STUDY OF CHEMICAL REACTIONS IN THE NUCLEAR UNDERGROUND EXPLOSION - INCIDENCE ON RADIOACTIVITY

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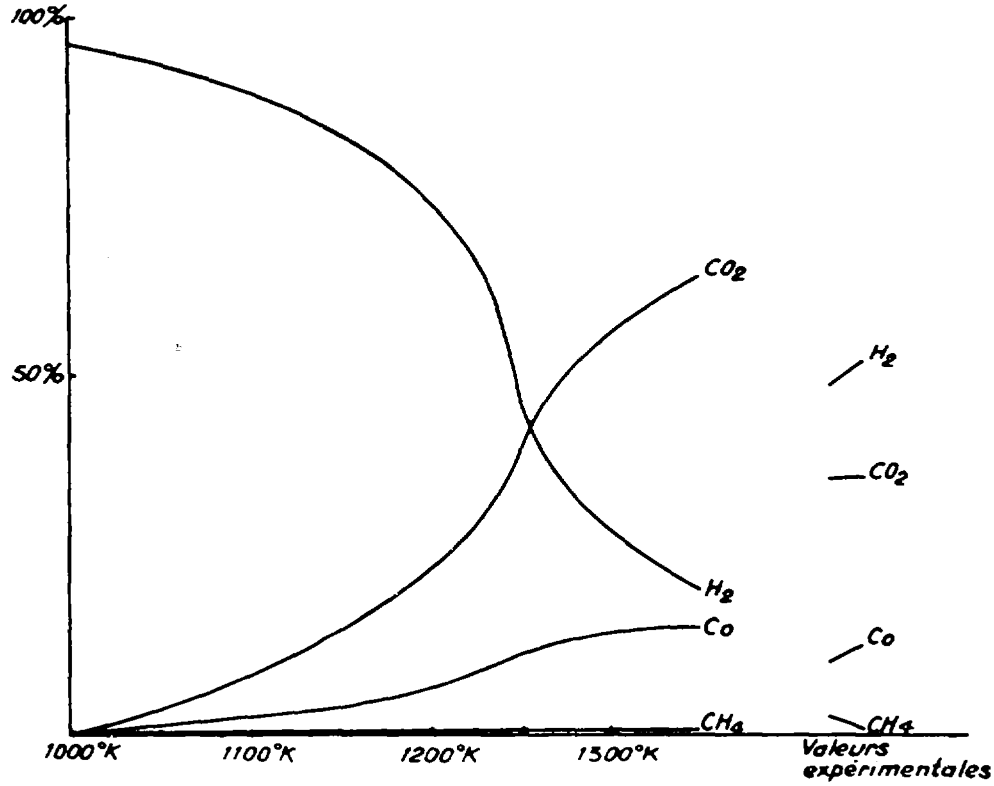
We have been working in order to find out and state the theoretical or semi-empirical laws governing the reaction of radioactivity in contained nuclear explosion. To do so, we are studying the chemical reactions during the different stages of the cavity and chimney formation, as well as thermal transfers. At the same time, we are carrying an experimental study on melted rock and gas samples taken from the French underground explosions. The results of which can be found in this paper are derived from our present experiments at the plant (have been obtained from partial studies).

During the French underground explosions, we took gaseous samples. The gas analysis, without taking water vapour into consideration, showed that those samples were composed of hydrogen, carbon dioxide, carbon monoxide with small quantities of hydrocarbones (chiefly methane - about one per cent). The total amount of gas being quite large and proportional to the burst power, we came to the conclusion that those gases were produced by rock reactions (that rock was granite). We considered the following reagents because they were found in sufficient quantities to alter the balance between the different components : ferrous ions contained in mica, biotite, carbon dioxide from carbonates and water, either free or in a component state, contained in the rock. A comparison between theoretical and experimental results led us to notice among other things (fig. 1).

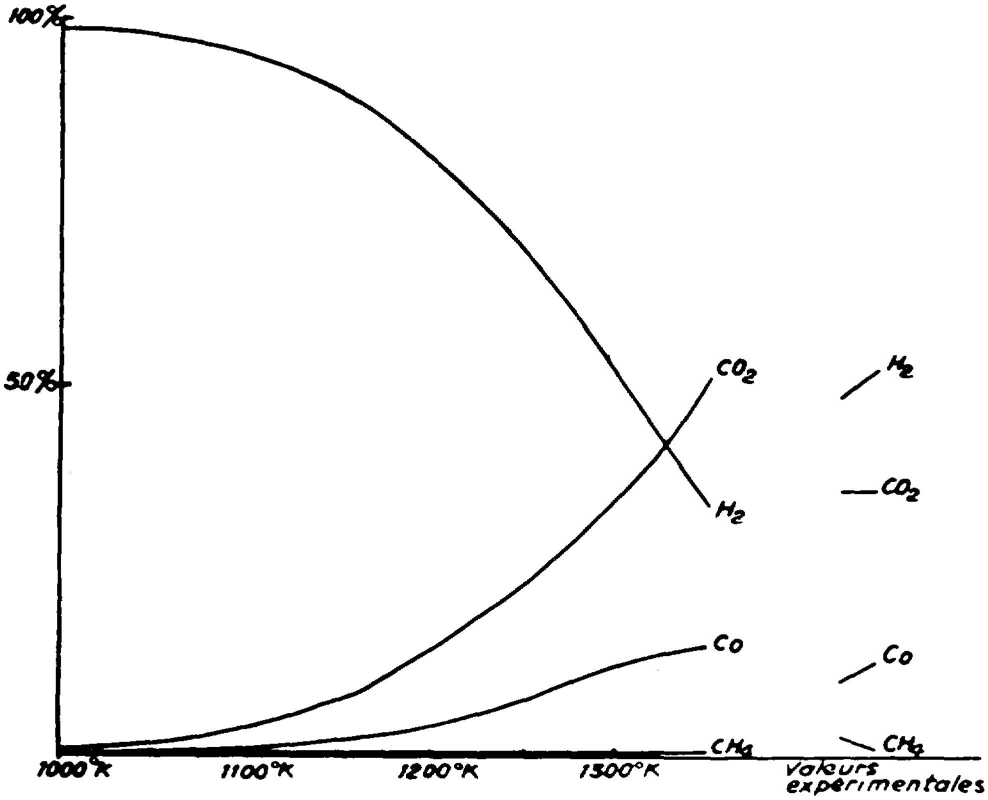
- temperature : the temperature of rock re-solidificating
- pressure : a pressure nearing lithostatic pressure

Since the components of the environment, water not included, is quite homogeneous, the gas volume produced by "1 kiloton" is quite constant. On the other hand, the relative proportion of the gases undergoes a few changes, particularly the ratio CO/CO_2 which normally depends on the quantity of water contained in the environment. This statement is verified by the calculation of thermodynamic equilibriums (figure 2).

In order to work out this calculation of simultaneous chemical equilibrium we have first selected five reactions. Our method of reasoning is as follows : we work by loop, studying each reaction in turn. After a certain number of loops, we reach a stable equilibrium. We felt



. — **Variation des concentrations des principaux gaz (100 atm, gaz secs)'**



— **Variation des concentrations des principaux gaz (200 atm, gaz secs).**

Fig. 1 — Variations of the relative proportion gases with variations of temperature and pressure.

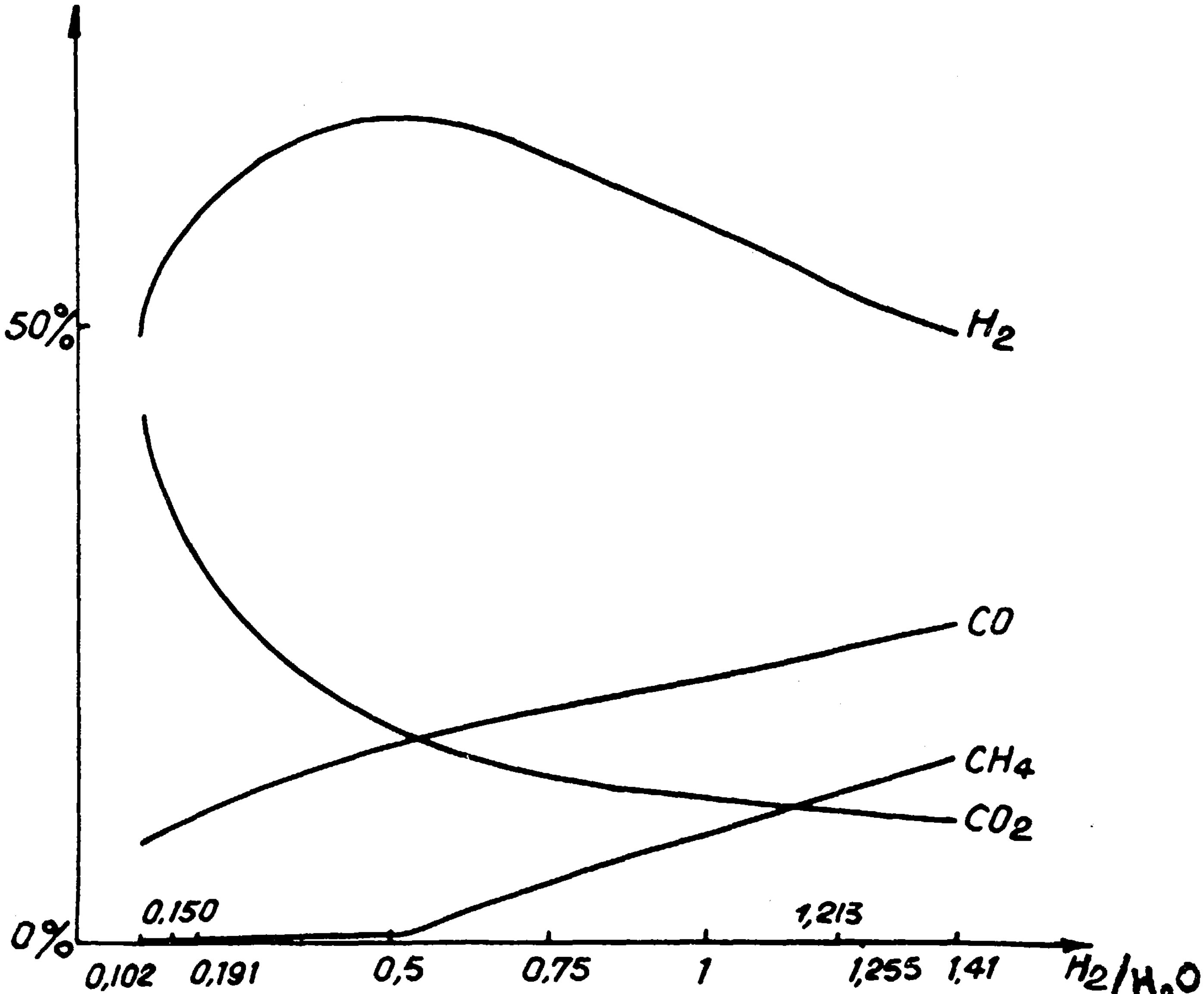


Fig. 2 — Variations of the relative proportion gases with quantity of water contained.

sufficiently satisfied with the results to set a programme in which we take all macroconstituants.

The programme is used for temperature as high as 6000°K. We cannot go beyond that temperature for lack of thermodynamic data. It enables us to study the chemical behaviour of refractory elements. Macroconstituents govern the reactional medium and microconstituents (chiefly radioactive elements) react in that medium.

Another part in our theoretical study is the incidence of chemical reactions inside a chimney on radioactivity. We noticed two main points : long term thermical evolution and chemical reactions in a hydrocarbon environment.

To study thermical evolution, we took the case of granite, where thermical conduction phenomena are preponderant.

In that study, we take as zero time the collapse of the chimney and as source part of a sphere whose radius, for granite, is roughly given by the formula $R = 10\ W^{1/3}$ for a 500 to 1000 m deep explosion. This source has an even temperature : about 900 to 1000°K corresponding to the calorific energy stored up by the rock : calorific energy is about 85 per cent of total energy.

The temperature determined by calculation is in agreement with the measurements taken on the 178th and 221st days after the explosion. These comparisons were made with horizontal drill, the only one we had for the considered explosion. To measure the temperature along a vertical drill, we must consider thermical transfers by convection due to condensable components : water, hydrocarbons... (not very important in the particular case of granite, containing little water)(figure 3).

The chemical reactions in the chimney were studied limitating the temperature range to oil shale decomposition. These cracking reactions as well as the exchange reactions determine the fixation of radioactivity in oil.

To study the rock reactions at high temperature, we have conceived a simulation apparatus based on inductive plasma. In an inductor surrounding the sample the generator produces a current as high as 10 000 V with a 3 to 9 megahertz frequency. The energy stored in the sample is about 10 kJ for a plasma lasting one second. The temperature in the center of the plasma is supposed to be more than 15 000°K. This plasma is made in a hole drilled in a sample of the rock to study. That rock, overheated, is partially vaporized and melted, and it releases gases which are cryogenically trapped out at the end of the heating cycle. Then these gases are analysed either in gaseous phase chromatography or with mass spectrometer (fig. 4 and 5).

We find some difficulties to know the temperature and pressure undergone by the samples to be analysed. The tests were made on different sorts of granite, then on marble chosen because, when heated, it

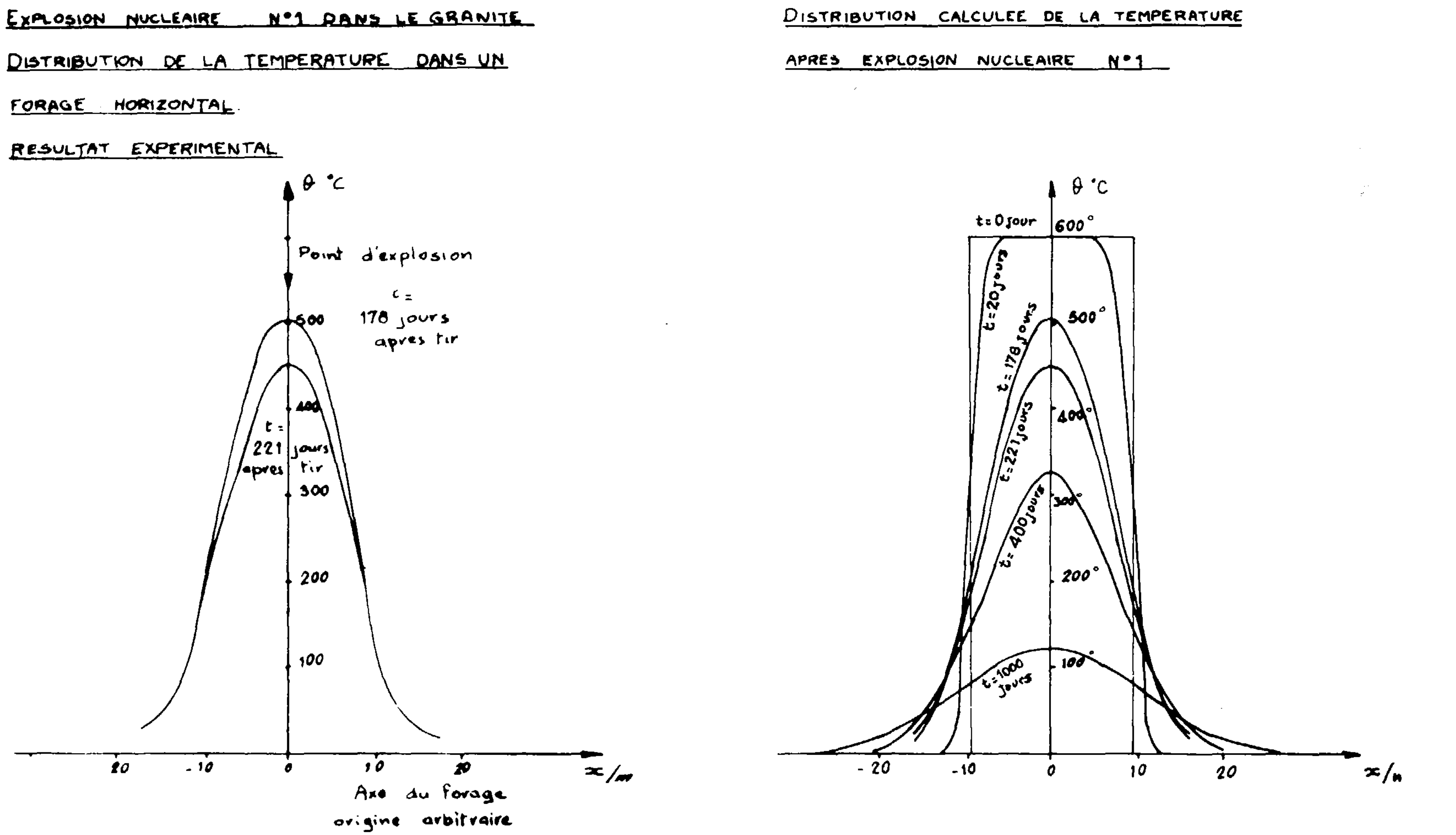


Fig. 3 — Temperatures by calculation and by measurement.

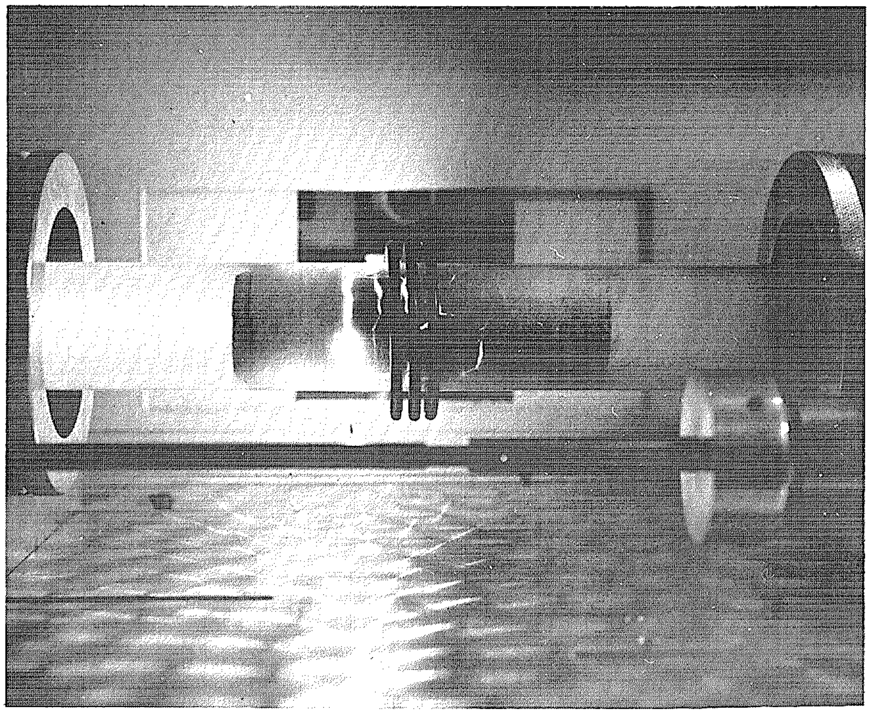


Fig. 4 Simulation apparatus.

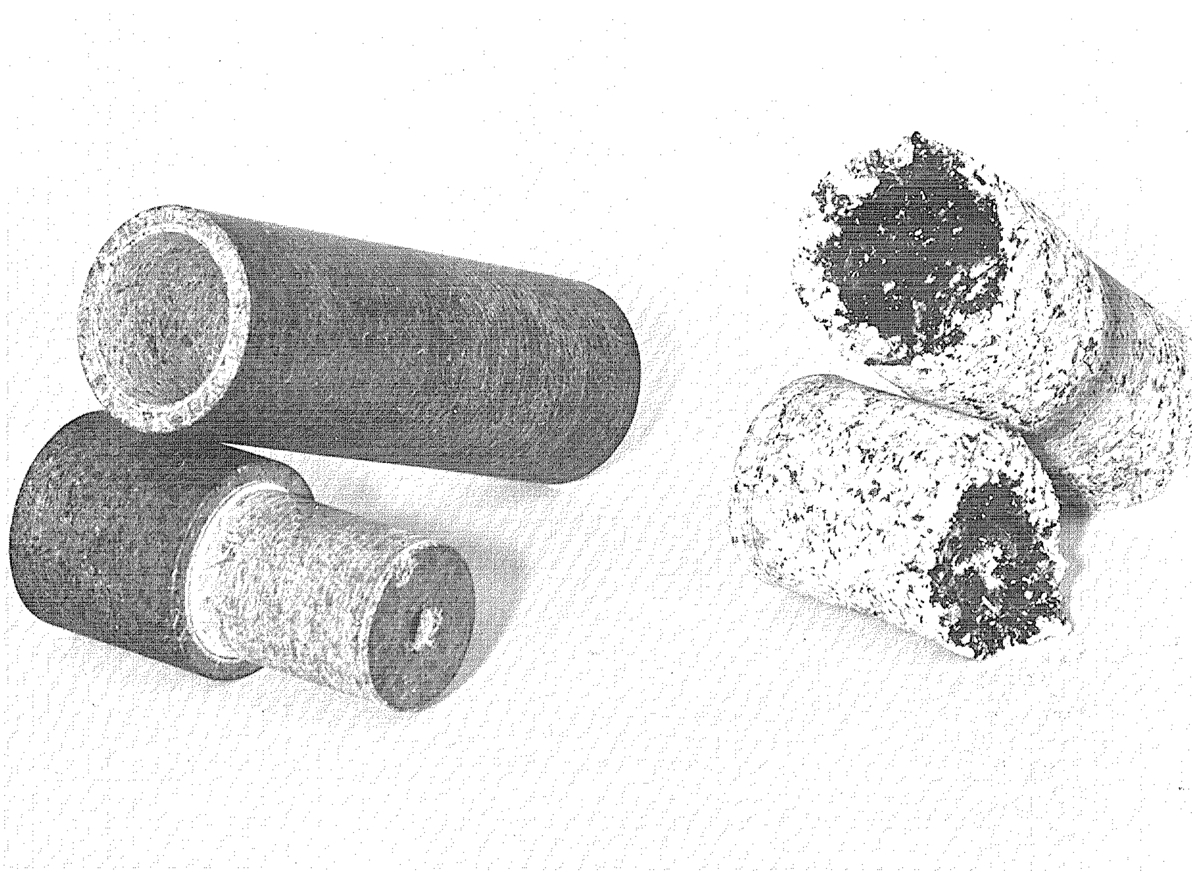


Fig. 5 Samples before and after plasmas heating.

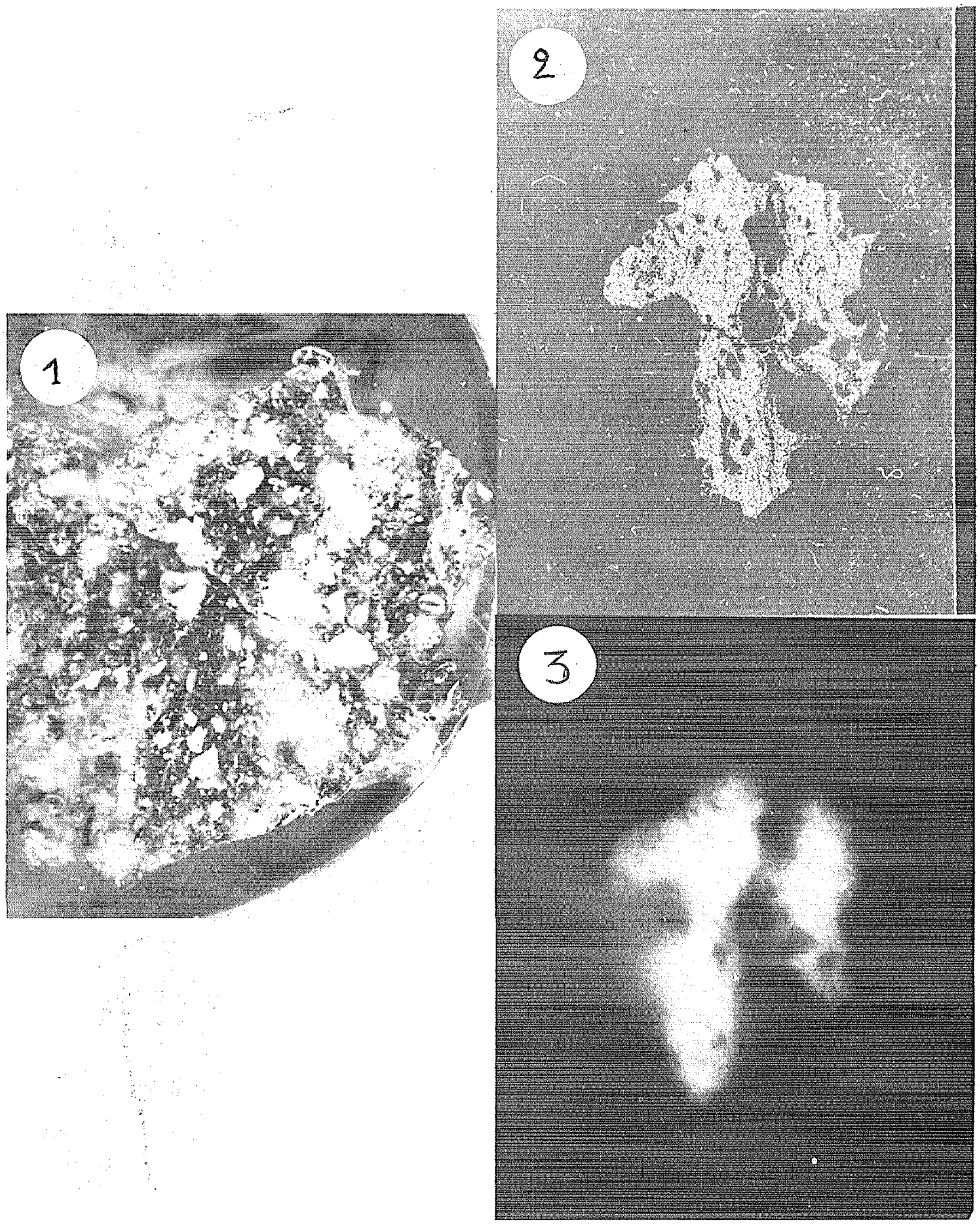


Fig. 6

1 - Photo of the sample
2 - α autoradiography
3 - β-γ autoradiography

Melted rock included no radioactivity

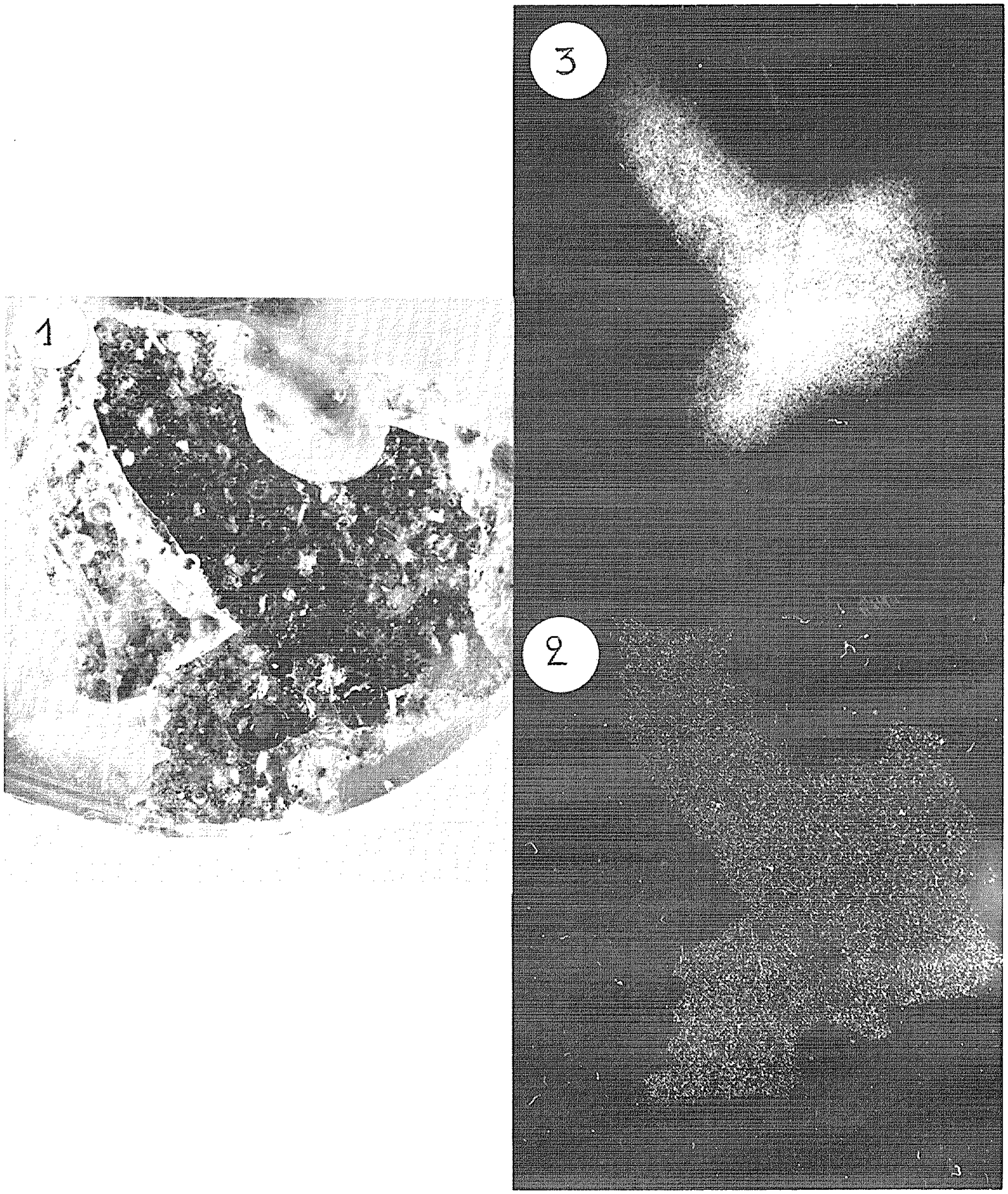


Fig. 7

1 - Photo of the sample
2 - α autoradiography
3 - β-γ autoradiography

Differences between β and γ radioactivity releases large quantities of gas. The melting of the lime resulting from the decomposition of marble showed that the temperature had been raised up to 3000°K, or more. As from granite samples, we obtained green melted rocks with lots of bubbles, quite like the samples taken out from nuclear burst.

We took a certain number of β - γ as well as α autoradiography snaps of the piece of melted rock from a nuclear explosion. We compared them with each other and them with a snap of the sample.

We noticed :

- some parts of the melted rock included no radioactivity.
- In the radioactive parts, α activity was quite homogeneous, while β - γ was not.
- The strongest β - γ activity parts corresponded to the parts of the melted rock scattered with bubbles. From the fact that the bubble lining is more radioactive than the melted product rock, we suppose that the elements concentrated in bubble linings are isotopes with gaseous antecedents having periods compatible with their inclusion as bubbles in the melted rocks, and we hope to obtain more concrete results soon (figures 6 and 7).

We also studied dissolved or occluded gases in the same melted rock. Occluded gases represent only a small part of the gases contained in the rock.

But, on the other hand, large quantities of active gases, particularly tritium, seem to be contained in those melted rocks. We think it may be interesting, particularly in case of an industrial application, and we mean to pursue these studies with other kinds of rocks.