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Developments in micro-sample ^{14}C AMS at the ANTARES AMS facility

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ABSTRACT

We continue development of micro-sample radiocarbon sample preparation and AMS measurement at the ANTARES AMS facility. We routinely prepare samples containing 10–200 μg of carbon using an iron catalyst with an excess of hydrogen in ~2.5 mL graphitisation reactors. These use a tube furnace to heat the catalyst to 600 °C and a Peltier-cooled water trap. Samples containing just a few micrograms of carbon can be prepared. We describe progress with a 0.5 mL laser-heated 'microfurnace' we are developing for the rapid and efficient graphitisation of ~5 μg samples. Following operating experience with a prototype unit, work has commenced on the development of a second-generation device with the goal of fully automated operation with minimal introduction of extraneous carbon.

Key to development of micro-sample ^{14}C AMS is the ability to reliably handle the graphite/iron sample and to mount it in the ion source target holder. We have developed a target holder that permits the sample to be loaded in a 1 mm diameter recess and rear pressed, ensuring a high quality surface finish, at a reproducible depth. Additionally we have developed a method for systematically aligning the sample stage with the cesium beam following ion source servicing.

1. Introduction

ANSTO uses two tandem accelerators for ^{14}C AMS: the 10 MV ANTARES (C^{4+} at 5.2 MV) and the 2 MV STAR (C^{3+} at 1.95 MV). Both machines have the same High Voltage Engineering (HVE) model 846B solid-sample, multi-target, high-intensity cesium sputter negative ion source. This enables efficient use of sample graphite by moving the target beneath the primary cesium sputtering beam. All ANTARES AMS uses fast isotope bouncing whereas STAR uses a recombinator system [1,2].

At ANSTO, sample CO_2 is reduced to graphite over Fe catalyst at 600 °C in an excess of H_2 in a 'graphitisation furnace' [3] by the Bosch reaction [4–7]. Although there are various intermediate reactions [8], the overall reaction is $CO_2 + 2H_2 \rightarrow C + 2H_2O$. The water vapour formed during the reaction must be trapped to promote the formation of graphite; this becomes increasingly important for the preparation of micro-samples [9]. In our standard reactors we use a tube furnace to heat 1–2 mg of iron catalyst in a Pyrex reaction tube and a two stage Peltier cell to cool a glass cold finger to −40 °C to trap water. A modified Swagelok UltraTorr fitting is used to seal the tubes in this system. The Fe catalyst is firstly prepared by reduction under 750 mbar H_2 at 600 °C for >1.0 h. The furnace and cold finger temperatures and reactor pressure (Sensortechnics stainless steel diaphragm piezoresistive gauge head) are monitored and logged. We operate a bank of 24 such furnaces for which we have minimised the internal volume to ~2.5 mL in order to maximise reaction pressure for more efficient graphitisation of 'micro-samples', samples which contain up to a few tens of micrograms of carbon. Samples containing ~10 μg of carbon can be readily prepared. Larger samples are prepared by using a larger cold finger.

There are a number of critical aspects to producing graphite from ultra-small CO_2 samples. Firstly, since the reaction is pressure-dependent the reactor volume must be minimised. Secondly, the Fe catalyst must be carefully chosen: it must be inherently pure (no C or Li), it must have a large and reactive surface area and it must have the correct mesh size. We have found that firstly oxidising the Fe catalyst with 500 mbar O_2 at 450 °C for 0.5 h then reducing it under H_2 as usual, can improve catalyst performance [3]. Thirdly, the H_2O produced in the reaction must be efficiently trapped [9]. The efficiency of H_2O trapping effectively dictates the reaction speed, whereas the reaction pressure and amount of catalyst essentially dictate reaction efficiency (yields). Finally, the quantity of extraneous carbon introduced into the sample must be minimised; this becomes more critical as sample size decreases.

2. Laser-heated microfurnace

Taking these ideas into consideration, a novel microfurnace aimed at reliable and efficient graphitisation of samples containing ~5 μg of carbon has been developed [10]. The challenge is to max-

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imise sample pressure and to minimise the addition of extraneous carbon. We use the same chemical reaction to produce the graphite, however a new approach to furnace design was required in order to reduce the reaction volume below 2.5 mL. Fig. 1 shows the basic microfurnace. A 25 W (continuous) fibre-packaged array infrared laser (805 nm Coherent) is coupled by an armoured fibre optic cable to a lens assembly mounted ~20 mm directly above a quartz window. The lens focuses the IR radiation to a ~1 mm spot on the Fe catalyst which is contained in a disposable quartz crucible, the only component which is significantly heated during the reaction. The crucibles are 4 mm diameter, 4 mm high and have either a hemispherical (1 mm radius) or conical (1 mm radius, 2 mm high) cavity. An infrared thermometer (1600 nm Raytek), focused to 1 mm diameter, views the heated Fe catalyst at angle of 30° to the normal through the quartz window. Both the laser and the IR thermometer are provided with visible aiming beams.

A stainless steel cold finger is welded to the furnace body, eliminating the need for an o'ring seal. An integrated valve isolates the furnace once the CO_2 and H_2 have been admitted and a sub-miniature stainless steel diaphragm gauge (Entran) is used to monitor the pressure in the reaction vessel. All materials in the reaction vessel are either stainless steel or quartz, and the window, gauge head and valve are sealed with Viton o'rings. Total internal volume of the prototype reaction vessel is just 0.5 mL, usable for a maximum sample size of ~200 μg of carbon as CO_2. The furnace is enclosed in a light proof box, fitted with electrical interlocks, for safe operation of the class 4 laser. The catalyst temperature, furnace body temperature and furnace pressure are monitored and logged by a LabVIEW interface.

To date, 67 samples have been produced with the prototype microfurnace. The first 24 were prepared from OX1 and ^{14}C-depleted CO_2 to develop the microfurnace and target loading techniques and are not considered further. The other 43 samples shown in Table 1 were all pressed into targets using the rear-loading technique discussed below. Of these, 19 were prepared from a cylinder of 'modern' CO_2 ('Manildra', sourced from an alcohol distillation plant) and 24 were prepared from two different cylinders of ^{14}C-depleted CO_2 (12 from each). Typically 0.4–0.6 mg of Sigma–Aldrich −400 Fe catalyst was used, firstly prepared by heating for >15 min under ~700 mbar of H_2 in a fresh quartz crucible which was pre-cleaned by baking in oxygen.

During a typical graphitisation, the cold finger (CF) temperature is changed between room temperature, −190 °C (using liquid nitrogen) and −65 ± 5 °C (using a thick aluminium rod immersed in an alcohol and dry ice mixture). The furnace is firstly evacuated then sample CO_2 is admitted and trapped with the CF at −190 °C. The microfurnace is isolated, the CF warmed and the amount of CO_2 volumetrically determined. The CO_2 is trapped again with CF at −190 °C, H_2 is added in excess and the microfurnace isolated. With the CF at −65 °C laser power is applied to rapidly bring the catalyst to the desired temperature, normally between 500 and 600 °C. The CF is maintained at −65 °C during the reaction to trap water vapour. To maintain a constant temperature, it is necessary to reduce the laser power as the reaction proceeds since convective losses change within the furnace as the reacting gases are consumed. Reactions are typically complete within 15–30 min although longer reaction times were sometimes used.

Following completion of the reaction, we estimate the efficiency of CO_2 conversion under the assumption that the residual gas is a mixture of H_2, CO_2 and H_2O. Firstly we measure the pressure of the residual gas, and then take the CF to −190 °C, trapping CO_2 and H_2O and pumping away H_2. The CF is warmed and the pressure measured. Next the CF is taken to −65 °C, trapping H_2O and pumping away CO_2. The CF is then warmed and the pressure measured. The latter two pressures always ~23.5 mbar, the vapour pressure of H_2O at 20 °C, suggesting that no measurable CO_2 remains and that it has all been converted to graphite.

3. Sample loading

We have developed a specially designed target holder to simplify and minimise the handling of extremely small samples and to guarantee optimum sample density and surface finish for best ion source performance. This is an adaptation of the sample pressing method used by National Electrostatic Corporation (NEC) with the SNICS series of ion sources.

The graphitised sample and catalyst are pressed into a 1.00 mm diameter bore hole in a detachable aluminium target cap. Firstly, the cap is placed upside down on a ground, hardened steel pin and the assembly is placed in a die. This lower pin protrudes ~0.5 mm into the bore. To avoid sample loss, a small stainless steel funnel can be screwed onto the cap to extend a conical surface machined into the back of the cap. The sample is loaded by tipping it directly from the quartz crucible into the funnel, under a microscope. Sometimes an electric vibrator or the 99.95% pure alumin-

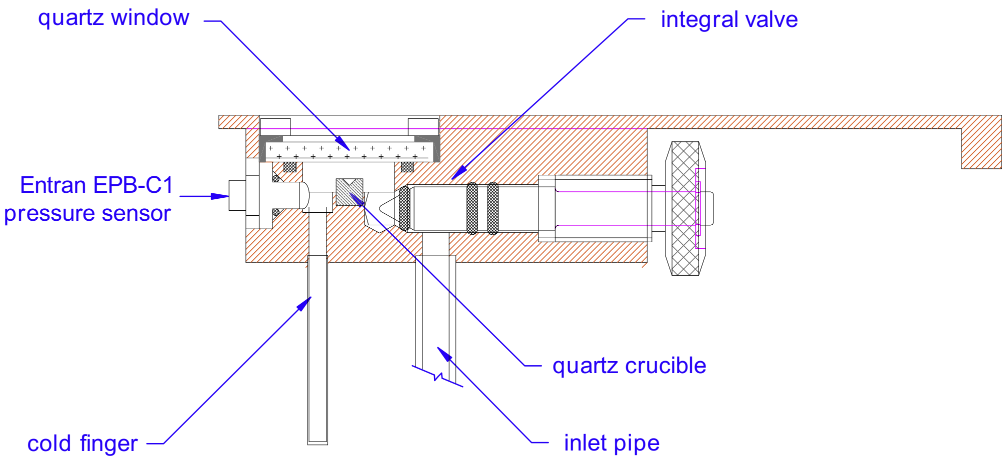

Fig. 1. The basic microfurnace. The body is machined from stainless steel and the 3 mm diameter stainless steel cold finger is fusion welded to the body. An integrated valve isolates the microfurnace from the inlet manifold and the brass valve spindle is sealed with three Viton o'rings. A 1.6 mm thick quartz window and a sub-miniature stainless steel diaphragm gauge are also sealed with Viton o'rings. The quartz crucible containing the Fe catalyst is located in a recess, directly below the lens assembly which focuses the infrared laser beam through the window to a ~1 mm spot. The heated catalyst is viewed through the window by an infrared thermometer, also focused to a ~1 mm spot. Total internal volume is ~0.5 mL.

Table 1

Samples prepared in the microfurnace, listed in order of preparation, with boxed samples prepared about the same time. Three different CO_2 types were used: M = Manildra (modern), while D1 and D2 were two different cylinders of ^{14}C-depleted CO_2. Graphitisation times were varied and often exceeded the average time of ~15 min needed for completion of the reaction. All samples except #1 (see Note 1) were prepared with Sigma–Aldrich −400 Fe (99.99%) catalyst, reduced under ~700 mb H_2 for >15 min. All used a cold trap maintained at 65 ± 5 °C to trap water produced during the reaction. All except #9 used hemispherical crucibles and all were new, cleaned by baking under oxygen, the exception being the crucible for #6 which was re-used without cleaning for samples #7 and #8. All results have been normalized to the mean of large OX1 and OX2 and were corrected for machine background using the measured value of ^{14}C-depleted graphite; no further corrections have been applied.

Date	Sample ID	Note	CO_2 type	Mass C (μg)	Error (μg)	Mass Fe (mg)	C/Fe (μg/mg)	Mean Fe temp (°C)	Reaction time (min)	Measurement time (s)	Mean $^{13}C^{4+}$ (nA)	pMC	Error
February 2007	1	1	M	8.28	0.09	0.74	11	560	85	300	85.0		
	2	5	M	5.18	0.06	1.15	5	576	23	300	0.3		
	3	5	M	6.17	0.07	1.09	6	529	54	300	1.4		
	4		M	6.61	0.07	0.85	8	551	29	540	22.4	98.18	2.67
	5		M	6.10	0.07	0.67	9	552	48	1200	41.0	100.96	1.25
	6		M	5.29	0.06	0.69	8	500	19	600	19.5	101.02	2.39
	7		M	6.79	0.07	0.76	9	605	23	900	29.5	102.96	1.98
	8		M	5.45	0.06	0.30	18	502	58	1200	37.0	102.09	1.24
	9		D1	5.71	0.06	0.93	6	567	47	337	24.9	2.28	0.60
	10		D1	5.68	0.06	0.59	10	570	43	300	21.2	3.78	0.63
	11	5, 6	D1	5.81	0.06	1.03	6	555	25	255	0.0		
June 2007	12	4	D1	5.51	0.06	0.97	6	482	27	2000	24.4	17.45	1.44
	13		D1	4.34	0.05	0.92	5	573	15	2000	33.1	4.73	0.53
	14		D1	3.53	0.04	0.23	15	511	34	1400	30.9	3.11	0.31
	15		D1	9.22	0.10	0.46	20	529	30	2000	54.4	1.66	0.13
	16		D1	12.41	0.13	0.44	28	573	44	2000	115.4	1.00	0.08
	17	2	D1	12.29	0.13	0.52	24	495	3	1400	39.4	1.13	0.13
	18		D1	52.57	0.57	0.48	110	577	28	2000	303.7	0.25	0.02
	19	4	D1	20.48	0.22	0.41	50	591	127	1400	153.9	7.01	0.21
	20	4	D1	10.82	0.12	0.40	27	580	26	1400	94.6	8.09	0.27
	21	3	M	5.47	0.06	0.64	9	570	18	300	32.6	94.99	2.62
	22	3	M	5.12	?	0.26	20	545	44	600	18.4	95.07	3.00
August 2008	23		D2	100.60	1.08	0.50	201	600	49	1160	585.8	0.37	0.04
	24		D2	101.50	1.09	0.56	181	643	36	1200	460.9	0.97	0.11
	25		D2	50.53	0.54	0.49	103	607	40	1180	384.2	1.09	0.07
	26		D2	50.04	0.54	0.62	81	609	31	1200	335.8	1.34	0.11
	27		D2	20.23	0.22	0.45	45	610	32	1180	228.1	1.44	0.10
	28		D2	20.06	0.22	0.62	32	634	42	1200	104.2	2.69	0.20
	29		D2	10.09	0.11	0.41	25	570	42	1200	118.9	4.18	0.38
	30		D2	9.86	0.11	0.45	22	612	25	880	121.5	5.84	0.51
	31		D2	5.06	0.05	0.41	12	617	17	900	81.1	18.45	1.20
	32	4	D2	5.16	0.06	0.42	12	613	48	860	62.1	32.08	1.17
	33	4	D2	2.54	0.03	0.52	5	559	46	900	24.8	48.83	10.24
	34		D2	2.48	0.03	0.52	5	606	30	860	15.2	25.60	1.72
September 2008	35		M	87.00	0.94	0.46	189	614	36	2400	332.7	105.77	0.33
	36		M	77.88	0.84	0.62	126	602	46	2400	304.9	106.77	0.56
	37		M	5.35	0.06	0.57	9	608	24	600	93.2	103.33	1.16
	38		M	4.89	0.05	0.55	9	610	30	600	112.5	103.78	1.06
	39		M	4.73	0.05	0.45	11	599	40	600	91.5	104.93	1.18
	40		M	5.00	0.05	0.56	9	599	53	600	110.2	105.88	1.08
	41		M	5.24	0.06	0.60	9	601	38	600	104.2	104.17	1.40
	42		M	4.82	0.05	0.55	9	617	41	600	99.0	103.54	1.12
	43		M	4.90	0.05	0.67	7	508	47	600	117.2	105.52	1.08

Notes:

1. This sample was deliberately graphitised for a long time to check on completion of reaction; Alfa-Aesar Fe (98%) catalyst was used. This catalyst was found to have unacceptably high lithium levels which overloaded the ionisation detector.
2. This reaction was deliberately stopped prior to completion.
3. These samples were used to test the laser following repair.
4. These results were discarded from the 1/*m* analysis.
5. No result obtained due to low current.
6. Oxidised in O_2 prior to reduction in H_2.

ium wire plug, held in tweezers, are used to aid loading. Once the entire sample has been loaded the plug is guided into the bore and the assembly is lifted onto the anvil of a hand press, fitted with a pressure indicator. The die is then moved until a long, hardened steel pin cleanly enters the bore and a pressure of 14 bar compresses the sample between the plug and the lower pin, providing a good quality surface and cold-welding the aluminium wire to the bore of the cap. At this stage it is safe to disassemble the components and to screw the cap onto the labelled target base.

We have used this system for loading BeO samples for ^{10}Be AMS and have tested it with Al_2O_3 for ^{26}Al AMS. In both cases the oxides are mixed with Nb powder to improve electrical and thermal conductivity. Variants of the detachable targets have been made from copper and aluminium with bore diameters of 1.0 and 1.6 mm. A 0.5 mm bore version is under development for ^{14}C micro-samples. Additionally, stainless steel target bases have been produced which can be re-used.

4. Ion source alignment

The primary cesium sputtering beam has a diameter of ~0.3 mm and the target is generally rotated under the cesium beam in a circle of radius 0.5 mm to optimise the use of sample material. With a target diameter of just 1.00 mm, good alignment between the cesium beam and the target center is needed. Following routine ion source service, we have found it necessary to check and adjust this alignment; the basic design of the ion source, coupled with worn, jet-abraded parts means that alignment jigs do not guarantee a sufficiently good alignment.

We firstly tried an alignment target incorporating a 0.3 mm diameter clutch pencil lead. This alignment target was moved in a ±0.5 mm XY grid under the cesium beam to generate a spatial ^{12}C contour map. This approach was unreliable and often gave a distorted map due to uneven sputtering of the carbon and because the Al cap sputters preferentially, leaving a raised carbon 'post' behind.

Currently we use the 1.0 mm diameter detachable targets for this purpose, either pressing Al wire into a Cu cap or a Cu wire into an Al cap. Spatial maps of ^{27}Al or ^{63}Cu are generated by moving the alignment target in a grid under the cesium beam and adjusting the position until the target is centered with the cesium beam.

All samples containing less than 50 μg of carbon are measured on the ANTARES accelerator because it provides higher yields of the measured charge state (4+) than does STAR (3+) and because it has better digitisation of low currents than does STAR. Presently, the $^{13}C^{4+}$ beam is used on ANTARES to derive $^{14}C/^{13}C$ and hence $^{14}C/^{12}C$, due to problems measuring the $^{12}C^{4+}$ beam.

5. Performance with 5 μg 'modern' CO_2 samples

Table 1 summarises results obtained from sets of ~5 μg and some larger Manildra samples, prepared in the prototype microfurnace on two different occasions (samples M). The expected activity for Manildra CO_2 is 105.51 ± 0.74 pMC. All samples were measured on ANTARES and were normalized to large-sized OX1 and OX2 samples prepared in our standard graphitisation furnaces. Measurements were considered unreliable when the $^{13}C^{4+}$ current fell below ~20 nA and this data was discarded from the analysis. Samples typically produced good currents for 10 min or more, generally long enough to obtain sufficient ^{14}C counts for a Poisson error of ~1% or less.

The first set of 8–5 μg Manildra samples were prepared in February 2007; samples 2 and 3 failed to produce useful currents. Sample 1 was the only sample in Table 1 prepared with Alfa-Aesar −325 (98%) Fe catalyst which was found to have an unacceptably high Li content and could not be measured. All other samples were prepared with Sigma–Aldrich −400 (99.99%) Fe catalyst. The remaining five samples gave an error weighted mean of 101.42 ± 0.73 pMC. The second set of 7–5 μg samples were prepared in September 2008 and gave an error weighted mean of 104.48 ± 0.43 pMC. Two larger samples (~90 μg) prepared at the same time measured near the nominal value. We also observe a lowering of the $^{14}C/^{12}C$ ratio for micro-samples prepared from reference materials in our standard furnaces. For this reason it is our practice to normalize the measured values of micro-samples against the measured values of commensurately-sized reference samples.

There are a number of possible explanations for measuring a decreased $^{14}C/^{12}C$ ratio. One possibility is that there is addition of less-active extraneous carbon during sample preparation which may noticeably affect ultra-small samples. Another possibility is isotopic fractionation during sample processing, but this is unlikely as we seem to see near 100% graphite yields. There may also be accelerator-related explanations, including fractionation in the ion source [11], current-dependent accelerator transmission or problems measuring the low, pulsed currents obtained from micro-samples with isotopic bouncing.

In order to investigate this further, five test samples containing ~3 μg of carbon per 0.5 mg of Fe catalyst were prepared by mixing 200 μg from a homogenised large OX1 sample (3.78 mg of C on 2.7 mg Cerac −325 Fe catalyst) with 22 mg of H_2-reduced Cerac −325 Fe catalyst. These samples produced 25–60 nA of $^{13}C^{4+}$ and gave an error weighted mean of 101.03 ± 0.72 pMC when normalized to the remainder of the parent OX1. This is comparable to normal experience with directly-prepared small samples and demonstrates that the majority of the fractionation occurs in the accelerator, rather than during sample preparation.

Tests with a precision current source have shown that steady (DC) currents of just a few nA can be reliably measured on ANTARES. The stable currents obtained from the test samples allowed us to check the linearity of the charge measurement system for the low-current pulsed $^{13}C^{4+}$ beams obtained with isotope bouncing (typically 2 ms ^{12}C, 10 ms ^{13}C and 188 ms ^{14}C) for micro-samples. No obvious problems were identified. Although there may be current-dependent accelerator transmission, we feel this is unlikely and that fractionation during the sputtering process in the ion source is the underlying explanation, due to the wide range of C:Fe ratios between micro-samples and larger-sized samples.

6. Extraneous carbon

A set of samples of varying masses were prepared in the prototype microfurnace in June 2007 from a cylinder of ^{14}C-depleted CO_2 purchased from Matheson (samples D1). Large samples prepared from this material in the standard furnace measured 0.50 ± 0.01 pMC. The results are shown in Table 1. Most samples gave a linear relationship between pMC and $1/m$, where m is the mass of carbon in the sample, that is consistent with the addition of 0.15 μg of 100 pMC carbon. Three outlier samples identified in Table 1 did not fall on this line and were rejected from the analysis.

A second cylinder (samples D2) of more ^{14}C-depleted CO_2 was obtained from BOC. Large samples prepared from this material in the standard furnace measured 0.08 ± 0.01 pMC. A set of samples were prepared in the prototype microfurnace in August 2008, in sequence from largest to smallest mass. The results are shown in Table 1. Most samples gave a linear relationship between $1/m$ and pMC, consistent with the addition of 0.70 μg of 100 pMC carbon. Two outlier samples identified in Table 1 did not fall on this line and were rejected from the analysis.

The increase in the apparent amount of extraneous carbon was disappointing, but perhaps not too surprising. Before it was first assembled, the microfurnace was only cleaned with solvents. It has never been cleaned since, and Manildra and OX1 sample have been prepared in it along with the blanks. It is likely that the extraneous carbon added to each sample varied in its activity; 100 pMC carbon was assumed for the above analyses but there were outliers that did not fit the trend. Table 1 shows that the first set of Manildra samples was prepared in February 2007 and the first set of blank samples was prepared shortly afterwards in June 2007. It is possible that more than 0.15 μg of carbon with a lesser activity than 100 pMC was added and that this is responsible for the lower mean of 101.42 ± 0.73 pMC obtained for this set of Manildra samples. The second set of blank sample was prepared in August 2008 and the second set of Manildra samples in September 2008, both by a different operator. The latter set gave a mean of 104.48 ± 0.43, close to the expected value. This could be due to the addition of extraneous carbon with a similar activity, which would not be noticeable. Another factor could be that the average $^{13}C^{4+}$ currents for the first set of Manildra samples were ~½ to ⅓ those of the second set of Manildra samples.

7. Discussion and future plans

Virtually all 67 samples prepared with the prototype microfurnace, even at the 5 μg level, have produced good currents and have demonstrated adequate longevity. With modern samples, ~10,000 ^{14}C atoms can be readily detected before measurement becomes unreliable, giving a statistical precision of ~1%.

The largest challenge is to understand the sources of extraneous carbon and to minimise and characterise them. This will require a systematic approach, using separate furnaces for ^{14}C-depleted and for modern samples, and adopting cleaning protocols. We have already implemented feedback control of the laser power to maintain a set temperature as the reaction proceeds and gas composition and pressure changes. Along with automation of the entire process, this will enable more careful experimentation for future sample preparation. We are introducing a heater so that the cold finger can be baked between samples. It may be desirable to introduce an oxidation step to further cleanse and condition the catalyst before reduction. We also need to improve the microfurnace pressure measurement since the zero drifts in the existing gauge and it is affected by H_2; this limits our ability to make reliable measurement of the residual gas pressures. We are investigating ways of directly analysing the residual gas composition to better determine the efficiency of the graphitisation reaction.

Following operating experience with the prototype unit, work has commenced on the development of a second-generation device with multiple furnaces designed for easier operation and to facilitate cleaning. The goal is fully automated operation through a LabVIEW interface of oxidation, reduction & graphitisation. This system will use a 30 W infrared laser system (807 nm nLIGHT) featuring a 90° mirror and lens for a compact design with a simple IR thermometer for temperature measurement. A temperature control system (Linkam) has been adapted for automatic control of cold finger temperature over the normal operating range −196 °C to room temperature and at elevated temperature for vacuum baking. Both of these systems have been purchased and are currently under test.

Effort is underway to improve measurement of the more intense $^{12}C^{4+}$ beam on ANTARES. Using $^{12}C^{4+}$, rather than $^{13}C^{4+}$ for AMS measurements will increase the effective measurement time and so improve the precision. Furthermore, it is desirable to measure $\delta^{13}C$ on the accelerator as this is the only opportunity to do so with such small samples.

Decreasing the amount of catalyst will also serve to increase beam currents. We plan to experiment with ~0.2 mg of catalyst but this will need a 0.5 mm diameter bore in the target cap for reliable loading. With such a small diameter target it will be unnecessary to move the target under the cesium beam during measurement as we currently do.

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