IF) steel substrate with a cross-sectional area of $2.5 \times 0.9\ cm^2$ [3]. The result of the combined *S* and *W* Doppler Broadening measurements on a as spin coated and 3 mm bend sample is shown in Fig. 4. It was observed that only the substrate was affected by the bending. No changes in the spin coated layer or the interface were detected, indicating that under these conditions the polymer seems to follow the substrate during deformation.

## 6. Hydrogen permeation cell

A well known method to study the diffusion and trapping of hydrogen in (industrial) steels is the so-called permeation technique. In this technique one face of a thin (steel) sample is exposed to hydrogen while the other side faces an ultra high vacuum (UHV) system equipped with apparatus for hydrogen detection such as a quadrupole mass analyzer or a sensitive membrane manometer. Hydrogen can be supplied in the form of a molecular gas ($H_2$, gas driven permeation) or as atomic hydrogen. The latter occurs for instance when an acid is used (pickling driven) or in case of electrochemical hydrogen loading. Dankert has applied these techniques to study the hydrogen pressure build-up in blowholes in IF steels [4]. Based on these concepts an electrochemical loading cell has been constructed for the detection of hydrogen arriving at defects (voids) or interfaces using the positron beam Doppler Broadening techniques. An overview of this system is shown in Fig. 5. Basically it consists of a tube which on one side is closed-off by the thin metallic sample to be studied. A sheet of platinum mounted on an insulating tube made of PVC is facing the sample surface. In this setup the sample and platinum sheet act as the cathode and anode, respectively. A suitable electrolyte (NaOH) is continuously circulating through the small volume created by the anode and cathode. A chromel/alumel thermocouple is attached to the sample for control of temperature during the electrochemical loading process. This interior is welded to a standard UHV flange which can be mounted horizontally at the end of the positron beam line. The positron beam is hitting the opposite sample face and the arrival of hydrogen in sub-surface voids or near surface interfaces is detected by monitoring the changes in the *S* and *W* parameter.

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