INERT ANNEALING OF IRRADIATED GRAPHITE BY INDUCTIVE HEATING

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Abstract. Fission neutrons change physical properties of graphite being used in nuclear reactors as moderator and as structural material. The understanding of these effects on an atomic model is expressed by dislocations of carbon atoms within the graphite and the thereby stored energy is known as Wigner Energy.

The dismantling of the Pile 1 core may necessitate the thermal treatment of the irradiated but otherwise undamaged graphite. This heat treatment – usually called annealing – initiates the release of stored Wigner Energy in a controlled manner. This energy could otherwise give rise to an increase in temperature under certain conditions during transport or preparation for final storage.

In order to prevent such an effect it is intended to anneal the major part of Pile 1 graphite before it is packed into boxes suitable for final disposal.

Different heating techniques have been assessed. Inductive heating in an inert atmosphere was selected for installation in the Pile 1 Waste Processing Facility built for the treatment and packaging of the dismantled Pile 1 waste.. The graphite blocks will be heated up to 250 °C in the annealing ovens, which results in the release of significant amount of the stored energy. External heat sources in a final repository will never heat up the storage boxes to such a temperature.

1. BACKGROUND

The two Windscale Pile Reactors were constructed between 1946 and 1950 and operated until 1957 when Pile 1 was shut down following a core fire. This reactor is now being managed by UKAEA – The United Kingdom Atomic Energy Authority. A contract to undertake the decommissioning of this reactor was placed in August 1997 to the consortium BNFL plc, NUKEM Nuklear GmbH and Rolls-Royce Nuclear Engineering Services.

Windscale Pile 1 is a graphite moderated, air-cooled reactor which was fuelled with uranium metal rods clad in aluminium with longitudinal fins to improve heat transfer. The moderator is essentially a cylinder of diameter 15 m and length 7,5 m, laid on its horizontal axis. The core is surrounded by a 2,6 m thick biological shield constructed of reinforced concrete.

The goal of the contract is take Pile 1 from its current state to an end state in which the core has been fully dismantled and all fuel as well as graphite have been processed and packaged for medium term storage. The Pile 1 bio-shield will be sealed. A waste processing facility (WPF) will be constructed and operated during the realisation of the project. Intermediate radioactive waste will be processed to a form acceptable for both interim storage and final disposal through United Kingdom Nirex Limited (Nirex). Nirex is responsible for providing and managing facilities for the safe disposal of radioactive wastes. A new waste store suitable for at least 50 years will be erected. The project should be completed in 2008.

One of the major objectives during dismantling and waste treatment is the prevention of a fire. There exists a possibility that the core or remaining fuel (up to 17 Mg) could catch fire from a number of causes. The most significant hazards are the possible presence of pyrophoric uranium hydride associated with the fuel elements and the release of Wigner Energy from the graphite. The core will therefore be fully inerted with argon to maintain a very low oxygen concentration 1–2%.

As Nirex is not yet in a position to give definite advice on the need for annealing of the Wigner Energy, provision for inert annealing of the graphite has to be made.

2. IMPLICATIONS OF THE WIGNER ENERGY CONTENT AND THE REQUIREMENT FOR ANNEALING

Dislocations of carbon atoms within the graphite grains induced by neutron bombardment of the graphite initiated by the radiation of fuel elements are restored by the influence of temperature. During restoration the graphite material releases energy. This energy is called Wigner Energy and the release can start a self-sustaining reaction under certain conditions.

The restoration of the dislocations of carbon atoms can be observed over a wide temperature range from above room temperature up to 1500°C. The release of this energy can be triggered by any heat source such as encapsulation of the graphite by grout or backfill grouting procedures later in the final disposal.

For the purpose of planning and executing all stages of the graphite-disposal process, it is necessary to address the question of whether an inadvertent release of stored energy (Wigner Energy) can occur during handling and storage such that deliberate partial annealing of the material is required.

This issue concerning Wigner Energy is not limited to the disposal of Pile 1 undamaged graphite. Rather the issue has been subject to deliberations by an industry-wide working group – the Nirex "NCTM Wigner Energy Sub-group". The Consortium is represented in this group thus allowing a common approach to Wigner Energy.

Input to the deliberations of the NCTM Wigner Energy Sub-group has culminated in the completion of two work packages:

Work Package 1: Derivation of equations for best-estimate and bounding isothermal releases of stored energy over the temperature range of concern (25–175ºC) including a fundamental review of international literature in this regard

Work Package 2: Use of the equations derived in Work Package 1 in scoping calculations to predict what temperatures may be expected in boxes of grouted graphite.

The results of these two work packages have been presented to Nirex for incorporation into a Nirex repository model to provide a realistic assessment of the affect of the Wigner Energy release during the backfill process. First results show, that the behaviour in the curing grout is very sensitive to the assumptions made in the calculations. It is very difficult to construct a bounding argument how high temperature excursions cannot occur. Nirex was of the view that even a substantial programme of further work may be unable to provide the necessary assurances. Further it is unclear whether the graphite containing Wigner Energy is consistent with the regulatory views on ′passive safety′.

NUKEM Nuklear GmbH started a programme to develop an annealing plant inside the WPF for the dismantled Pile 1 graphite.

The current Waste Processing Facility (WPF) design incorporates equipment to anneal the graphite blocks classified as undamaged graphite, with this provision subject to the conclusions drawn by the Nirex NCTM Sub-Group.

3. QUANTITY AND NATURE OF THE GRAPHITE TO BE ANNEALED

3.1. Waste type and physical description

The graphite is Intermediate Level Waste in the form of graphite blocks, slats and tiles from outside the fire affected zone (FAZ) of the Pile 1 core (accounting for approximately 80% of the core bulk graphite).

The Pile core is constructed from graphite blocks stacked in such a way as to make a horizontal cylinder 15.32 m diameter and 7.43 m deep. The blocks are stacked vertically and located to the blocks above and below by graphite slats and tiles. The lattice of slats and tiles are all assembled to the same pitch, this positions the centre of the graphite blocks which rest on the cruciform. Three grades (purity) were used in construction, as defined in terms of their nuclear cross-section. Due to the different types of graphite and the many holes and channels to accommodate the shut off rods, control rods, fuel and isotopes there were literally hundreds of brick designs. For the purposes of annealing, however, it is adequate to consider the blocks, slats and tiles as graphite pieces each with the following overall dimensions:

- full height blocks – 210 · 210 · 790 mm
- half height blocks – 210 · 210 · 370 mm
- slats – 400 · 26 · 90 mm
- tiles – 180 · 180 · 52 mm

3.2. Waste quantity

The core consists of approximately 27,440 full-height blocks, 10,960 half-height blocks, 37.600 slats and 37.600 tiles. This is equivalent to approximately 1.760 Mg of graphite occupying a raw waste volume of approximately 1.100 m^3

Only undamaged graphite will be annealed. The quantity of undamaged graphite is largely determined by the definition of the FAZ of the Pile 1 core. The above inventory represents approximately 80% of the total core graphite.

3.3. Definition of undamaged graphite

The undamaged graphite waste stream may be defined as the graphite from that part of the Pile 1 core that is outside the fire affected zone. In addition, assay and monitoring will determine that undamaged graphite will meet the criteria from the IAEA Transport Regulations for non-fissile, LSA-II material prior to box loading. Undamaged should not be interpreted as a description of the physical appearance of the graphite pieces. For example, it is perfectly feasible that undamaged graphite blocks may be broken during core dismantling.

3.4. Pile 1 graphite chemistry

The waste consists entirely of graphite removed from the Pile 1 core plus any associated contamination.

3.4.1. Chemical analysis of swab samples

An indication of the chemical contaminants present on the surface of Pile 1 undamaged graphite is given by the results of chemical analysis of swab samples taken from four channels located above the FAZ. It is recommended [1] that large uncertainties should be applied to the analytical data, especially on account of the small sample size. The bulk graphite is essentially pure carbon with other elements as impurities being present at the ppm level only. From a physical/chemical point of view the undamaged graphite waste will behave as pure carbon.

Carbon, as graphite, is a chemically low reactive solid at normal temperatures and pressures. Carbon is very slowly oxidised to carbon-dioxide in the presence of oxygen.

3.4.2. Thermal oxidation

Chemical analysis of swab samples taken from Pile 1 and 2 fuel channels have demonstrated the presence of several chemical elements which are potentially capable of enhancing the rate of graphite oxidation. As the swab samples represent only channel-wall surfaces, where certain potential oxidation catalysts derived from failed cartridges etc. are likely to be located, the data may be expected to give a pessimistic impression of the reactivity of the underlying bulk graphite. More meaningful information on thermal oxidation rates relevant for the bulk graphite is determined from the trepanned samples.

Measurement of the oxidation rates for the Pile 1 trepanned samples were very variable [2]. Ref. [1] reports that this is apparently an intrinsic property of the graphite used, quite apart from any subsequent catalytic contamination. A few samples showed unusually high results; the highest measured oxidation rate was 7405 $\mu g\ g^{-1}\ h^{-1}$. These high results are likely to be the result of contamination by the chemical contaminants mentioned above e.g. lead is known to act as a catalyst for graphite oxidation.

The majority of measured thermal-oxidation rates were made at or very close to 400°C. Utilisation of the measurement data, together with typical activation energies determined from Pile 2 samples, enables oxidation rates to be estimated at other temperatures. Thus, using the activation energy data, the most chemically active sample at 400°C, having an oxidation rate of 7405 $\mu g.g^{-1}.h^{-1}$, would have an oxidation rate of only around 0.35 $\mu g \cdot g^{-1} \cdot h^{-1}$ at 200°C [2]. At such a low oxidation rate, the exothermic heat generation from the reaction with air is small.

In order to minimise oxidation of graphite the annealing process is designed to in an inert argon atmosphere.

3.4.3. Wigner energy

During operation, Pile 1 was annealed several times to remove the low temperature component of stored energy (Wigner Energy) and it was during such an annealing in October

1957 that Pile 1 caught fire. Pile 1 had accumulated a dose of 41488 MW d prior to the final annealing. Prior to the analysis of the trepanned graphite samples the opinion was that the fire would have released much of the stored energy from the Pile, but the results of the tests show that any heating effect was limited to immediately above the FAZ or towards the discharge face, where the stored energy would naturally have been less because of the higher irradiation temperature.

The results of the Wigner Energy related measurements on the Windscale Pile 1 trepanned graphite samples are reported in [3].Samples from 11 fuel channels were thoroughly investigated in a survey recently in 1997/1998. The essential results and the consequences for an annealing process are given in [4].

3.5. Physical/mechanical properties

3.5.1. Density

The densities of the nominally 7 mm by 2 mm thick specimens of undamaged graphite trepanned from the core ranged from 1,48 to 1,70 $g.cm^{-3}$. The mean density was 1,59 $g.cm^{-3}$ [3].

3.5.2. Strength

The compressive strength measured on the trepanned cores from Pile 1 [3] in the range of from 39,6 to 67,2 MPa.

3.5.3. Thermal conductivity

The thermal conductivity of irradiated graphite reduces rapidly with irradiation. The lowest value measured for Pile 1 was 2.1 $W\ m^{-1}\ K^{-1}$ compared with not irradiated graphite which has thermal conductivity of 238 $W\ m^{-1}\ K^{-1}$ parallel to the extrusion direction and 135 $W\ m^{-1}\ K^{-1}$ perpendicular to the extrusion direction [2]. Reference [2] reports that this low value will be typical of most of the graphite in Pile 1, with the exception of blocks in the reflector and at the front and back of the Pile which may have experienced a low irradiation dose. The highest value measured on Pile 1 samples was 10,2 $W\ m^{-1}\ K^{-1}$.

3.5.4. Dust

The Pile graphite is resistant to abrasion, as demonstrated for example, by the requirement to slide graphite boats and fuel cartridges along the fuel channels during loading/unloading of Pile 1. Irradiation has also resulted in hardening of the graphite. Processing the graphite through the WPF and the annealing ovens includes several of handling sequences of individual graphite blocks. Minimal dust may be expected to be associated with the undamaged graphite waste.

4. DEVELOPMENT OF THE ANNEALING PROCESS

The decommissioning of the Windscale Pile 1 is conveniently broken down into two areas; the dismantling of Pile 1 and the processing/packaging of the retrieved waste. This description concentrates on the processing of the graphite. The annealing process is an integrated part of the WPF and must therefor be suitable for.

4.1. Waste processing facility

The dismantling of Pile 1 will be performed remotely. Four manipulators will be used from above each accessing one quarter i.e. front right, front left, back right and back left when viewed from the top. The manipulators will dismantle Pile 1 in a regular pattern starting at the corner furthest from the centre and working towards the centre in a layer by layer manner. The retrieved graphite will be placed in skips (approximately 1m^3 capacity) held within Pile 1. Once a skip is full, it is transferred to the Buffer Store in the Waste Processing Facility via the modified Pile 1 air duct. The dismantling operations will take place in an argon environment. The ducts between Pile 1 and the Waste Processing Facility will also be inerted.

4.1.1. Waste receipt, storage and identification

The waste will be received into the Buffer Store part of the Waste Processing Facility. The Buffer Store will provide a hold-up of skips to compensate for peak arisings from the dismantling operations. The Buffer Store is inerted by argon. The skip bar code is read on entry to the Buffer Store and the skip contents are correlated from the numeric skip number input during retrieval.

4.1.2. Processing of graphite

The operator will request a skip to be transferred into the Sort/Test Area based on the skip identifying number and the waste contents. Skips of waste are transferred into the Sort/Test Area through an argon door using cranes and a conveyor. Large items of waste (i.e. graphite blocks, fuel cartridges, isotope cartridges and boron rods) will be removed from the skip using manipulators.

The receipt, sorting and identification operations will be undertaken in an inert argon atmosphere in order to avoid any potential problems with any pyrophoric material present e.g. uranium hydride.

When undamaged graphite, i.e. from outside of the FAZ, is removed from the skip it will be visually examined to detect the presence of any part of a fuel or isotope cartridge associated with graphite. A gamma camera is also used to confirm the visual examination. If the graphite passes the visual examination, it is placed on a tray and transferred to the annealing ovens where the heat treatment takes place. To meet the throughput requirement the annealing process is designed to anneal a full size block or two half size blocks every ten minutes. After the heating procedure the graphite will pass a cooling station. to be cooled down to less than 50°C.

4.1.3. Waste packaging

After cooling and radiological measurements the undamaged graphite will be loaded into the Nirex 12 m^3 standard box for intermediate level waste (4m ILW) at the box loading station in the Waste Processing Cell. The anticipated loading is ca. 8,3 to 8,7 m^3. Weighing ensures that the 55 Mg limit of the 4m ILW box is not exceeded. After closing and monitoring the box is transferred to the ILW store.

4.1.4. Selection of annealing temperature

Measurement of the release of Wigner Energy over increasing temperatures show many peaks. A typical release peak occurs at about 200°C especially for the samples from relatively cold irradiation temperatures [4]. The irradiation temperature was low in front of the core as well as in lower and corner regions. The release of energy at this peak is sometimes greater than the specific heat of the graphite and therefore has the potential to initiate a self sustaining release.

The next peaks above specific heat were found at 1200°C to 1600°C. It is nearly impossible to remove all stored Wigner Energy as temperatures of about 2000 °C would be required. It is not possible however to reach such temperatures in a disposal repository.

Specialists generally agree that an annealing procedure should concentrate on the low temperature peak at 200°C.

A selection of annealing temperature is discussed in detail in [4].

4.2. Selection of Heating Process

Different heating techniques have been assessed as alternatives for the proposed annealing process. Convection heating is an indirect heating method and was mainly dismissed as being too slow. Radiation heating represents a fast, direct heating method for the "black body" graphite material but was dismissed as the requirement to replace the radiation heaters several times during the proposed plant life-time was regarded as being too difficult and time consuming. In addition, the extreme reduction of the thermal conductivity of the irradiated graphite in comparison to the virgin material by a factor of 50 to 100, renders a process which relies on the transport of heat from the surface through to the bulk material as unsuitable.

In contrast, inductive heating is a heating method for any electrical conductor and offers the advantage that the electromagnetic energy penetrates directly into the bulk material resulting in complete heating. Physical laws, which describe the penetration of the electrical current show that this effect is depending on the electrical resistance of the material to be heated and the frequency used. As graphite is ideally suitable for an induction process as a result of its comparatively high electrical resistance, process optimisation is essentially an assessment of the frequency. A calculated penetration depth of approximately 50 mm which may be achieved at a frequency of a few thousand Hertz seems ideal for the dimensions of the Pile 1 graphite blocks.

Another advantage of induction heating compared to any other alternative is that the proposed equipment does not get very hot during the heating procedure. This results from the fact that the copper coil installed to induce the electrical current in the graphite is cooled by a flow of water through its inner volume. Furthermore, the use of other materials suitable to couple electrically to the energy produced by the copper coil is not allowed within its electromagnetic fields. Thus the energy generated is almost totally concentrated on the material to be heated. This advantage supports meeting the expected throughput with a comparatively small number of annealing ovens.

4.3. Tests with inductive heating

Orientating tests have been performed in order to prove the inductive heating method and to obtain better physical parameters for the equipment.

The test equipment, shown in Fig.1, consists of:

- induction coil
- control cabinet with frequency converter
- cooling water supply

The induction coil was constructed from copper tube and consisted of 17 coils in a cube shaped formation with the dimensions 900 × 320 × 320 mm. The inner volume of the coil was fitted with 10 mm thick isolation material.

Four heating tests have been performed with unirradiated graphite type UC 312/496 with a density of 1.800 kg $\cdot$ m^{-3}. The tested graphite parts correspond to the dimensions of the slats (26 × 90 × 370 mm) and tiles (52 × 180 × 180 mm) from the Pile core. It was considered that the electrical coupling would be easier for graphite pieces with dimensions similar to full height blocks and half height blocks.

During each heating test the electrical data (power, voltage, frequency) were kept constant. The coil was water-cooled.

The inner temperature of the arrangement was measured by a thermocouple and the surface temperature was measured by an infrared pyrometer in order to obtain the heating curves of graphite test pieces. The temperatures were read every minute.

The following parameters were tested:

- arrangement of the graphite parts
- electrical power20 – 30 kW
- voltage 360-580 V
- frequency 3 – 3,2 kHz

The orientating tests gave the following results:

Inductive heating of graphite is practicable without any difficulty. Because of the relative good electrical coupling of the tiles in the induction coil, heating up by 350°C can be achieved over 15 minutes (slats 30 minutes) with a power input of 30 kW and a frequency of 3 kHz (Fig. 2.).

The heating curves show that the inner graphite temperature rises ahead of the surface temperature. The hottest temperatures are expected close to the surface as the current flux is the highest. The surface is, however, cooled by convection and radiation. This means, that during operation, it is sufficient to measure the surface temperature and one can be quite sure that the temperature inside the blocks will be even higher.

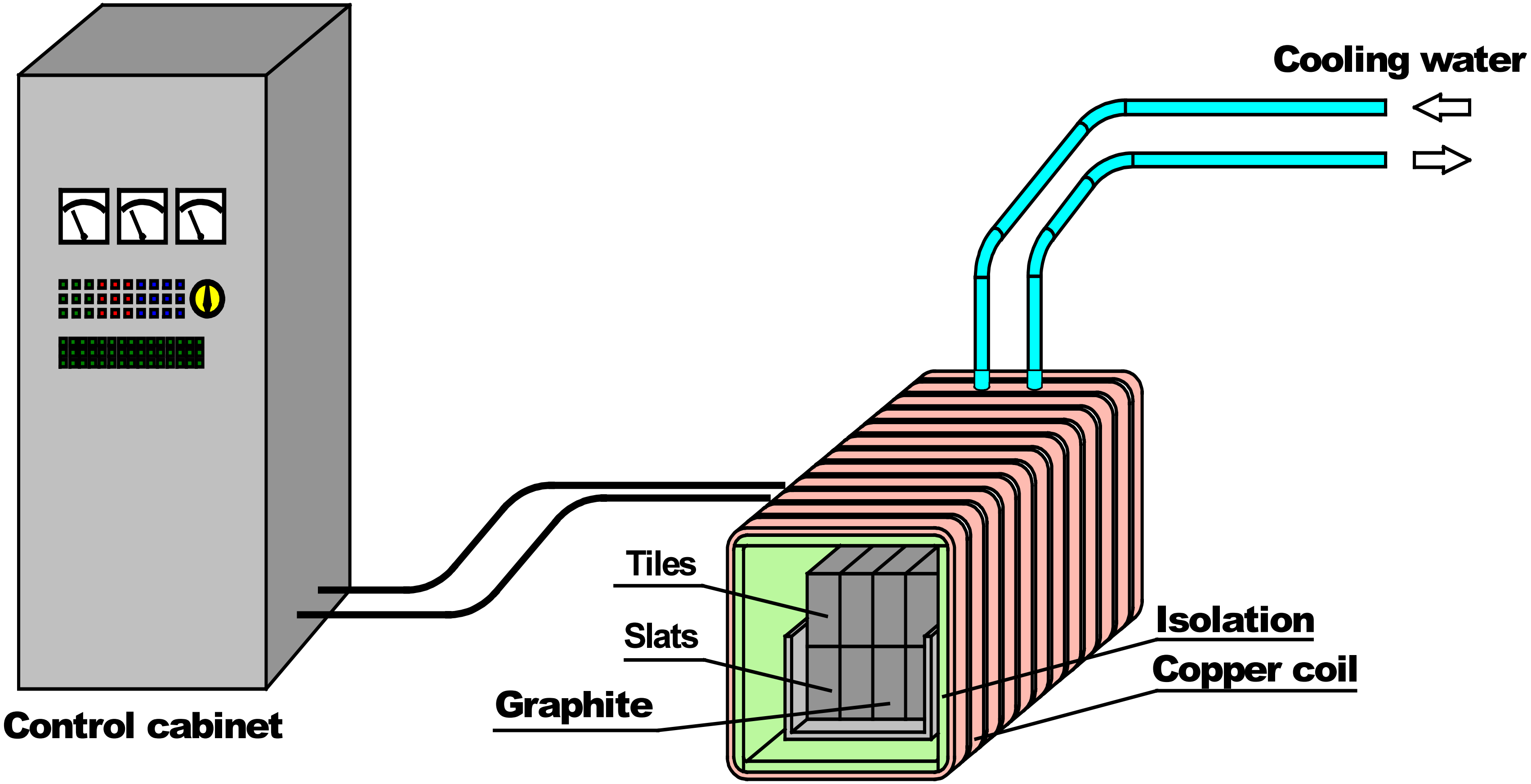

FIG. 1. *Test arrangement with inductive heating.*

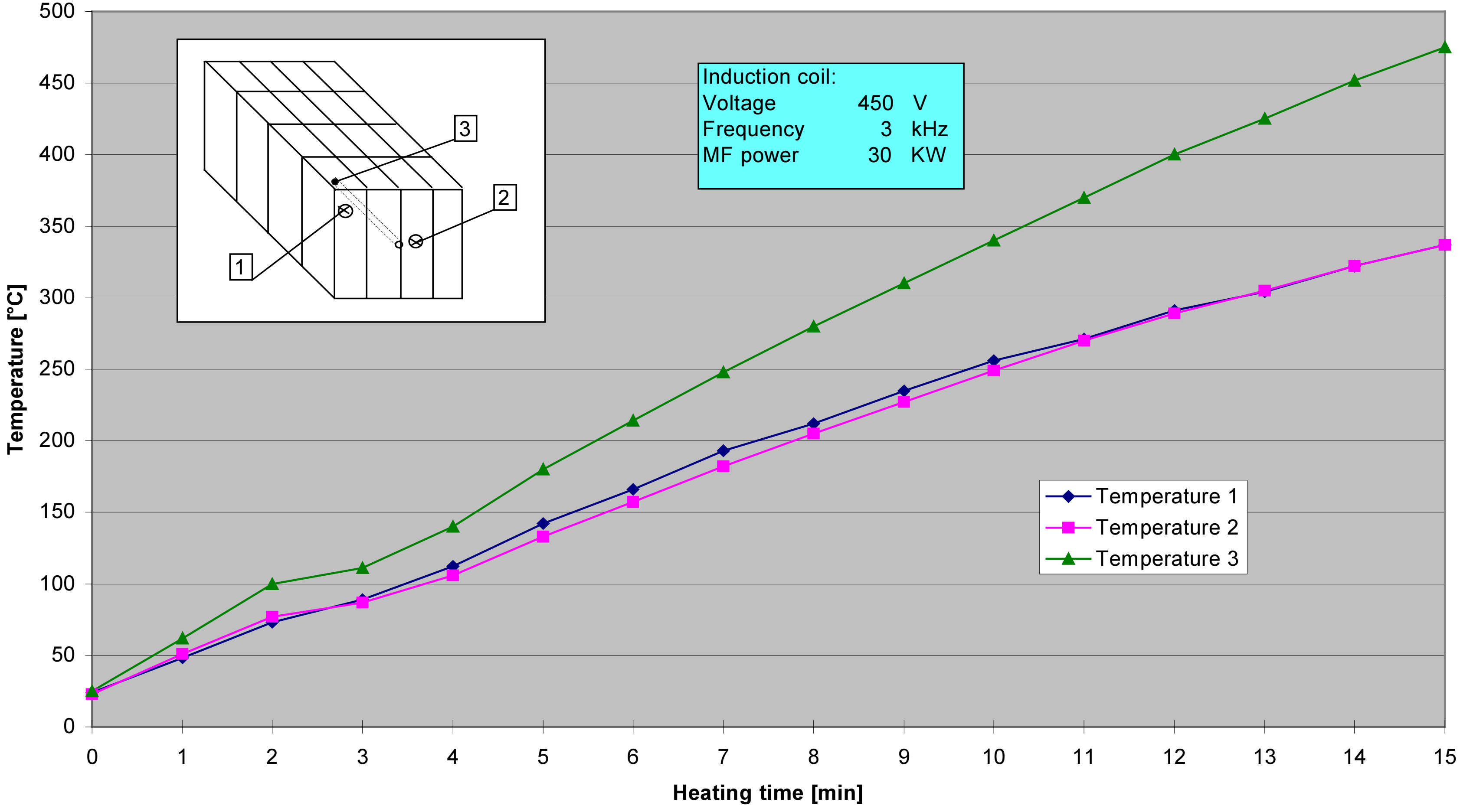

FIG. 2. Inductive heating of graphite-tiles heating curve.

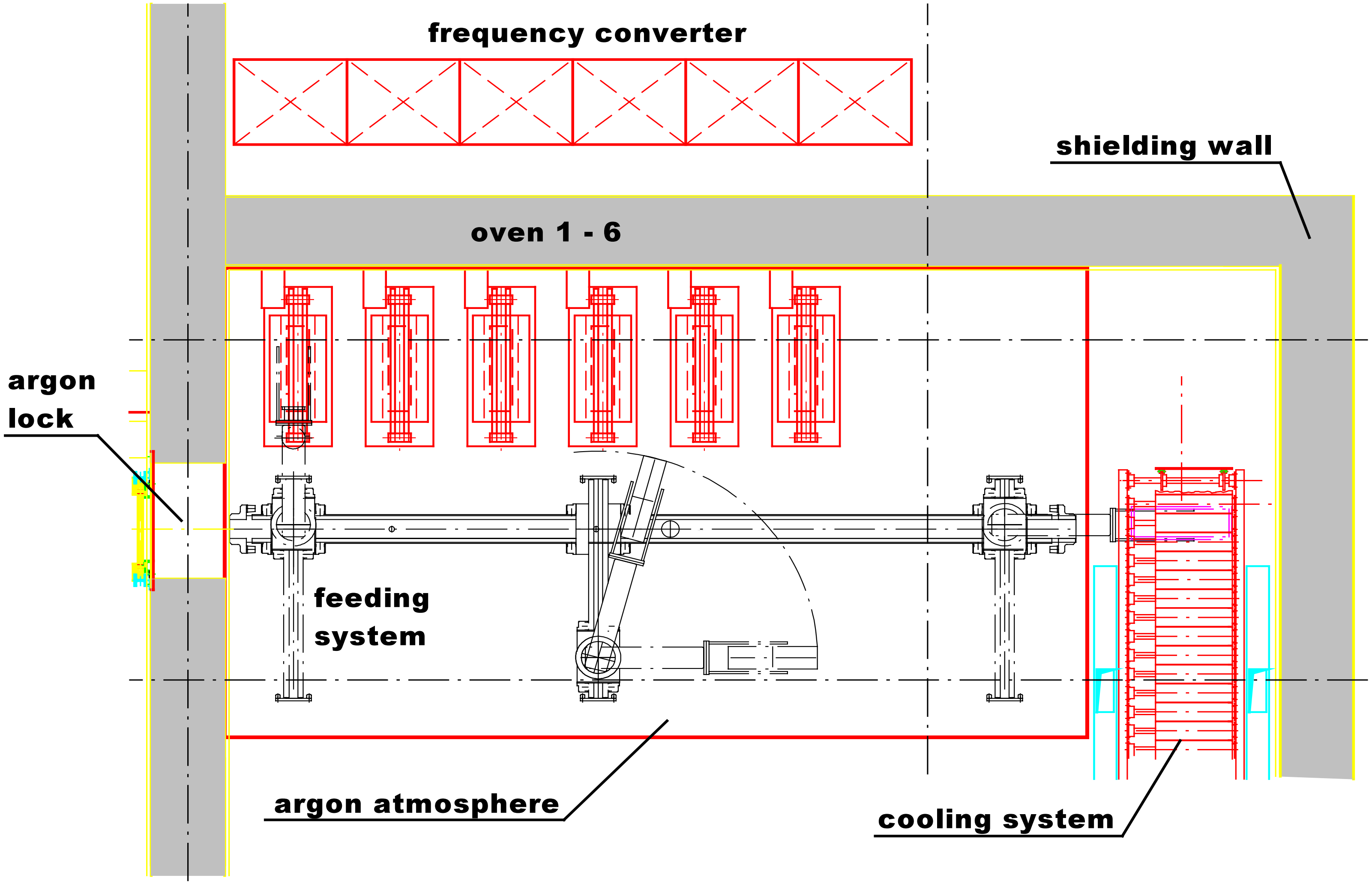

FIG. 3. Arrangement of annealing equipment in the WPT.

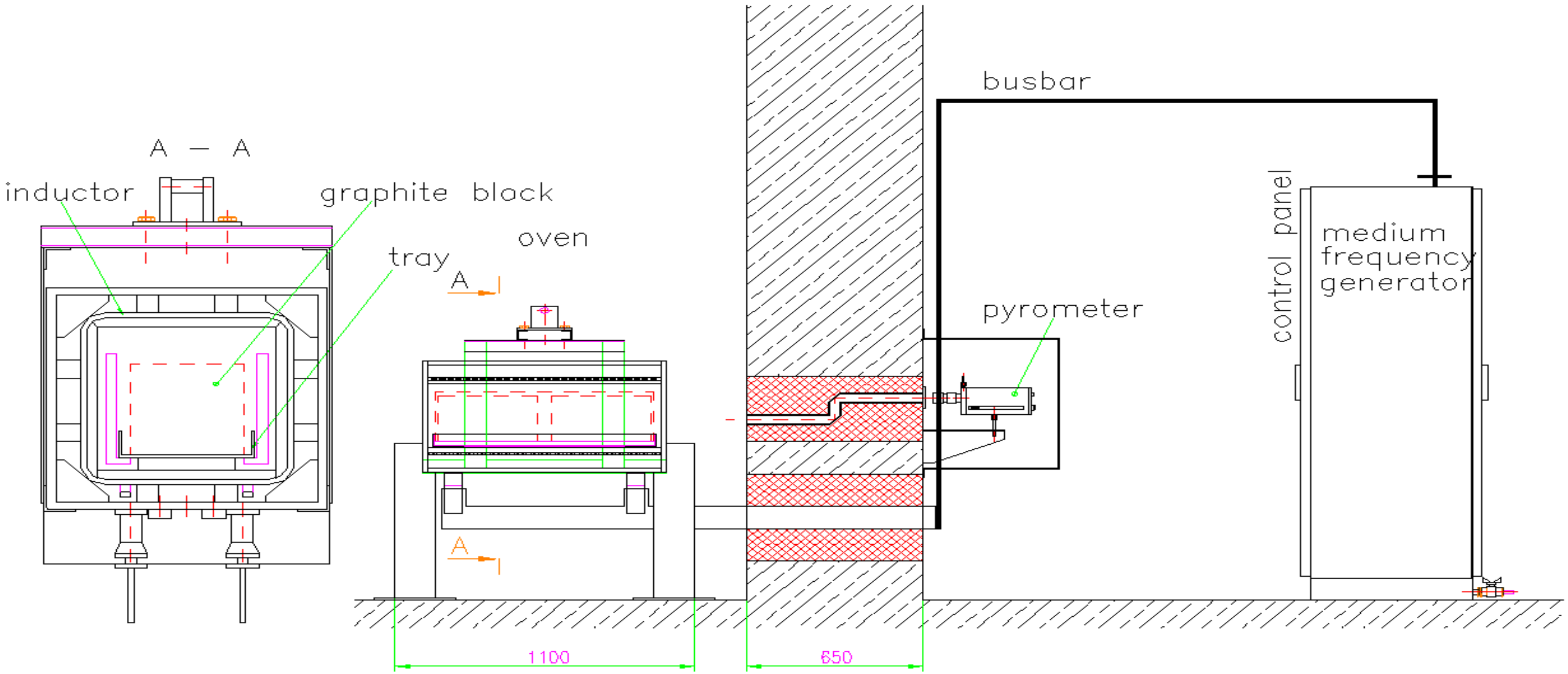

Fig. 4. Annealing oven section.

4.5.Annealing equipment in the Pile 1 waste processing facility

To guarantee the required throughput in the WPF, six annealing ovens working in parallel have been selected. Figure 3 shows the arrangement in the facility. The annealing equipment and associated graphite feeding system is located in an argon inert atmosphere inside a caisson. The feeding system takes a ceramic tray, on which one full height block or two half height blocks are positioned and pushes it into the annealing oven (Fig. 3, Fig. 4). Within about seven minutes the graphite will be heated up to about 200°C. After a pause of three minutes the heating process starts again to increase the temperature to 250 °C. The temperature will be measured on the surface of the graphite by pyrometers located outside the hot cell shielding wall. The heat radiation is transferred through the wall using a mirror system. The graphite will be held at 250°C for about twenty minutes to observe the behaviour. If the release of Wigner Energy results in higher temperatures the heating will be switched off. Following this heat treatment process, the tray with the graphite is transported by the feeding system through a double lock into a cooling station were cooling in air to less than 50 °C is performed.

Characteristics of each oven:

Maximum output	40 kW
Phased current	80 A
Frequency	3 kHz
Working frequency range	2,25-3,75 kHz

Frequency converter located outside the hot cell.
Inductor water cooled.

The first annealing oven unit is will be constructed within one year. It is planned to optimise the parameters in a new test sequence before the next five units will be produced.

5. CONCLUSION

Inductive heating of Pile 1 graphite blocks in an inert atmosphere to anneal the fist peak release of Wigner Energy promises to be successful and it will be implemented in the new Waste Process Facility.

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