*Francesco Barbesino & **Mario Moroni

# Effects of neutron irradiation on linen fibres and consequences for a radiocarbon dating

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* Graduate in Industrial Chemistry • fra.bar@libero.it
** Industrial Expert - Member of the International Center for Sindonology of Turin • liliana.tagliaferri@alice.it

The results of the analyses performed on the linen sample of the Shroud of Turin, already since they were published in 1988, have raised many doubts because they contradicted the results previously obtained in a variety of different disciplines.
Many hypotheses have been formulated to explain the presence of the high $^{14}C$ content which produced the medieval dating. [1]

On our part, we formulated and took into consideration two assumptions :

- an increase of $^{14}C$ caused by the fire of Chambery occurred in 1532
- an increase of $^{14}C$ produced by a neutron irradiation.

In order to obtain results which were comparable with each other, we have chosen:

- to obtain the analyses by a single laboratory: the IsoTrace Radiocarbon Laboratory of the University of Toronto (Canada), equipped with an accelerator mass spectrometer (AMS)
- to carry out the tests with the same old linen: the Lyma mummy linen coming from the "Museum of tissues" of Lyon.

## 1. Dating of the Lyma mummy linen “as received”

We had received the piece of Lyma mummy linen from prof. J. B. Rinaudo. This linen had already been irradiated at the “*Centre de datation par le radiocarboni - Université-Claude Bernard Lyon 1*”, but we preferred to repeat the analysis not knowing the criterion used to evaluate the errors in that laboratory.
Later, for the reasons we’ll explain herein after, a second analysis was performed, preceded by a very strong cleaning pretreatment The results are presented in Table I[2].

| Sample | Apparent age (years BP) | Yield % | Probability % | Calibrated age | Confidence interval 68,3 % | Confidence interval 95,5 % |
|---|---|---|---|---|---|---|
| TO-Ly 6509 | 2110 ± 60 | 40 – 80* | 100 | 115 BC | 195 BC - 40 BC | 250 BC - 20 AD |
| TO-13583 | 2020 ± 50 | 12,6 | 100 | 40 BC | 55 BC - 30 AD | 170 BC - 80 AD |
| | | | 100 | 10 BC | 55 BC - 30 AD | 170 BC - 80 AD |
| | | | 100 | 0 BC | 55 BC - 30 AD | 170 BC - 80 AD |

Table I

1 Marino J. G., Benford M. S.,: “*Evidence for the Skewing of the $^{14}C$ Dating of The Shroud of Turin due to Repairs*”, www.shroud.com/pdfs/marben.pdf
Rogers R.: “*Studies on the radiocarbon sample from the Shroud of Turin*”, Thermochimica Acta, Vol. 425, Issues 1-2 , 20, Jan. 2005, pp. 189-194.
Alonso M.: Shroud Science Group on Yahoo discussion.

2 For the meaning of individual columns and calibration data of Tables I, II, III and V, see Appendix.

* 40 - 80 . = Maximum interval of the yield % in the conventional cleaning pretreatments of samples (see R.L.Otlet – J.Evin: “*The present state of radiocarbon dating*” in “The Turin Shroud – Past, present and future.”, International Scientific Symposium, Turin 2-5 March 2000, Sindon - Effatà ed. (p. 455-478, p.470.)

## 2. Simulation of the fire occurred in Chambery

The sample was subjected to heat treatment at a temperature of 200° C for 90 minutes in a mixture of $CO/CO_2$ [3]
The radiocarbon analysis gave the following results (Table II):

| Sample | Apparent age (years BP) | Yield % | Probability % | Calibrated age | Confidence interval 68,3 % | Confidence interval 95,5 % |
|---|---|---|---|---|---|---|
| TO-6897 | $1810 \pm 60$ | 63 | 100 | 235 AD | 135 AD - 260 AD | 75 AD - 390 AD |

Table II

Also another sample of the Lyma mummy, which was subjected to a thermal simulation with different modalities (see note 3) and analyzed by the AMS Laboratory of Groningen on behalf of the Beta-Analitics of Miami, turned out to have a $^{14}C$ content lower than that above mentioned.
Therefore it does not seem possible to attribute to the heating the large increase in the $^{14}C$ content found on the samples of the Shroud.

## 3. Neutron irradiation

T.J. Phillips in 1989 had already advanced the hypothesis that a neutron flux could be the cause of the strong increase in $^{14}C$ [4].
Subsequently, our team started a fruitful collaboration with Professor Jean Baptiste Rinaudo of the Faculty of Medicine of Montpellier. He presented a model in order to explain both the formation of the Shroud and the increase in $^{14}C$.
According to this model, a sudden disintegration of the deuterium nuclei, which are present on the surface of the body, may have originated protons and neutrons. The first ones could be the origin of the image, the second ones could be the cause of the increase in $^{14}C$ [5]. On the basis of this hypothesis, we estimated the proton flux necessary to produce the coloring of the Shroud and, from this value, we calculated the corresponding neutron flux.
(see Appendix).

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[3] The modalities of both simulations, as indicated respectively by Koutznetsov and Moroni, are described in the report : M.Moroni-F.Barbesino-M.Bettinelli. "*Verification of an hypothesis of radiocarbon rejuvenation*, III International Congress of Studies on the Shroud, Turin 5-7 June 1998.

[4] Phillips, T.J., "*Shroud Irradiated With Neutrons*?," Nature, Vol. 337, 16 February, 1989, p.594; http://creationevolutiondesign.blogspot.com/2007/05/bogus-shroud-of-turin-4.html

[5] Rinaudo J. B.: "*Nouveau mécanisme de formation de l'image sur le Linceul de Turin, ayant pu entraîner une fausse radiodatation médièvale*", L'Identification Scientifique de l'Homme du Linceul, Jésus de Nazareth, Actes du Symposium Scientifique International, Rome 1993, F.-X. De Guibert, Paris 1995, pp. 293-299.
Rinaudo J. B., "*Image formation on the Shroud of Turin explained by a protonic model affecting radiocarbon dating*" III International Congress of Studies on the Shroud, Turin 5-7 June 1998.

Prof. Rinaudo gave us a piece of the Lyma mummy linen irradiated with a neutron flux of $1.13 \cdot 10^{13}$ n/cm$^2$. From this piece we obtained three samples: one was directly radiodated and the two others were radiodated after the heat treatments simulating the Chambery fire previously performed. (see note 3).
These analyses' results are shown in Table III.

| Sample | Apparent age (years BP) | Yield % | Probability % | Calibrated age | Confidence interval 68,3 % | Confidence interval 95,5 % |
|---|---|---|---|---|---|---|
| TO-5305 | $1750 \pm 50$ | 10,3 | 100 | 260 AD | 240 AD - 385 AD | 145 AD - 215 AD |
| | | | 100 | 290 AD | 240 AD - 385 AD | 145 AD - 215 AD |
| | | | 100 | 325 AD | 240 AD - 385 AD | 145 AD - 215 AD |
| TO-6898 | $990 \pm 60$ | 79 | 100 | 1025 AD | 1005 A D - 1050 AD | 965 AD – 1210 AD |
| TO-6420 | $720 \pm 50$ | 51,9 | 100 | 1290 AD | 1275 AD - 1300 AD | 1230 AD – 1315 AD |

Table III

From these results, at first we arrived at the conclusion that the heat treatment carried out on the irradiated samples was able to produce a significant increase in the $^{14}C$ content. However, after reviewing the results, we noticed that:

- The $^{14}C$ content in the irradiated sample (TO-5305) is much lower than we could expect after a neutron irradiation.

- The yield of this sample was much lower than the percentage obtained from the normal cleaning pretreatments made before the analyses performed with AMS equipment.

Therefore, on the advice of Dr. R.P. Beukens, we sent to the IsoTrace Laboratory a piece of the irradiated Lyma mummy linen, with which they started a series of analyses by applying increasingly severe cleaning pretreatments. The results are summarized in Table IV.

| Pretreatment | Apparent age (years BP) | Yield % |
|---|---|---|
| no pretreatment | $-520 \pm 50$ | 100 |
| washed | $590 \pm 50$ | 85 |
| AAA* | $940 \pm 50$ | 65 |
| cellulose** | $1040 \pm 50$ | 17 |
| AAA + cellulose | $1750 \pm 50$ | 10 |

Table IV

* AAA: strong acid and alkali extraction followed by an acid wash .
** cellulose:. hot acid/chlorite bleach extraction followed by alkali extraction and an acid wash.

We can therefore state that, in irradiated samples, the more severe is the pre-cleaning and the yield decreases, the more the $^{14}C$ content decreases. This result was confirmed by the analyses of two new Lyma mummy samples, also provided by Prof. Rinaudo, which were irradiated with $2.59 \cdot 10^{13}$ n/cm$^2$ : one sample was subjected to a very strong cleaning pretreatment (yield 7.8%), the other one to a conventional cleaning.

| Sample | Apparent age (years BP) | Yield % | Probability % | Calibrated age | Confidence interval 68,3 % | Confidence interval 95,5 % |
|---|---|---|---|---|---|---|
| TO-12553 | $1340 \pm 60$ | 7,8 | 100 | 660 AD | 645 AD - 690 AD | 600 AD -775 AD |
| TO-12553-2 | $260 \pm 50$ | 55,9 | 100 | 1645AD | 1630 AD -1665AD | 1605 AD - 1680 AD |

Table V

**Conclusions**

The most evident result of our research is the increase of the $^{14}C$ content after neutron irradiation[6] and the impossibility, even with the most severe pretreatments, to reach the "historic" age (calibrated on dendro-chronological curves) of the non irradiated sample (TO-13583 - 40/10/0 BC).
**Therefore, if an irradiation had happened, the result would explain the anomalous data obtained in the analyses of the Shroud made in 1998.[7]**
It is evident that the $^{14}C$ content increases with the increase of the neutron flux. The sample TO-12553 (Table V), although subjected to a cleaning pretreament with a yield of 7.8%, shows a result of the $^{14}C$ content greater than the sample TO-5305 (Tab.III).

We can add some considerations:

- Every linen fibre is composed of a certain number of pure cellulose fibrils associated with complementary substances (hemicellulose, lignin, pectin) that form interfibre lamellae and cementing textiles which go with the cellulose up to the finished product. Their number varies depending on the conditions of production and processing. It is also probable that their distribution along the axis of fibres is not homogeneous. Although it is impossible to ascribe to the cellulosic chains the $^{14}C$ increase corresponding to 1000 years and more, this could be possible with the complementary substances.
  If the amount of $^{14}C$ which combines with the complementary substances is high, it is impossible to obtain from the analyses the "historic" (or balanced) age that would be obtainable from pure cellulose; in fact, if we push the cleaning pretreatment up to the limit, also the cellulose will disappear with the additional substances.

[6] In the current state of research, we are not able to indicate what mechanisms have produced the increase occurred in the $^{14}C$ content. Several hypotheses have been made: adsorption or chemo-adsorption of the $^{14}C$ produced by the collision of neutrons against the nitrogen atoms present in the environment, the creation of free radicals and many others.
[7] Professor Robert Hedges (consulted on the pretreatment of a second analysis performed after several years on the Shroud samples delivered to the Laboratory of Oxford - this analysis confirmed the previous results) said that, as he recalled, the yield ranged between 70 and 75%.
(fax addressed to M. Moroni on 30/01/08).

- Also on not-irradiated antique linens a severe cleaning pretreatment may be useful. This is detectable from samples in Table I. In fact, it is known that the possibility of interaction between fabric and external agents also depends on the degree of order of the cellulose chains closely packed with each other by means of hydrogen bridges. In areas where such a compactness decreases (indicated as semi-crystalline areas) the action of external agents becomes more effective.
  In the past, we examined with X-rays diffraction spectra the degree of order of three substantially coeval antique linens: since their results were significantly different among them, the degradation had to depend on the historical events happened to each of them.[8]

## Appendix

Protons irradiation

It should be noted that Professor Rinaudo had previously established by calculation that if we consider the thickness of a body surface of 10 μm, this surface contains for each $cm^2$ an amount of deuterium enough to provide the required protons. Only the surface layers of the body were considered since the protons from the deeper layers would have suffered drastic reductions of energy while crossing the tissues overlying.
The flux was obtained by a particle accelerator type Van der Graaff installed at the "Centre d'Etudes Nucleaires Bordeaux-Gradignan". The linen used, recently manufactured, was "white" and was washed. The flux deemed necessary was of 2 $\mu C/cm^2$.
We obtained the confirmation that the colour of the linen irradiated with protons was similar to that of the Shroud by comparing the spectroreflectometric registration of the first linen with that obtained on the Shroud by Roger and Marion Gilbert in the campaign conducted by STURP in 1978 (see Gilbert R. Jr. MM and Gilbert: *"Ultraviolet-Visible Reflectance and Fluorescence Spectra of the Shroud of Turin*", Applied Optic 19 (1980), p.1930 - 1936.
A series of exploratory campaigns have confirmed some features expected on the basis of the original investigation. It was found that the image is three-dimensional, superficial and indelible and that the "singeing" does not produce furfurals (see M. Moroni - F. Barbesino: "*Different formation mechanisms of the bloodstains and the body image on the Shroud of Turin*" , The Dallas Third International Conference on the Shroud of Turin, Dallas, Texas, September 8-11, 2005.)

Neutron irradiation

Professor Rinaudo pointed out that in his hypothesis the neutrons, emitted as a result of the rupture of atoms of deuterium, are fast neutrons (1.1 MeV) which, slowed by the successive shocks, become thermal neutrons (0.0025 eV) capable of producing $^{14}C$.
The neutron irradiation of the samples was performed by the "Service du Reacteur Nuclear Universitarie" of the University L.Pasteur in Strasbourg (samples in Table I, II and III) and by the "Istitute Laue Longevin" of Grenoble (samples in Table V).

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[8] M.Bettinelli - E.Cartoni - M.Moroni - F.Barbesino*: Impiego di tecniche fisico-chimiche per lo studio dell'invecchiamento delle fibre di lino (*Use of physico-chemical techniques for the study of the linen fibres*)*, 2000 Worldwide Congress on the Shroud of Turin, Orvieto. 27-29 June 2000. www.sacrasindone.135.it/

<u>Meaning of items in Tables I, II, III and V</u>

Apparent age (or radiocarbon age) – Radiocarbon dating referred to the year 1950.

Yield (%) – final weight of the pretreated sample / initial weight x 100.

Probability % – 100 when the radiocarbon date intersects the dendro-chronological curve in one or more points.

Calibrated age – age obtained by the radiocarbon dating on the basis of the dendro-chronological curve.

Confidence interval 68,3 % e 95,5 % – the limits 1σ and 2σ of a normal distribution.

## Calibration

The results of Table I, II e III were calibrated with bidecal data set INTCAL93 from: M.Stuiver and P.J. Reimer, Radiocarbon 35 (1993) p. 235-230.

This data set uses the dendro-calibration results from:
M.Stuiver and G.W.Pearson: Radiocarbon 35 (1993), p.1-23.
G.W.Pearson and M.Stuiver: Radiocarbon 35 (1993), p.25-33.
T.W.Linick, A.Long, P.Damon, C.W. Ferguson: Radiocarbon 28 (1986), p. 943-953.
B. Kromer and B.Becker: Radiocarbon 35 (1993), p.125-135.

The results of Table IV were calibrated with INTCAL04 Terrestrial Radiocarbon Age Calibration, 0-26 cal kyr BP from: P.J.Reimer et al.: Radiocarbon 46#3 (2004), p.1029.

<u>Credits</u>

We thank Dr. Roelf P. Beukens of the IsoTrace Laboratory of the University of Toronto who was always generous with prompt critical comments and useful advises in a collaborative relationship that lasted almost twenty years.