

Applications of nuclear physics in archaeology

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1. Introduction

The use of low energy particle accelerators for fundamental nuclear physics is of decreasing interest since 1960-1970, but of high interest for analytical purposes in various fields outside physics. Their use in Archaeology was growing and the Museum of Louvre has been equipped for that purpose at the end of the 80's. LARN laboratory was very active in that field since the late 70's and we report here only a few applications on the study of "Objects of Eternity": a gold jewellery artefact of the Ancient World and a confident model for the construction of pyramids of Egypt.

2. Useful interactions for analytical purposes.

The main interesting interactions giving rise to signals of atomic or nuclear origin are schematically illustrated in table 1. References to fundamental works on analytical applications may be found in previous works (1-4).

Table 1. Useful interactions of ion beams in Archaeology.

PIXE (Particle Induced X-ray Emission): atomic reaction suitable to analyse medium and heavy elements (1).
PIGE (Particle Induced g-ray Emission), nuclear reaction suitable to analyse light elements (2).
RBS (Rutherford Backscattering Spectroscopy): elastic scattering on the atomic nucleus suitable to give depth information on medium and heavy elements, but less convenient for light elements (3).
NRA (Nuclear Reaction Analysis): true nuclear interaction inducing the transmutation of light elements and suitable to determine their depth profile (4).

3. Ancient gold artefact with various solders.

The high level of workmanship of ancient goldsmiths can hardly be equalled even today. Among the items of Iranian and Syrian jewellery that we have studied, the wonderful Achemenide pendant of the 4th century B.C. which belongs to the Department of Iranian Antiquities of the Musée du Louvre, clearly illustrates the skill of ancient Iranian goldsmiths.(5). The pendant is shown in Figure 1. The total width of the disk decorated in "repoussé" is 5.5 cm. The surrounding ornaments were made with 28 identical motifs. Each of them includes a hollow cylinder and two hemispherical caps. Regions of solders are clearly visible. These regions were non-destructively analysed for composition, and the results are

summarised in figure 2. All measurements have been performed with proton-induced X-ray emission (PIXE) in a microprobe assembly. A PIXE microprobe facility has been used to irradiate small regions (15 μm wide) of the sample put in a vacuum chamber. The pendant was fixed on an X-Y frame and moved in the proton beam by stepping motors with a reproducibility better than 2.5 microns. Four joins were made from regions A to D of Figure 2, over a distance extending less than 5 mm. The elemental composition at solders (impacts in regions A, B, C and D) shows that three different joining procedures were performed. At site C, the gold content is significantly greater than in the neighbouring regions and indicates that the join was made by welding without adding any external material: the end of the cylinder and the bottom of the first hemispherical cap were simultaneously heated to an early stage of fusion. In this process, metals like copper and silver were selectively eliminated, mainly by oxidation, so that the gold content was enhanced. The temperature was around 1050°C.

At site D, an increase in copper concentration was observed simultaneously with a proportional decrease in gold and silver concentrations. The two caps, previously well-fitted were joined by the process known as solid-state diffusion bonding with copper salts in a reducing atmosphere. The process took place at about 890°C. This relatively low temperature makes the joining at site D without de-soldering the join at site C, only 1.5 mm distant. This joining method was widely used by the Etruscans in their famous granulation work.

An increase in copper and silver and a decrease in gold were simultaneously observed at sites A and B, a result indicating that at both sites a brazing alloy has been used. The lower concentration of gold in region A indicated that the temperature of fusion of the alloy used at that site was lower than that used at site B (possibly around 820 and 860°C respectively). These temperatures are close to 890°C: the soldering temperature at site D. However, the latter join did not desolder. The process of diffusion bonding produces a join that cannot be de-soldered. Reheating after joining results in further copper diffusion. That diffusion induces a local decrease of the copper content with a consequent increase in the local melting temperature. The high degree of workmanship displayed in a piece as complex as the Achemenide pendant shows that, 2500 years ago, the goldsmith recognized and used different temperatures for various joining processes.

This process of soldering was easily reproduced in our laboratory. Natural malachite was finely powdered and mixed with some fat. The mixture was spread on a sheet of pure gold which was then covered with another gold piece. The whole assembly was heated in order to induce the reduction of malachite with carbon from the burned fat and to allow copper to diffuse in both parts of the system to ensure the bonding. A modern improvement of this process of bonding has been experimented using the formation of a low melting point gold binary alloy: the eutectic bonding with Au-Si (6).

4. From Etruscan solid state bonding to modern eutectic bonding.

Silicon alloyed with gold may form a eutectic alloy which can be used as a low melting point solder. We have experimented with a new process of gold soldering involving two steps: (a) the formation of a microscopic gold-silicon eutectic alloy during silicon diffusion through polycrystalline gold foils along their grain boundaries and the re-diffusion of a part of this eutectic alloy to form the joining with a second pure gold element. Both steps are performed at a low temperature (below 400°C).

Particle-induced-X-ray emission (PIXE) and nuclear reactions (NR) induced by a deuteron microbeam have been used to characterize the gold-silicon alloy formed by the diffusion of silicon into gold foils.

Pure polycrystalline gold foils are rolled at room temperature down to thicknesses ranging from 10 to 20 μm . A film of silicon is then deposited through a mask on a part of these foils

with an electron gun. As the adherence of silicon on gold is extremely difficult at room temperature, gold foils are maintained at 400°C during the silicon deposition. This temperature is slightly higher than the eutectic temperature (363°C) so that any Au-Si mixture at 400°C always contains a liquid phase. This liquid rapidly diffuses in the gold foils, but only along well-defined paths: the gold grain boundaries.

Nuclear reaction induced by deuterons giving rise to proton emission is used to measure the 3D distribution of silicon in gold. Deuterons can scarcely hit or graze a light nucleus (like Si) and be stripped, leading to the capture of the neutron and the emission of the residual proton (stripping reactions). This nuclear reaction on silicon is highly exoenergetic ($Q = 6.249 \text{ MeV}$) due to greater binding energy of the exchanged neutron with the silicon nucleus by comparison with that into the deuteron. This nuclear reaction involving the neutron capture to leave the final residual nucleus in its fundamental state has its maximum intensity if the proton emission is achieved in the forward direction. We have then chosen to detect the emitted protons in the forward direction. A 25 μm thick absorber of pure gold is inserted between the sample to be studied and the detector, in order to completely stop the incident deuteron beam. The choice of a material containing only a heavy element as absorber prevents any emission of additional protons. On account of the low cross section of nuclear reactions induced by low intensity microbeam, the sensitivity for silicon determination in volumes of 30 μm^3 lies around 0.5%. The experimental procedure is illustrated in figure 3. The results of figure 4 have been obtained on a gold foil of 12 μm thick irradiated by 2.8 MeV deuterons incident on the back side of the foil prepared by silicon diffusion (the side opposite that on which silicon has been deposited). The first map (on the left) corresponds to the front surface of the foil (that on which silicon has been deposited). The last one corresponds to the silicon concentration at the rear, 12 μm below. (7).

With the same experimental arrangement for the analysis and the same experimental procedure (analysis followed by the reproduction of the process in our laboratory) we may propose a confident mode of construction of several Egyptian pyramids.

5. The composition of the pyramid of Kheops .

The Cheops' pyramid, with a volume of $2.7 \cdot 10^6 \text{ m}^3$, was completed over a period of 20 to 25 years. One can then estimate the average daily cadence at 300 to 400 blocks having all an average volume of 1 m^3 (i.e. 750 to 1000 tons per day). This represents one block put at the right place every two minutes. To achieve this goal, 1 m^2 of hewn face would have been ready every 20 seconds! What a performance with tools made of stone or soft copper! Hoisting huge blocks of more than two tons with rudimentary means (wheels and pulleys did not exist at that time) is evidently an impossible task. As several dozens of those monuments have been constructed on the left bank of the Nile by the Pharaohs of the first dynasties, we cannot imagine the average time of construction of each pyramid to be much longer. When looking carefully at the surface of the blocks of the pyramid of Kheops (those visible today and therefore those underlying the casing blocks, which totally disappeared), one clearly sees irregularity in the shape, but a remarkable close fit of adjacent faces. It would be surprising that these blocks could have been so badly cut but so perfectly joined. This admirable close fit would have been easier to achieve if the blocks had been hewn with perfect rectangular shapes! Furthermore, this care in this optimal juxtaposition was useless because these blocks (visible today) were originally hidden under the casing. We can also see that blocks appear to be more porous in their top part than in their bottom part. This porous feature on the top of the blocks cannot be explained by some climatic erosion of natural limestone but could be understood if we propose a construction similar to our modern concrete.

Narrow channels, with a section of 20 cm x 20 cm, starting from the Queen's chamber and investigated by R.Gantenblink's robot (8) clearly indicate that they were not carved . There is no gap between the two lateral sides (walls) and the ceiling of this conduit. On a TV show of the Gantenblink's expedition, one could see that no protrude (convex) defect appears in the walls and in the ceiling of this narrow tunnel. Irregularities are only of hollow (concave) shapes. A carving procedure would have given convex and concave irregularities in equal proportions. When thinking about a moulding procedure, the apparent cavities could be understood by some loss of material during the de-moulding.

Many other arguments including (a) the chaotic organisation of nummulites in the blocks, with respect to parallel alignment of shells in natural stones, (b) the high water content (about 13%) of the whole pyramid measured by the transmission of electromagnetic waves, (c) traces of mortar mostly at the base of the blocks, play in favour of another way of construction: not natural hewn and hoisted stones but the agglomeration of natural limestone using a binder ... which contains natron, alumino-silicates and certainly water. All ingredients have been transported in small quantities, dropped in moulds installed progressively onto or on the side of blocks which were previously moulded.

In the 70's, J.Davidovits (9) proposed that the great pyramids were made of a kind of concrete whose basic binding element was natron: a sodium carbonate. Natron was indeed widely extracted from a region of the North of Egypt, on the left bank of the Nile, very close to the site of Giza. The binder is obtained by some chemical reaction giving rise to a geopolymer (name given by Davidovits to a class of X-polysialates, X being an alkaline nucleus, in particular sodium). Natron, lime and water form caustic soda, which reacts with aluminous limestone to yield the basic geopolymer. A mineral ore containing arsenic (scorodite or olivenite) is added to produce sodium arsenate acting as an activating ingredient that could have been used in various concentrations to control the speed of the hydraulic setting. The invention is attributed to Imhotep, the architect of the pyramid of the Pharaoh Djoser.

In addition to the analyses carried out by J. Davidovits with X-ray fluorescence (9) and X-ray diffraction, which showed that the blocks mainly consisted of limestone (85 to 92%), we have also performed investigations on a little number of samples from the Kheops' pyramid: elemental analysis was performed by ion beam analyses, PIXE and PIGE and structural characterization by NMR-Spectroscopy. (10 - 11)

By using the PIGE-PIXE techniques (Proton Induced Gamma-ray Emission or X-ray Emission) we have determined the elemental content of small fragments. The light elements F, Na, Mg, Al, Si were quantitatively determined by using PIGE , and Ca, Fe and other trace elements by using PIXE (11). A small sample from the Kheop's pyramid (figure 5) is made of a central compact structure containing mainly limestone with traces of other elements The outer part contains a large amount of F, Na, Mg, Al, Si, indicating that a material to aggregate the limestone has been used . The ratios of concentrations of F, Na, Mg, Al and Si in the coating relative to the bulk are given in Table 2.

Table 2
Intensity ratios (coating/bulk)

F	7.5
Na	8.5
Mg	12
Al	2
Si	21

Except for Al, those ratios are much more greater than one, indicating a complete different structure. The high concentration of sodium is certainly due to the use of **natron** for the binder. Furthermore, the significant signal of As in the coating may be attributed to some additional ore which could be scorodite as more extensively discussed below. In addition to PIGE and PIXE measurements which allow us to have insight to the elemental composition, the Nuclear Magnetic Resonance Spectroscopy (NMR) of Al²⁷ and Si²⁹ enables us to determine the type of synthetic medium (basic pH) and to differentiate a natural environment (neutral pH) from an artificial one. We have then fabricated the binder based on the geopolymer formula of J.Davidovits (9). The NMR-Spectra of Al and Si on this modern synthetic material shows typical resonances assigned to Si [Si(OSi)₄] and Al (tetrahedral) in this synthesized material which is highly chemically basic (pH around 10). The NMR spectra of several samples of Kheops pyramid indicate that the tetrahedral Al content is 10-15% of that obtained for the pure synthetic mixture reproduced in our laboratory and which exhibits a very fine adherence with small gravels. This value of 10-15% of the NMR signals is in direct relation with the amount of geopolymeric binder and, consequently, also related to the original water content of the blocks. Si-NMR leads to the same conclusion.

6. Manpower for the construction of the pyramid.

The moulds could have been made of grooved small boards fitting one into the other in the way of modern wooden floors. The fixation by two crossbars of the first and of the last small board of the mould (and only these two boards), as represented in figure 6 , allows the intermediate small planks to slide in a movement going from the top downwards to prevent any leakage in the region of contact with the block of the inferior step. These small boards have probably the width of an Egyptian palm (1/7 of cubit, about 7.5 cm). The construction of the moulds with a definite number of small boards explains the modular width reported by Davidovits for the blocks of -'s pyramid. Davidovits mentioned this modular dimension (an integer of a reference board width) for the blocks of Khephren and not those of Kheops because blocks of Khephren have a more regular dimension. This second pyramid of Giza would have profited from the experience acquired during the construction of the first pyramid at Giza: the Kheops pyramid. Those blocks of Khephren have then a higher quality in their finish.

The irregularity noticed on the blocks in the Kheops pyramid would be due to possible "accidents" during the removing of the planks of the mould after the solidification of the liquid binder. Any loss of building material occurring during de-moulding would have been corrected by a partial covering of the de-moulded block with the next mould. With this method, the hardening of the new block provided a close fit with the precedent, even though this last one had lost its surface regularity during the de-moulding.

A system of two boards constituted of a variable number of one palm wide small planks was sufficient for the constitution of a mould: the lower blocks, the side blocks and the leaned blocks provide the other four faces that insure the tightness of the volume in which the mixture is poured. This procedure explains the modularity noticed for the widths of all the blocks in Khephren's pyramid and also the close fit between the irregular blocks in the pyramids of Giza, even the one of Kheops. The height of a single block depends on the height of the shortest plank used in for mould. The modular width of the blocks of the Khephren's pyramid depends on the number of small planks constituting the board.

The solid ingredients brought in bow nets are poured into the mould that has been made waterproof by the application of a mortar. This mortar is applied to the internal base of the mould. The mortar, already visible today, is mainly present at the base of the blocks. As for moulding a block, liquid ingredients are first poured, waterproofing is necessary at the bottom

of the mould. The solid ingredients introduced in this liquid are limestone aggregates brought from the immediate neighbourhood of the construction site.

Instead of having teams pulling blocks weighing several tons, the workers transported the ingredients by small loads (of 25 to 50 kg each) that do not require any wide ramp (that anyway would badly support the extensive heavy carriages during twenty years at a rate of 300 to 400 blocks of one cube meter per day). Workers could also save their energy by taking the load and bringing it upwards, passing it from man to man, without having the need to climb the pyramid with their load. Any transportation of a small charge would give rise to a maximum efficiency. In the case of heavy hewn blocks, the efficiency would have been extremely lower. The fabrication of moulds (static structure of 0,5 to 2m²) would require at least 100 times less wood (wood is indeed rare in Egypt) than the fabrication of sledges: less quantity of wood for a (static) mould than for a (moving) sledge and shorter time of use for moulds (one day) than for sledges (several days ... or even several weeks).

Assuming that the aggregates were extracted from quarries close to the building site and distant by at most 3 km from Kheops pyramid (as may still be seen in the surroundings today), we have estimated the cadence of the construction and the number of workers occupied on the site in the following way:

- a) 6 men are bringing one cube meter of ingredients per day, from the nearby quarries to the bottom of the pyramid (400 kg per man per day);
- b) for the lifting of ingredients from the plateau level to the desired step, we estimate that the cadence would represent 1 m³ of material to be lifted by each worker during one hour from one step to the next one above. Bearing in mind that the height of the steps varies from 100 cm to 50 cm when one passes gradually from the base to the top of the pyramid, there is a progressive decrease in the yield from the bottom to the top. This performance is completely similar to the one accomplished today if we have in mind that modern associated mason have to transport 7000 kg of bricks, sand or cement up to a 3 m floor per day.
- c) two workers are dedicated to the maintenance of each mould: survey of waterproof of the plank assembly of the boards, kneading during the pouring of ingredients.

The distribution of the manpower is illustrated in figure 7.

7. Ancient descriptions and illustrations.

In a well-documented report on the construction of the pyramids, Herodotus (Vth century B.C.) reports:

*"This pyramid was made, as I am going to say, in terraces that some name steps, and others small altars. When the base had been built, the rest of stones was raised by means of machines fabricated with **short** wooden pieces; the force of a machine acted from the ground until the level of the first tiers at first; once transported there, the stone was put on a second machine, which was fixed there. From there it was risen on the second tier, and on a third machine. There were as many rows of terraces, as there were machines. It is possible however that there was only a single portable machine: in this case, it was taken up from tier to tier, having brought up there the stone. It is indeed necessary to report these two processes as they were said to me. **The summit of the pyramid was finished before the rest**; and afterwards they completed the parts in the following tier, and one ended by the lowest, by the one that touches the ground. It was written in Egyptian characters, on the pyramid, how much was spent **for garlic, onions and parsley** for the workers. As I well remember it, the text (that the interpreter explained to me) means that the sum amounts to sixteen hundred talents of silver (58 tons of silver!). If these things cost so much, what did they spend in metallic tools, in **foods** and in clothes, since they took the time that I have mentioned to build this edifice*

*without even counting, in my opinion the time for quarrying the stones, their transportation and the construction of **underground** excavations?*

The description supports the following explanations:

- a) **short** wooden pieces cannot be long beam of levers necessary for the lifting, but could be the planks of the mould,
- b) the completion of the pyramid began with the installation of the **facing blocks at the summit** first. In order to be sure he would be well understood Herodotus insists in the next sentence with the following statement: the builders then continue on the following floor, downwards, to end with the one that touches the ground. These facing blocks are still present today at the top of Khephren's pyramid and they hold firmly even after more than four millennia. They have then evidently been put in place at the top first,
- c) **garlic, onion and parsley** cannot be understood as food supply for the workers. A comment on the cost of these ingredients provides us with a hint on the nature of the materials used and not of food. In the sentence which closely follows this statement, Herodotus argues about the supplementary cost for food, tools and clothes. Garlic and onion have a direct link with ingredients occurring in the manufacture of blocks. According to J.Davidovits (9) two of the three ingredients appear in the text of the Famine Stele (9) describing the "Revelation of Imhotep : "hedsh" is a disaggregated stone smelling like onion and "tem-ikr" is a mineral containing arsenic which has the characteristic smell of garlic and garlic stone could be scorodite as observed by the PIXE results on the coating of the small aggregate of figure 5. The famine stele relates to the description of various skills of Imhotep, the architect of the Pharaoh Djerer, who describes the procedure to make some kind of concrete.
- d) **underground excavations** cannot refer to open air quarries but could be associated with excavations in mines located on the left bank of the Nile in the northern region close to Giza to extract natron. The name of this region is already today: Ouadi Natrum.

This method of construction based on a moulding technique is explicitly described on a painting of the tomb of Rekhmire dating from the XIInd dynasty. Various operations are illustrated on it: loading of ingredients in bow nets (on the left), transport and pouring upright into a mould (in the top centre); one plank of the mould is maintained vertically by a civil servant (in the bottom centre). Contrarily to some modern transcription, the original painting does not show any modular structure in this large block that some scholar had interpreted as an assembly of bricks.

8. Proposals for future investigations on pyramids.

To put an end to the debate involving partisans and opponents of the construction method based on agglomeration, we suggest to improve the physico-chemical analyses by increasing the study of samples, but in addition, by carrying out several simple experiments such as:

- a) critical study of the slope for each of the three Giza's pyramids, compared with the lower slope of an anterior pyramid like the one of Snefrou. We think that the slope is connected to the type of building material used for the construction. Civil engineers know the relation existing between slope and ingredients that constitute a heap of material. The longevity of the pyramids is, in our opinion, rather linked to these technical data than on computation of "magic" numbers;

- b) dating of straw in mortars taken precisely at the bottom of the blocks. The determination of the C^{14} content by accelerator mass-spectrometry would require a minimum sampling;
- c) removal of some blocks for the observation of an internal face. The presently visible faces underwent climatic erosion which eliminated any imprint of board or ... carving marks;
- d) study of the well fitted faces of the blocks should reveal whether their faces adhere or not.
- e) sampling of small size fragments and their elementary analysis in terms of elements of light atomic weight, using modern techniques and their quantitative comparison with the contents of these same elements in the limestone found in the neighbourhood of the site;
- f) further study of texts, especially in hieroglyphs, and their interpretation by an interdisciplinary team: philologists, historians, archaeologists, with the help of physicists and chemists to comment on scientific interpretation;
- g) reconstruction of blocks of a size similar to those of the pyramids by moulding and carving techniques under the control of independent experts.

Conclusions

Since the end of use of small particle accelerators for fundamental research on mechanisms of nuclear reactions several laboratories continue to use them for fundamental researches in other fields. Their use in Archaeology brings some new insight in fundamental problems like provenance, ancient technologies in complementarity with conventional analytical techniques used in laboratories of museums. Pertinent results may be obtained on materials which are not altered by wearing. We have selected to give here only two examples : one on the identification of several types of solders on a gold jewellery artefact and the second one on a new proposal to explain the way the Egyptian pyramids have been build.

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Figure 1. The gold Achaemenide pendant (Musée du Louvre) : 6.4 cm wide.

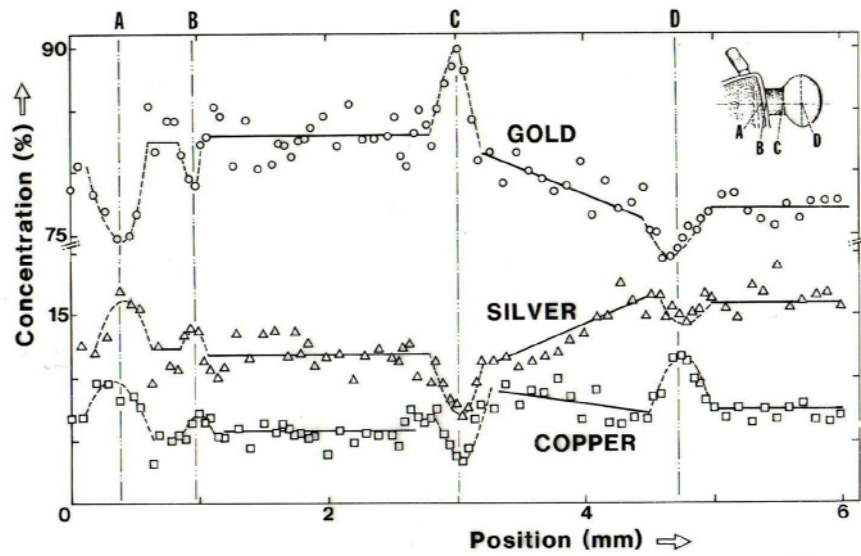


Figure 2. Concentration of the main elements measured with a PIXE-microprobe.

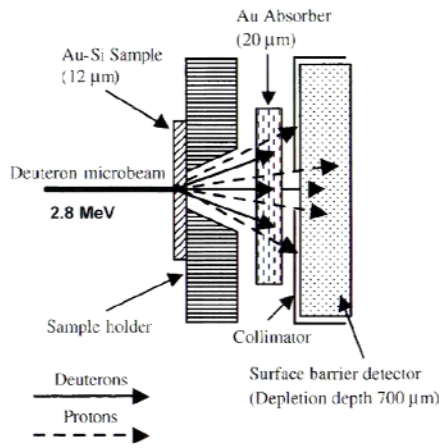


Figure 3. Experimental arrangement for the measurement of Si depth profile

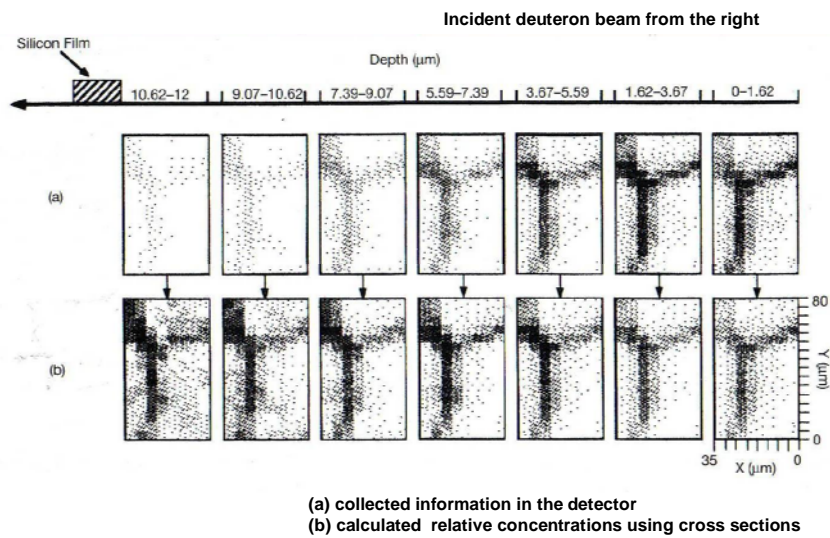
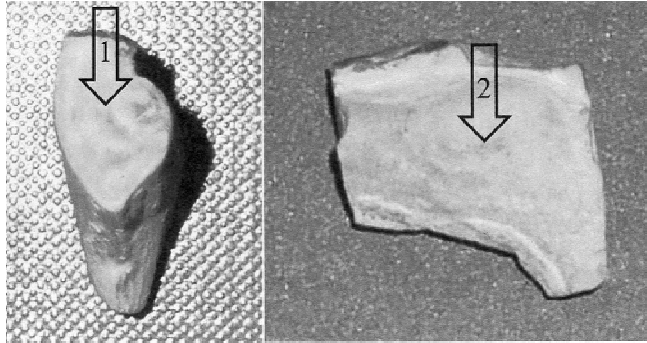


Figure 4. Silicon concentration in the grain boundaries.



Regions of analysis : (1) in the bulk, (2) in the coating

Figure 5. Fragment of the pyramid of Kheops (length : 18 mm).

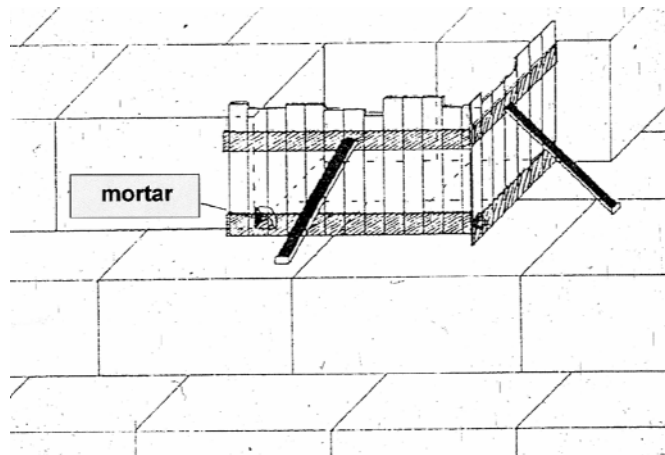


Figure 6. The model of mould made with small planks one Egyptian palm (7.5 cm) wide.

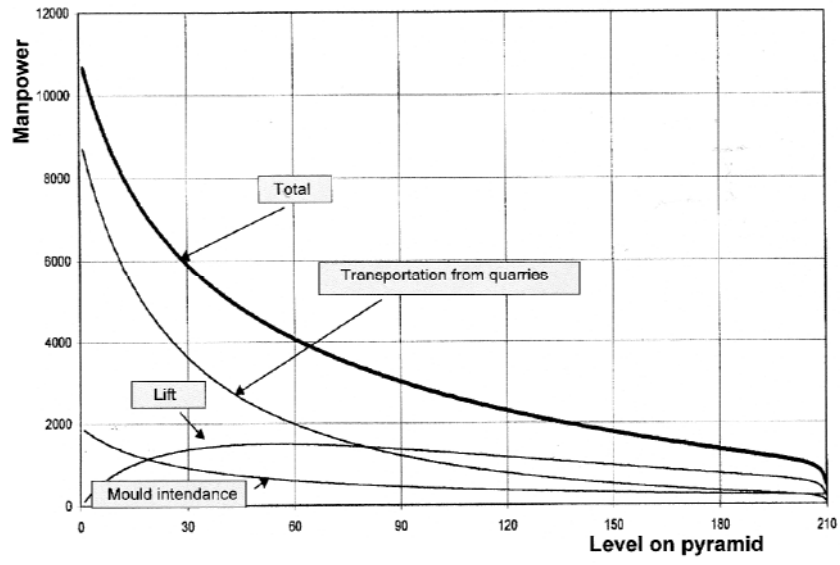


Figure 7. Manpower for the construction of the Kheops pyramid.