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Second generation laser-heated microfurnace for the preparation of microgram-sized graphite samples

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ABSTRACT

We present construction details and test results for two second-generation laser-heated microfurnaces (LHF-II) used to prepare graphite samples for Accelerator Mass Spectrometry (AMS) at ANSTO. Based on systematic studies aimed at optimising the performance of our prototype laser-heated microfurnace (LHF-I) (Smith et al., 2007 [1]; Smith et al., 2010 [2,3]; Yang et al., 2014 [4]), we have designed the LHF-II to have the following features: (i) it has a small reactor volume of 0.25 mL allowing us to completely graphitise carbon dioxide samples containing as little as 2 μg of C, (ii) it can operate over a large pressure range (0–3 bar) and so has the capacity to graphitise CO_2 samples containing up to 100 μg of C; (iii) it is compact, with three valves integrated into the microfurnace body, (iv) it is compatible with our new miniaturised conventional graphitisation furnaces (MCF), also designed for small samples, and shares a common vacuum system. Early tests have shown that the extraneous carbon added during graphitisation in each LHF-II is of the order of 0.05 μg, assuming 100 pMC activity, similar to that of the prototype unit.

We use a 'budget' fibre packaged array for the diode laser with custom built focusing optics. The use of a new infrared (IR) thermometer with a short focal length has allowed us to decrease the height of the light-proof safety enclosure. These innovations have produced a cheaper and more compact device. As with the LHF-I, feedback control of the catalyst temperature and logging of the reaction parameters is managed by a LabVIEW interface.

1. Introduction

At ANSTO, we are equipped with two tandem accelerators: the 2 MV (million volt) STAR and the 10 MV ANTARES machines, both used for radiocarbon measurement. Two new radiocarbon-capable tandem accelerators, a 1 MV and a 6 MV machine, are currently being added to the Centre for Accelerator Science at ANSTO. With the increasing number of AMS machines, we need to increase our graphitisation capability for the preparation of radiocarbon samples.

Since establishment of radiocarbon AMS, many laboratories worldwide have worked to improve their capability for AMS of smaller and smaller carbon samples. Notable amongst the laboratories that prepare and measure 'micro' graphite samples are the KCCAMS Laboratory, with a 2 μg of C capability [5], and the VERA laboratory with a 1 μg of C capability [6]. Some laboratories have bypassed the graphitisation step completely and operate gas (CO_2) ion sources, for instance the ETH laboratory with a 1 μg of C capability [7]. Comparison of the performance of these and other laboratories, for the preparation and measurement of 'micro-carbon' samples, is beyond the scope of this paper.

At ANSTO we have chosen to graphitise our small samples for AMS. Development of the laser-heated microfurnaces LHF-II is one path to improving our throughput of ultra-small radiocarbon samples [1–3]. Based on our operational experience with the prototype laser-heated microfurnace (LHF-I), we designed and manufactured a second generation device, LHF-II, with the aim of producing ~5 μg carbon samples with high reliability and ease. One important application of the LHF-II is for radiocarbon measurement of carbonaceous gases and particles in ice sheets – the ability to reliably prepare and to analyse ultra-small samples is required in such investigations due to the very limited quantity of CO, CO_2 and CH_4 in ice core bubbles and of carbon particulate matter in the ice. Reliability is important for such projects as expedition costs are high. Moreover, there are many other applications at ANSTO that involve small samples for ^{14}C analysis, in fields such as chronology, archaeometry, palynomorphs (pollen, micro-foramifera, phytoliths), diatoms, organic fractions from archaeological material (e.g. mummies, cooking vessels, tool residues), biomedical, DNA (e.g. for brain/neuron cell age) and specific compounds, separated, for example, by preparative gas chromatography.

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2. Construction of LHF-II

2.1. Overall system

The LHF-II is designed to produce very small samples with low background and high reproducibility. It is compact, low cost and user friendly. Figs. 1a and 1b show the overall system: the control and display system, the graphitisation reactor, the vacuum system and the cold finger cooling system. The graphitisation reactor is comprised of four parts: (i) the core of the reactor, with three integrated valves for gas and vacuum control, (ii) the laser lens housing, used to focus the IR beam from an optical fibre to ~0.3 mm diameter on the catalyst, (iii) the fibre optic infrared thermometer lens probe, which views the heated target spot, and (iv) the light-proof safety enclosure, including a bottom plate, back plate and a rotatable top box. The IR laser diode module is installed on the back plate inside the enclosure and is connected to the lens housing by an optical fibre. Fig. 1c is an exploded view of the graphitisation reactor.

2.2. Reactor core

The core of LHF-II is machined from a solid block of stainless steel (64 × 64 × 26 mm) and internal surfaces are electro-polished. Viton o-rings are used to seal the valves, miniature pressure transducer and the 3 mm thick, 25 mm diameter quartz window, transparent to IR from the incident laser beam and from black body emission from the heated target. Fig. 2a shows a plan view of the cross section of the core, showing three valves and three ports. The three ports and the cold finger are laser welded to the block. One port is connected to the vacuum/H_2 manifold and another port is connected to the CO_2 sample transfer flask or breakseal cracker. The third port is spare for future development. The valves permit connection to vacuum/H_2 manifold and sample CO_2 and allow the reaction volume to be isolated. Fig. 2b is side view of the central cross section of the core, revealing the reaction volume and the positions of the stainless steel cold finger, crucible and quartz window. The volume of the isolated reactor is 0.25 mL, calculated by measuring the pressure of a known quantity of CO_2 gas in the reactor. The catalyst is placed in a gold-plated quartz crucible (4 mm high, 4 mm diameter, 2.0 mm radius hemispherical recess) directly under the quartz window.

2.3. Vacuum system

Fig. 3 is a photograph of the vacuum manifold assembled on the work bench, also partially shown in Fig. 1a. This manifold has six ports for connection to graphitisation modules. The main vacuum pipe is placed horizontally under the bench and is connected to a turbomolecular pumping station, a MKS Baratron gauge for measuring hydrogen pressure and for calibrating the miniature pressure sensors used in the microfurnaces, a cold cathode gauge

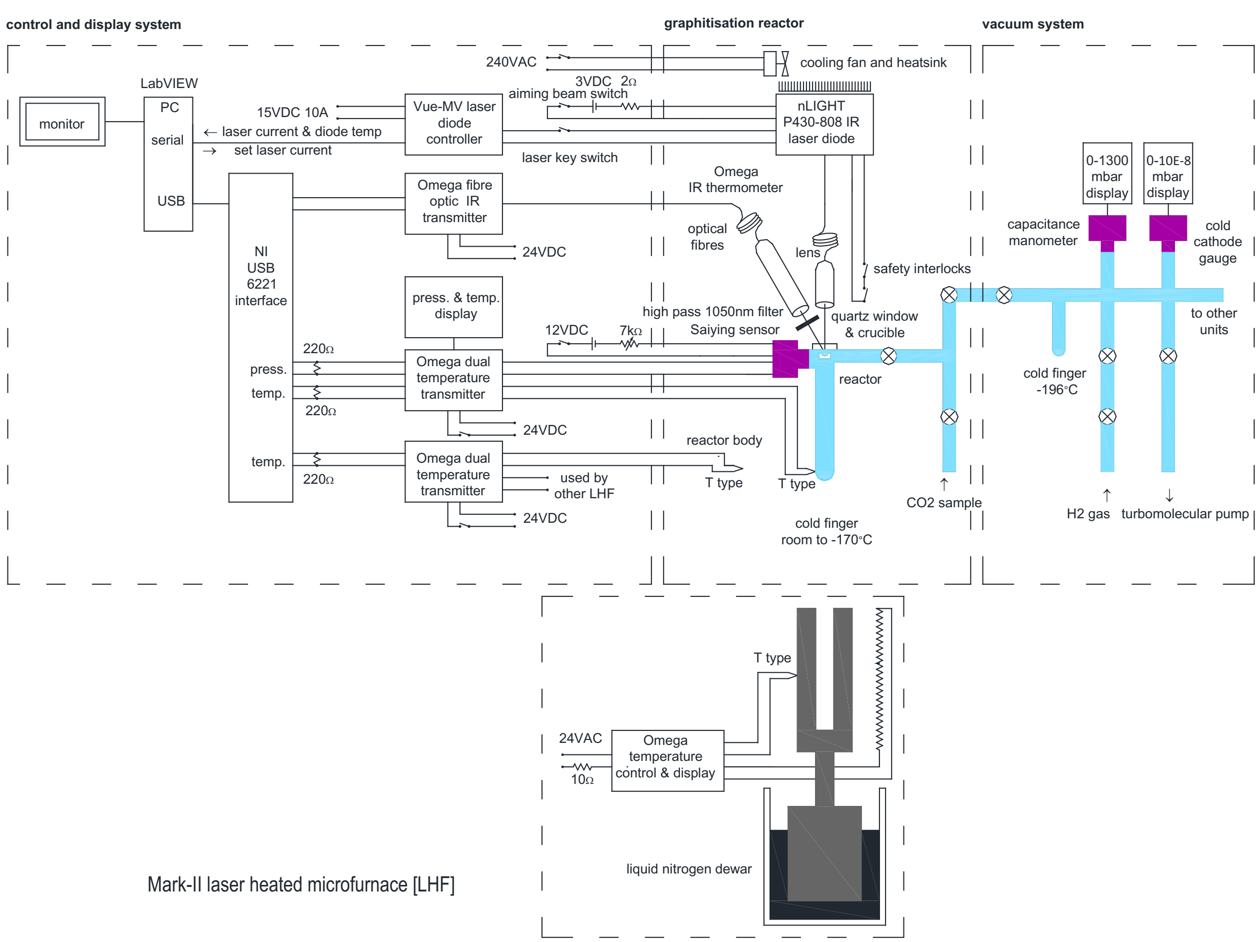

Fig. 1a. Block diagram of one LHF-II: units a and b share the same PC interface, LabVIEW control and vacuum/gas manifold. The National Instruments interface NI USB-6112 accepts 0–4.8 V DC signals, provided by 220 Ω resistors in the 4–22 mA current loop from the Omega temperature transmitters. The miniature Saiying pressure transducer is 0–3 bar.