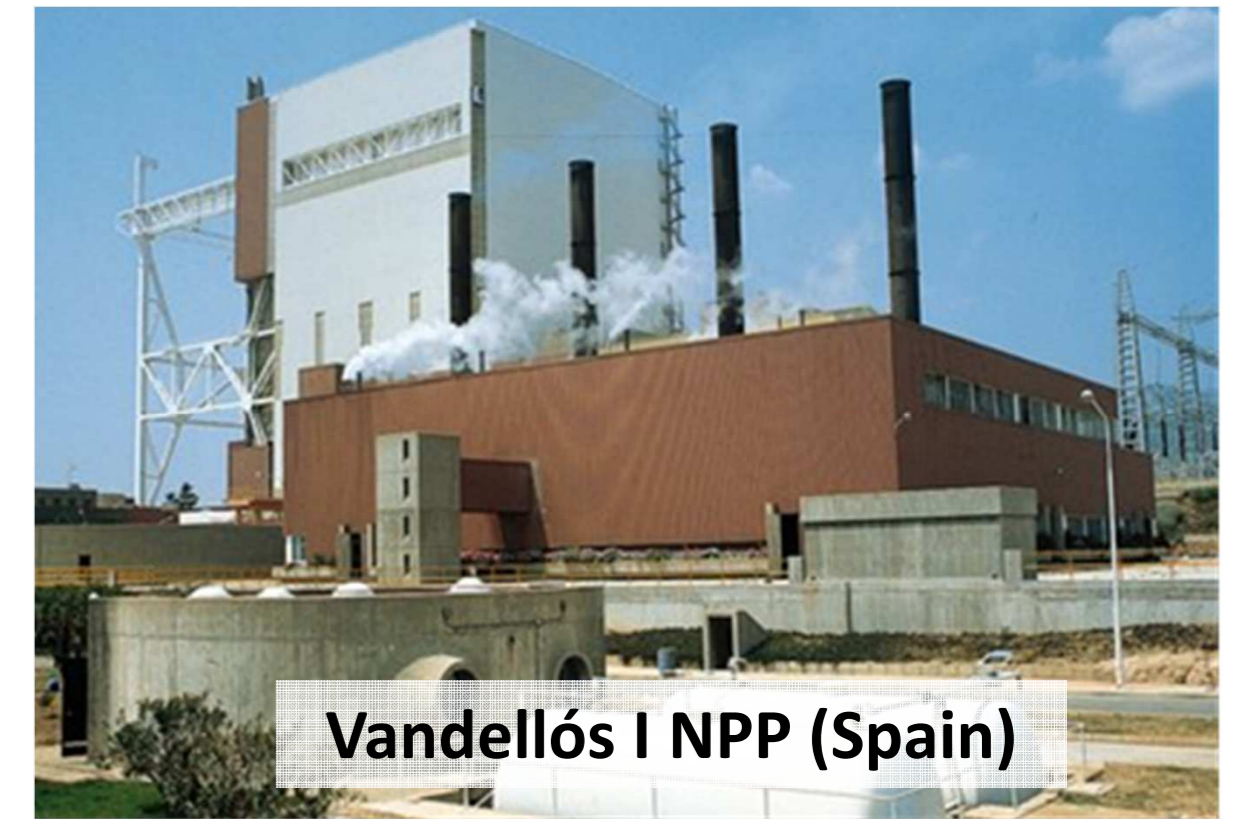


¹⁴C LEACHING AND SPECIATION STUDIES ON IRRADIATED GRAPHITE FROM VANDELLÓS I NPP

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CONTEXT OF THE STUDIES

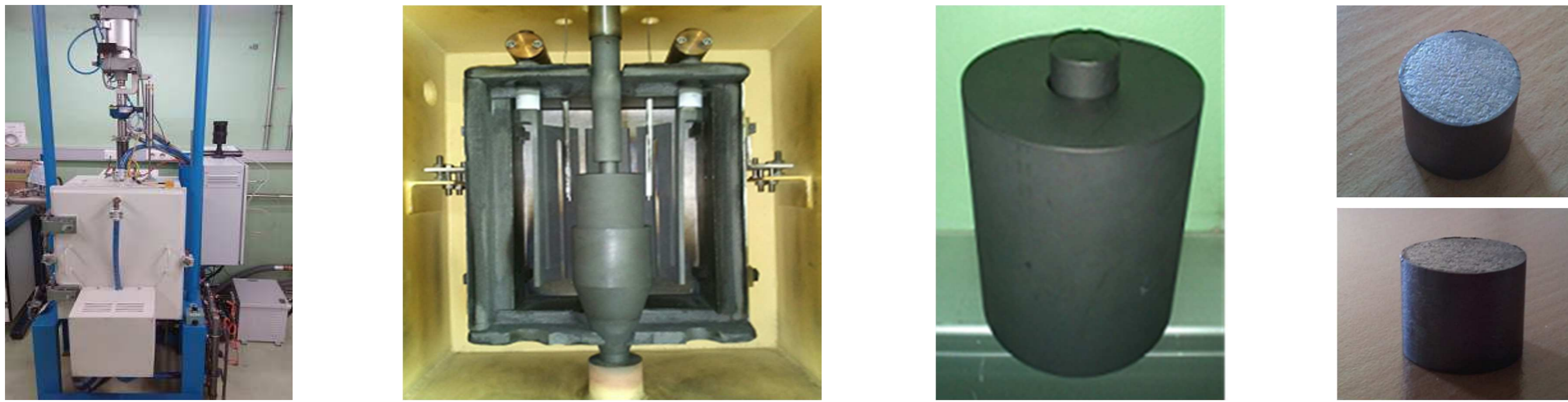
- One of the **R&D activities planned in the Spanish context** for the next years should provide support for the drawing up and/or **revision of the radwaste packages management strategies**. These activities should be based on the **better knowledge of the** physical, chemical, environmental and radiological properties of such **wastes**.
- The understanding of the **¹⁴C behavior in waste packages** (main objective of the CAST project) could lead to a **reevaluation of the near surface repository** for the disposal of wastes containing this radionuclide in high concentrations.
- The **CIEMAT contribution to CAST** focused on the **release of ¹⁴C as dissolved and gaseous species from irradiated graphite** from Vandellós I NPP **testing two types of samples; sleeve graphite core samples** and powder graphite solidified and encapsulated in the form of cylinders in a new glass matrix called Impermeable Graphite Matrix (**IGM**).



MATERIALS AND METHODS

SAMPLES TYPES & PREPARATION

IGM SAMPLES



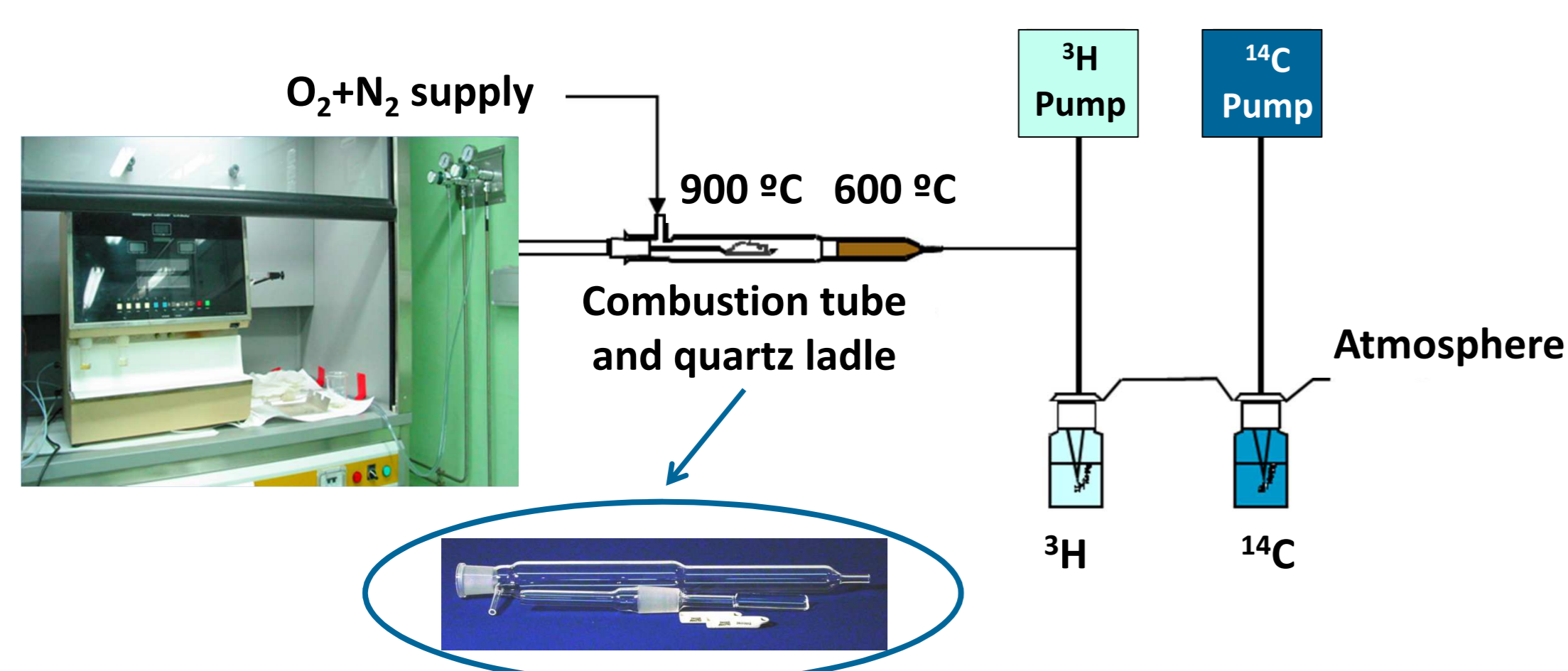
IGM samples of 20 mm x 10 mm (Ø x H) (right) are **manufactured in a hot vacuum press** (left) where a mixture (8:2) of powder graphite (ca. 6 g Ø ≤ 250 μm) and glass matrix is introduced into a **pressing tool mounted into the furnace** (middle).

CORE GRAPHITE SAMPLES



To the right core - drilled sample of ca. 2 g and 11 mm x 12 mm (Ø x H) from **Vandellós I sleeve graphite (UNGG reactor)**. Bulk sample to the left.

CHARACTERIZATION OF THE INITIAL SAMPLE



Ca. 20 mg of powder sample is burned in a **combustion oven able to transform**, by means of a catalyst bed (CuO, Pt), **carbon compounds to CO₂** which is **trapped in a suitable scintillation cocktail** for its further **LSC analyses** in a low background system (Quantulus)

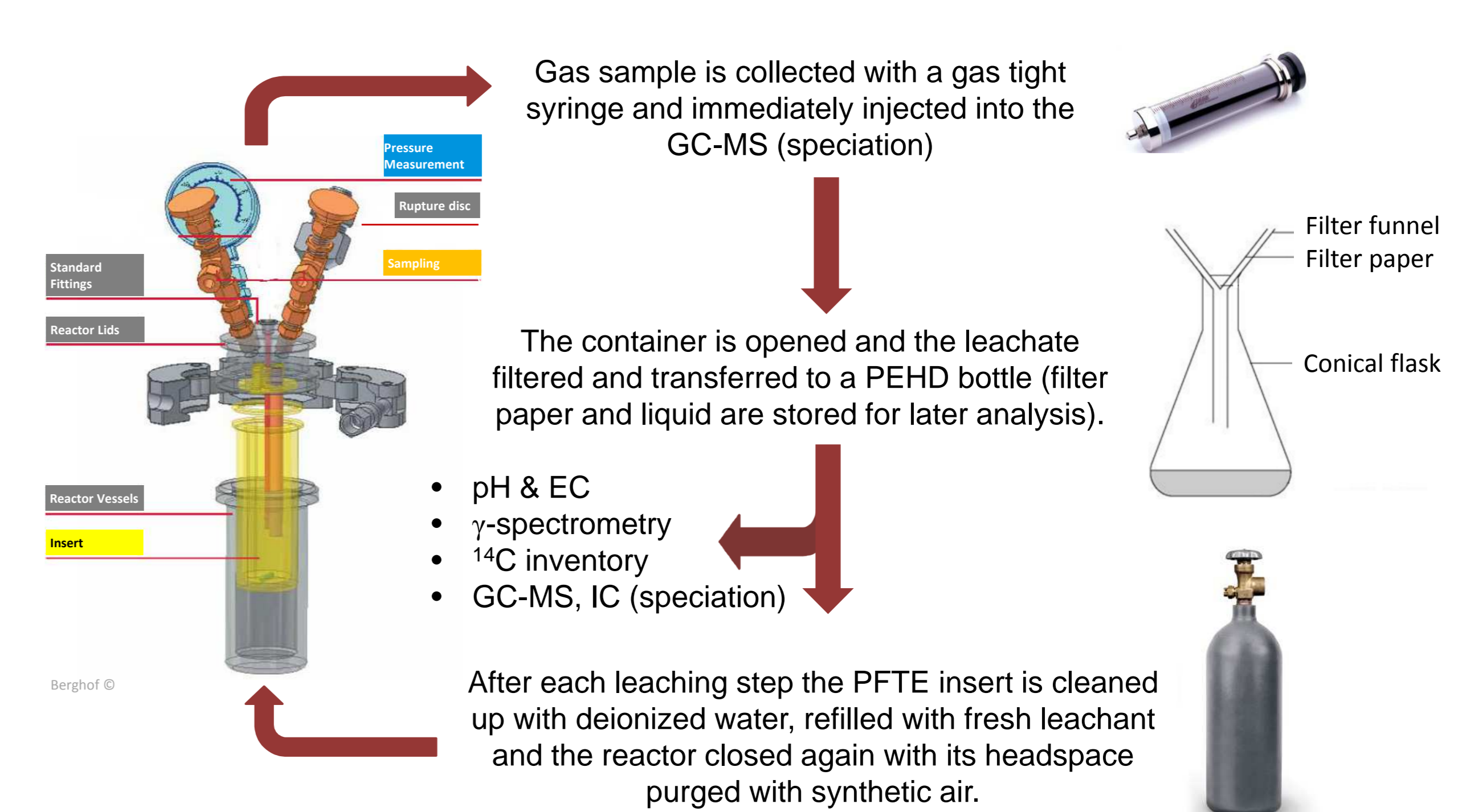
Sample	Activity (Bq/g)					
	¹⁴ C	2U (%)	⁶⁰ Co	2U (%)	¹³⁷ Cs	2U (%)
V-I-1	1,22E+04	7,42	5,56E+01	3,57	2,03E+03	6,27
V-I-2	1,35E+04	7,34	4,98E+01	3,71	3,98E+02	6,28
IGM-4	1,13E+04	7,26	8,39E+01	5,70	1,14E+02	12,07
IGM-5	1,13E+04	7,27	8,49E+01	5,70	1,19E+02	12,02

LEACHING PROCESS

PARAMETER	LEACHING CONDITIONS
Specimen geometry	Graphite Cylinder 11mm x 12 mm (Ø x H) IGM cylinder 20 mm x 10 mm (Ø x H)
Leachant	Graphite: Pure / GBW (synthetic)
Temp. (°C)	Room temperature
Vessel / Stirring	PTFE / No
V _{leachate} /S _{specimen}	0,1 m - 0,2 m
Initial gas phase	Synthetic air (21 % oxygen, 79 % nitrogen)
Regime	Total renewal of the leachate (static).
Sampling	14, 28, 56, 90, 180 and 360 days from 1 st immersion 14, 14, 28, 34, 90 and 180 leachate age
Analyses	Pre-leaching: Solid phase: ¹⁴ C inventory & γ-spectrometry Leachant: Background ¹⁴ C inventory, pH & EC
	Post-leaching: Leachate: ¹⁴ C inventory, speciation (GC-MS, IC), γ-spectrometry, pH & EC Gas phase: Speciation (GC-MS)
Evaluation of results	Incremental leaching rate as a function of time of leaching (cm/day) $R_v = \frac{a_v}{a_s} \times \frac{V}{S} \times \frac{1}{t_v}$



SAMPLING PROCEDURE & SPECIATION ANALYSES



The methodology to **determine permanent gases (CO, CO₂), C1 - C5 hydrocarbons, alcohols and aldehydes** has been developed using a **GC-MS** (to the right). Short chain **carboxylic acids** have been analyzed with an **Ion Chromatography System (IC)**.



RESULTS & FINAL REMARKS

- Although leaching rates has been determined for ¹³⁷Cs and ⁶⁰Co, **only has been found one value of ¹⁴C above the detection limit** (graphite core sample), and it is more likely that this could be because of bad filtering of the leachate or cross contamination in the equipment than because of the leaching process itself.
- Using deionized water as leachant, in the IC analyses, **acetate, formate and oxalate has been detected after some stages**. However, **this technique cannot be used to analyze GBW solutions** because of the high concentration of anions and cations present in this media.
- Both **alcohols and aldehydes in leachates have not been detected** in any step of the leaching process and, regarding gas samples, **nor was CO**, except for the first and second leaching period of IGM samples using GBW as leachant (this could be related to the carbonate content in the leachate).

