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HIGH VOLTAGE PERFORMANCE DEGRADATION OF THE 14UD TANDEM ACCELERATOR*

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Abstract

The 14UD at the Australian National University's Heavy Ion Accelerator Facility (HIAF) operated at a maximum voltage of 15.5 MV after the installation of tubes with a compressed geometry in the 1990s. In recent years, the performance of the accelerator has shown a gradual decline to a maximum operation voltage of ~14.5 MV. There are some fundamental factors that limit the high voltage performance, such as SF_6 gas pressure, field enhancement due to triple junctions and total voltage effect. In addition, there are non-fundamental factors causing high voltage degradation. These are: operation with faulty ceramic gaps; operation at inappropriate voltage and SF_6 pressure combinations; SF_6 leaks into the vacuum space; use of SF_6 and O_2 as a stripper gases; poor electron suppression in the high energy stripper and frequent use of highly reactive ions such as sulphur and fluorine. In this paper we will discuss factors that limit the high voltage performance. The main outcomes of a preliminary investigation of titanium (Ti) electrodes removed from the accelerator after a few decades of operation will be reported. The investigation confirmed contamination of Ti electrodes with unstable films containing traces of oxides, sulphur and fluorine. The rehabilitation strategies for the accelerator will be discussed.

INTRODUCTION

The Heavy Ion Accelerator Facility (HIAF) at the Australian National University (ANU) operates a National Electrostatics Corporation (NEC) 14UD pelletron tandem electrostatic particle accelerator [1]. This accelerator has been in operation for over forty years after the first successful experiment in 1974. The original configuration used corona voltage distribution system, in which a maximum voltage of 14.8 MV was achieved for experiments in 1981. However, there was a consistent deterioration from this level that eventually lead to an examination of aging effects [2] and a major accelerator upgrade in around 1990 [3].

This upgrade was comprised of two components. The first was the installation of "compressed geometry" acceleration tubes that removed dead space and allowed the installation of additional acceleration tubes thereby reducing the field across each insulating gap. The second replaced the corona voltage distribution system that was causing corrosion of accelerator components [4] with a resistor grading system. With these upgrades, a peak conditioning voltage of 16.7 MV was achieved, with experiments performed at 15.5 MV. However, the maximum voltage available for experiments has now degraded to around 14.5 MV.

Since January 2016, the 14UD has had voltage on its terminal for 56% of the total time, with 16% of the time spent on major accelerator maintenance and repairs. Of the time with terminal volts, 27% has been with terminal volts above 13.5 MV.

Even with continued operation near or above the original design voltage of the 14UD, user demand for both operational time and even higher terminal voltages is increasing. Therefore, the degradation mechanisms of the 14UD should be understood in order to extend the useful life of the accelerator.

HIGH-VOLTAGE BREAKDOWN

Mechanical issues in an electrostatic accelerator can be readily dealt with, but it is the electrical breakdown mechanisms of the insulating gas and vacuum spaces that determine the limits on high-voltage performance. A thorough review of the mechanisms – and the damage created – is provided in [5] and discussed here with specific reference to the ANU 14UD.

SF_6 Breakdown

The insulating space between the accelerator column and the containing pressure vessel of the 14UD is filled with approximately 22 tonnes of sulphur hexafluoride (SF_6) gas at a pressure of 100 psia. Although considered to be inert, chemical reactions during and after electrical breakdown of SF_6 create long-lived toxic and corrosive by-products [6, 7].

The basic reaction scheme, even in high purity SF_6, is [8]:

$$SF_6 \rightarrow SF_4 + 2F \quad (1)$$

$$SF_4 + H_2O \rightarrow SOF_2 + 2HF \quad (2)$$

$$SOF_2 + H_2O \rightarrow SO_2 + 2HF \quad (3)$$

$$2F + M \rightarrow MF_n \quad (4)$$

where M is any exposed metal such as titanium electrodes or aluminium structural components. The breakdown product yield is influenced by the SF_6 pressure, H_2O content and spark discharge energy [6, 9].

In the era of corona voltage distribution in the 14UD, hydrofluoric acid (HF) products in particular may have caused repeated fracturing of the nylon links in the pelletron chains [4]. A move to resistor voltage distribution configurations ameliorated the problem, but the use of corona terminal voltage stabilisation and arc discharges of the terminal voltage between accelerator electrodes and to the

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tank wall still provide a means of breakdown product formation.

In addition to chemical breakdown products, voltage breakdown in the SF_6 space results in deposits on exposed metal surfaces and the dispersion of particulate material throughout the 14UD, which affect the insulating capabilities of the gas.

The SF_6 gas in the 14UD pressure vessel is recirculated continuously through alumina, with purity, SO_2 and moisture content analysed regularly using a Cambridge Sensotec Rapidox 3100CA system. Since 2014, the SF_6 purity, SO_2 content and H_2O content has averaged 96.6%, 2.2 ppmV and 5.7 ppmV respectively (at around 20°C and 14.3 psi). This moisture level is similar to that measured in gaseous nitrogen sourced from liquid nitrogen boil off.

Even with a low moisture content, significant breakdown products are still produced inside the 14UD, evidenced by the particulates discussed earlier, a pungent smell and the mild acidification of water used to clean the accelerator during periodic maintenance.

Vacuum Arcs

Even at 10^{-8}–10^{-7} Torr, electrical breakdown can occur between the electrodes in the accelerator vacuum space. Breakdown is triggered via the release of electrons through [5, 10]:

- field emission from protrusions on electrode surfaces, either from manufacture or previous discharges;
- field emission from foreign particulates embedded in the electrode surfaces;
- ionisation of gas or other low vapour pressure contaminants desorbed from electrode surfaces that then cause micro-discharges;
- hot field emission from impacting particles that deform the electrodes and;
- ionisation of metal vapour from high-energy particle impact.

Clean electrode surfaces and good vacuum are therefore necessary prerequisites for good high voltage performance, both of which become more challenging as the 14UD ages even further.

Electrode surface conditioning is necessary to achieve good insulating strength. This is a process of safely removing protrusions and moving contaminants through the repeated use of electrical discharges. In operating accelerators, this is most often done in-situ, with or without sections of the accelerator column shorted. The terminal voltage is increased just until qualitative evidence of conditioning, such as x-ray emission from high-energy electron impact [10] and small vacuum excursions, is observed. This evidence will disappear as conditioning progresses. Overzealous attempts at conditioning can lead to dissipation of the majority or all of the stored energy in the accelerator and cause violent breakdown across other gaps in the accelerator column, distributing particles and damaging electrodes leading to deconditioning. Deconditioning can also occur through loss of vacuum from outgassing after discharges or from even the smallest vacuum leak.

Although conditioning can also be performed ex-situ via fast spark, glow or arc discharges or active gas preparation [5], removal of tubes from the accelerator inherently requires venting the acceleration tube and compromising the vacuum.

14UD DEGRADATION FACTORS

The factors of an aging accelerator that affect ultimate performance of the ANU 14UD are many and varied. For example, erosion of spark-gap electrodes in the SF_6 space due to breakdown products increases surface roughness and the likelihood of electron field emission precipitating breakdown. Issues such as this are relatively simple to diagnose, track and rectify during routine maintenance. However, others issues such as small vacuum leaks, the condition of ceramic insulators and the condition of internal acceleration tube electrodes, are not.

SF_6 Injection Into Vacuum Space

The Australian Nuclear Science and Technology Organisation (ANSTO) reported on the injection of SF_6 gas into the vacuum space for use as a gas stripper medium [11]. While there was no report of an effect on accelerator performance, installation of a similar system into the ANU 14UD resulted in a decrease in the ultimate achievable terminal voltage and in rapid deconditioning. The conditioning limit could not be sustained even after short operation at lower terminal voltages.

More recently, the 14UD has suffered from pressure-sensitive vacuum leaks from the SF_6 space into the acceleration tube vacuum. Residual Gas Analysis (RGA) of the vacuum at the both ends of the acceleration tube showed a sudden appearance and sometimes subsequent disappearance of SF_6 without relation to any external event. This occurred on multiple occasions accompanied by up to a one and a half order of magnitude degradation in vacuum. Even with a recovery in vacuum, and no significant levels of SF_6 detected with RGA scans, accelerator performance degraded in a similar manner to that observed when using SF_6 as a stripper gas. This perhaps suggests adsorption of the gas onto the electrode surfaces that is then involved in discharge events.

Ceramic Insulators

The ceramic insulators in the acceleration tubes and column support posts are of course a critical component in the operation of an electrostatic accelerator. The majority of these in the 14UD are original to the machine and some are showing their age. Over time, high-voltage insulators can degrade due to operation in polluted environments [12] and suffer x-ray radiation-induced degradation in resistivity in the presence of an electric field [13].

As with an earlier investigation into ageing ceramics in the 14UD [2], current leakage across insulation gaps in the supporting columns posts is still observed. It is often accompanied by feint cracks in the surface of the ceramic or metallic track marks that bridge the gap. Leakage currents up to tens of microamperes at five kilovolts are measured

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and are sometimes extremely dependent on the surrounding humidity.

Physical damage of the ceramic is also observed without current leakage, with significant amount of debris visible in the surrounding area.

As the posts form a structural component of the accelerator, damaged posts are replaced with urgency and exclusively with new posts. A previous programme to refurbish posts through sandblasting and high pressure rinsing seems to have had no benefit on extending the lifetime of older, used, post assemblies.

It is unclear if the mechanical failure of ceramics is due to electrical stresses from voltage breakdowns or from mechanical stresses from pressure cycling, movement of the terminal spinning during maintenance or from the release of energy during large terminal voltage sparks.

Current leakage is also measured across some ceramic gaps in the acceleration tubes. These are more problematic as the tubes cannot be removed as readily as posts and the only viable solution in the short-term is to electrically short those gaps. There are currently 21 shorted gaps of a total of 980 gaps in the 14UD. While in theory this is only a 2.1% reduction in the achievable terminal voltage, the shorted gaps are mostly grouped in particular regions and have a detrimental effect on the homogeneity of the voltage gradient throughout the acceleration column.

Unlike ceramic gaps on posts, there is sometimes no obvious external physical evidence accompanying a leaky ceramic gap in the acceleration tube, suggesting an issue in the vacuum space.

Electrode Surface Condition

The surface condition of electrodes has a great influence on the voltage at which conditioning will first appear in the vacuum space and therefore the achievable operational voltage. Unfortunately, the condition of electrodes in the vacuum space cannot be monitored in-situ and controlled testing at full operational voltages during maintenance periods is not possible, since the high breakdown voltage SF_6 insulating gas is replaced with atmospheric air to allow entry into the pressure vessel.

ELECTRODE SURFACE CONDITION

Five acceleration tubes were replaced with viable used spares during a maintenance period in 2016. The replacements were to troubleshoot and repair issues created by a poor high-energy stripper design that allows burst carbon foils to escape the region and electron emission to be unsuppressed (which is another limiting factor on voltage performance). Three eleven-gap tubes above the high-energy stripper were removed from unit 19 and two eleven-gap tubes were removed from below the stripper in unit 20. The titanium insert electrodes within the tubes were removed and examined using SEM EDX analysis.

Figure 1 and Fig. 2 show the top of the first titanium electrode in unit 19, acceleration tube 2 (exposed to the ion beam) and the bottom of the last electrode in the same tube (chiefly exposed to electron flux). SEM images of the same electrodes are shown in Fig. 3 through Fig 6. These images show varied but common features observed, such as porosity, metal vaporisation and material build up. Titanium sheet samples were also analysed as a comparison (Fig. 7) and the surface was shown to be relatively pure, with sparse embedded particles with a high oxygen and carbon content.

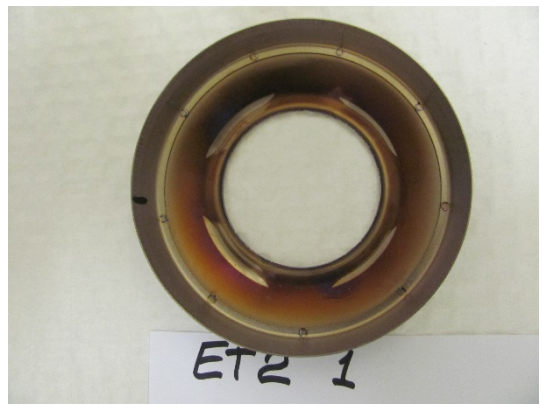

Figure 1: Top face of electrode 1 in unit 19, acceleration tube 2 after over 40 years of operation.

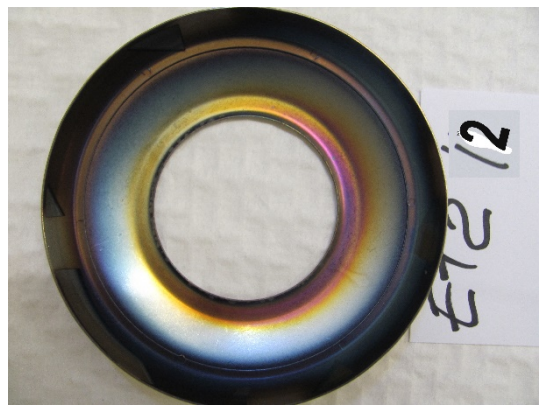

Figure 2: Bottom face of electrode 12 (last) in unit 19, acceleration tube 2 after over 40 years of operation.

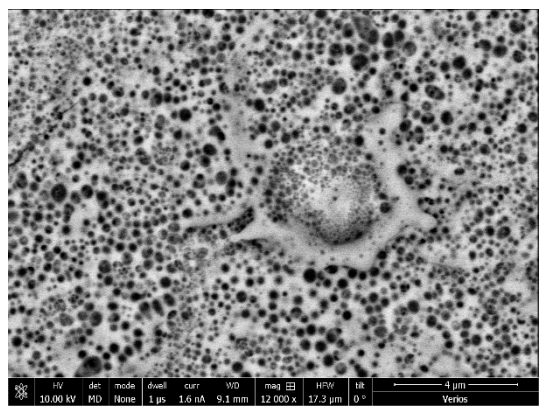

Figure 3: SEM image of the bottom of electrode 1 in unit 19, acceleration tube 2, showing metal vaporisation and surface porosity.

EDX analysis measured the composition at up to twenty radial points on the top and bottom of seven of the fifty-five electrodes removed. Three of these electrodes were the

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first electrode in a tube, three the last and one from the middle of a tube. Within measurement taken from a single side of an electrode, the composition was relatively homogeneous, but small differences were observed between the top and bottom sides of the electrodes.

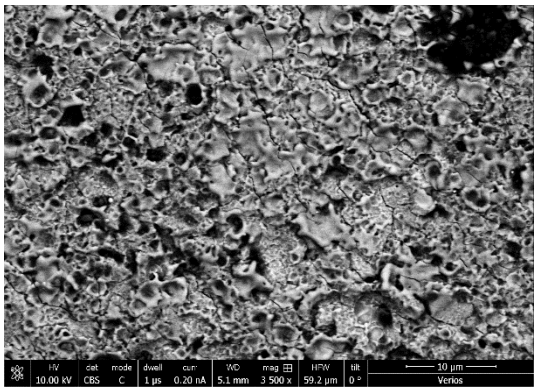

Figure 4: SEM image of the top of electrode 12 (last) in unit 19, acceleration tube 2 showing re-crystallisation of the surface layer of the Ti electrode.

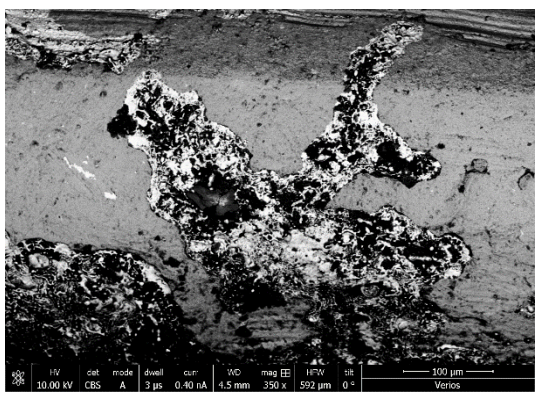

Figure 5: SEM image of the bottom of electrode 12 (last) in unit 19, acceleration tube 2, showing metal vaporisation.

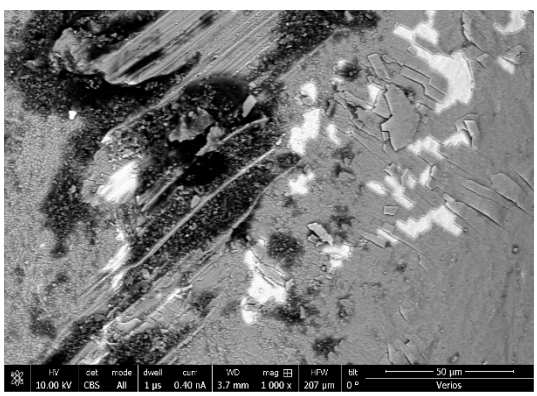

Figure 6: SEM image at the inner edge (toward the beam axis) of the bottom side of an acceleration tube electrode showing severe layer build up and subsequent possible flaking. The black regions are carbon rich layers and the white areas are pure Ti. The grey areas are loose films contaminated with O, F, S.

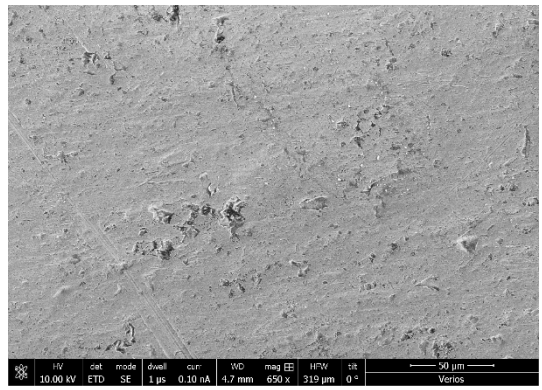

Figure 7: SEM image of titanium sheet as a comparison to used titanium electrodes in acceleration tubes.

Aside from titanium, the results showed the primary composition to be of oxygen, carbon, fluorine and aluminium, with trace amounts of silicon, argon, magnesium, sulphur, sodium calcium and iron. Across the seven electrodes analysed, oxygen percentages were 20–40% and generally higher on the underside of the last electrode in a tube. Carbon percentages were 10–20%. These are both more than double the amounts found on unused titanium. Fluorine, which is not seen on measurements of unused titanium, was found at percentages of 3–7%. Furthermore, very slightly higher levels of fluorine were found toward the axis of the acceleration tubes. Small amounts of aluminium were also detected, at 1–3%.

In combination with the SEM images, the oxygen and carbon content suggests a build-up of oxide and carbon layers, which aside from impacting the voltage gradient, would provide particulate material for increased initiation of vacuum breakdown. The supply of oxygen comes from residual gas in the acceleration tubes and oxygen gas that was historically used as a gas stripper medium. Most of the carbon contamination is likely to be from broken carbon stripper foils.

The presence of fluorine is further evidence of SF_6 leakage into the vacuum space.

Other features were also observed upon disassembly of the acceleration tubes. Pure physical damage to electrodes was observed, with craters in electrode surfaces and sputtering of titanium. This then creates protrusions that then act as a trigger for the release of electrons. An example of cratering is shown in Fig. 8.

REMEDIATION OPTIONS AND CONLCUSION

From the observed conditions of the accelerator tube insert electrodes, their remediation through cleaning or replacement should be a priority. However, even new electrodes may need processing. New tubes and electrodes installed during the compressed geometry upgrade of the 14UD were processed via magnetron sputtering using argon, followed by baking in a vacuum oven at 600 °C [3, 5].

NEC offer refurbishment of accelerator tubes via high pressure rinse of the tube and replacement or electro-polishing of the titanium insert electrodes. However, this does not remediate issues with possible deterioration of the bulk

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dielectric properties of the ceramic insulation, nor any damage to the aluminium bonding layer between the titanium and ceramic.

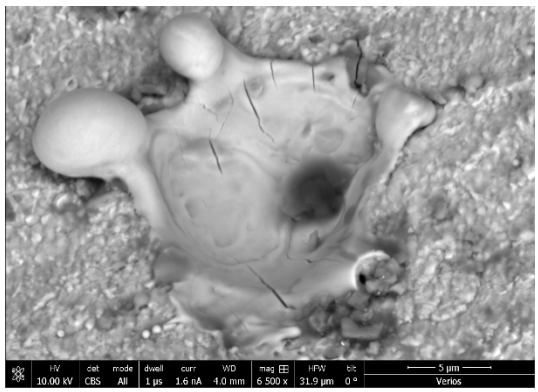

Figure 8: SEM image at the inner edge (toward the beam axis) of an acceleration tube electrode showing severe metal vaporisation.

The most obvious and efficient remediation option is the installation of new acceleration tubes. However, consideration of the large capital outlay required, facility downtime and the strategic future of the facility is necessary. Incremental replacement over time to reduce downtime requires regular venting of the vacuum space, compromising the good, low vacuum required to minimise vacuum breakdown.

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