FAST AND EPITHERMAL NEUTRON RADIOGRAPHY USING NEUTRON IRRADIATOR

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ABSTRACT

The neutron radiography technique (NR) with neutrons in the energy range fast to epithermal is a powerful tool used in non-destructive inspection of bulky objects of diverse materials, including those rich in hydrogen, oxygen, nitrogen and carbon. Thus, it can be used to identify, inclusions, voids and thickness differences in materials such as explosive artifacts and narcotics.

Aiming at using NR with fast and epithermal neutrons, an Irradiator was constructed composed by: a $^{241}$Am-Be source, with 5 Ci activity; a collimator with adjustable collimation rate, L/D; and a shielding device composed by plates of borated paraffin and iron.

The test specimens chosen were a Beam Purity Indicator (BPI) and an Indicator of Visual Resolution (IVR).

The neutron radiography images obtained had a resolution of 444.4 $\mu$m and 363.6 $\mu$m respectively when registered in: 1) the sheet of the nuclear track solid detector, CR-39 type, through $X(n,p)Y$ nuclear reaction; and 2) Kodak Industrex M radiographic film plate in close contact with a boron converter screen, both stored in a Kodak radiographic cassette.

1. INTRODUCTION

Fast Neutron Radiography – FNR is a technique that uses epithermal and fast neutrons in an energy range from 0.5 $eV$ to 20 $MeV$. The use of this technique brought progress to material inspection, because the penetration capability of neutrons with those energies in some materials is undeniable [1], allowing thicker samples to be inspected, because higher energy neutrons have to travel a longer path before being completely attenuated.

The detection of fast neutrons and consequently obtaining neutron radiographies is not an easy task to be accomplished. Fast neutron radiography provides images of lower quality than thermal neutron radiography. One of the main difficulties is to convert fast neutrons into radiation capable of sensitizing a radiographic film. However, a process that is being widely used increasing the number of FNR applications uses solid state nuclear track detectors – SSNTD [1].
2. THEORETICAL FOUNDATIONS

2.1 – Neutron Radiography System

To implement a Neutron Radiography system with fast/epithermal neutrons for non-destructive inspection of bulky objects, a device called Neutron Irradiator, which allowed obtaining high quality Neutron Radiography images, was constructed. The Neutron Irradiator is composed by: source, shielding and collimator, the dimensions of which were optimized by computer simulation with the Monte Carlo N-Particle- MCNP code [2], which allowed designing and constructing them. The merit parameters estimated for the construction of those complementary devices were the maximization of the fast neutron flux at the image plane and the smallest shielding thickness of the Irradiator to keep gamma and neutron radiations at dose allowable levels, according to CNEN NN-3.01 technical Regulation [3].

The source used was $^{241}$Am-Be [4], manufactured with a compact mixture of americium oxide and beryllium metal powder [5], double encapsulated with AISI.316 stainless steel walls. The neutron emission rate of a $^{241}$Am-Be source with activity 185 GBq (5 Ci) is of the order of $10^7$ n/s and its half-life 433 years.

The merit parameter to be considered on the construction of the collimator is the intensity of the neutron beam at the image plane. The collimator was designed with two removable aluminium modules, one divergent and the other parallel, with cadmium internal coating, thus allowing changing the L/D rate. The first module, the dimensions of which are shown in figure 1.a, was designed aiming at obtaining Neutron Radiographies that require great neutron flux at the image plane but with small L/D = 14, causing a low geometric resolution. The second module, the dimensions of which are presented in figure 1.b, is of the parallel type and has two functions: increase the L/D rate from 14 to 20, improving the image geometric resolution and align the neutron beam by absorbing the diffuse neutrons which, when interacting with the collimator walls, would be deviated from the primary direction and reach the detector from another direction, causing noise in the expected image.

![Collimator's Divergent module](image1)
![Collimator's Parallel Module](image2)

**Figure 1. (a) Collimator’s Divergent module (b) Collimator’s Parallel Module**

The dose equivalent rates at 1 cm from the shield surface of the neutron irradiator were estimated, to meet the domestic CNEN NN -3.01 and international ICRP 60 [3] standards that establish 25μSv/h as the limit of dose equivalent rate at that surface.

The determination of the shielding thickness for fast neutrons involves three main stages: slowing the fast neutrons, absorbing the thermalized neutrons and absorbing the gamma radiation beam from the Am/Be source (primary beam) and from the reactions of the neutrons with the material employed to encapsulate the source and the moderator material, which originate the secondary beam.

The calculated final configuration of the shielding device for the neutron irradiator with an Am/Be source with 5 Ci activity was, as shown in figure 2, 15 cm thick paraffin moderator.
(light yellow); 2 cm thick iron (green); 15.5 cm thick 20% borated paraffin (gold yellow). Thus, the total thickness of the shielding device, taking into account the thickness of the aluminium structural walls (about 0.5 cm), resulted in 33.0 cm.

![Diagram of shielding device](image)

**Figure 2 – Final setup of the neutron irradiator with an Am/Be source with 5 Ci activity.**

The distribution of the dose equivalent rate, at 1 cm from the irradiator external surface, according simulation with code MCNP, forecasts contributions of gamma radiations from the reaction of neutrons with the encapsulating material of 5.8 µSv/h and of 0.29 µSv/h of photons ($^{241}$Am) [4], causing a total dose equivalent rate of 20.36 µSv/h.

The final setup of the irradiator for NR with fast neutrons is represented in figure 3 and figure 4. The structure of the irradiator was manufactured in aluminium and the support table was made in iron, on wheels and coupled to the irradiator, allowing mobility and portability. Besides the Irradiator setup we can observe an auxiliary table coupled to the irradiator that serves as support for the samples to be radiographed and for the imaging system. This table was also constructed in iron and on wheels.

The proposed Neutron Radiography system is of small size, easy handling and not expensive. Besides, it can be transported to the location of the test specimen. Although it provides a low flux, it was designed to obtain NR with fast neutrons with the best possible resolution, and it can be used in quality control of samples for the detection of defects such as cracks and inclusions.

![Diagram of irradiator setup](image)

**Figure 3 – Final setup of the Irradiator for NR with fast neutrons.**
2.2 - Test Specimens

To characterize an Imaging System, identifying the materials that compose the test specimens is of the utmost importance. Thus, the images obtained with the neutron radiography Imaging System have to be qualified, to supply adequate data regarding neutron beam attenuation, when they pass through the sample (test specimen) [6].

The BPI used was constructed according to recommendations of ASTM norm and 545-91. It was made in a Teflon block with square cross-section of 645.16 $mm^2$ and 8 $mm$ thickness, and a central hole with 15.9 $mm$ diameter, containing two lead disks (99.999% pure Pb), two boron nitride disks (BN), all of them with 2 $mm$ diameter, and two cadmium wires (99.999% pure Cd), with 0.64 $mm$ diameter and 12 $mm$ length, arranged as shown in figure 5.

The IVR used was constructed according to ASTM-E-1025-84 norm [7], for the qualification of neutron radiography images, manufactured in cadmium plate with 0.5 $mm$ thickness, holes of 250 $\mu m$, 500$\mu m$ and 1.00 $mm$ diameter, spaced at distances equal to the diameters. A schematic drawing and a picture are shown in figure 6.

The test specimens, Beam Purity Indicator – BPI and Indicator of Visual Resolution – IVR were fixed to a CR-39 sheet, and also in a radiographic cassette, previously fed with film and conversor screen to reduce the effect known as geometric unsharpness. Figure 7 shows BPI and IVR fixed: on a CR-39 sheet and (b) on a radiographic cassette, ready to be submitted to the beam from the Neutron Irradiator.
Figure 5 – (a) Perspective drawing of the BPI, [6]; (b) Picture of the BPI; (c) Original design of the BPI of the Real Time Neutron Radiography Laboratory – LNRTR [8]

Figure 6 – Indicator of Visual Resolution (IVR) (a) Schematic drawing (b) Picture [6]

Figure 7 – Quality indicators, BPI and IVR used as test specimens fixed on CR-39 sheet and (b) on radiographic cassette.
2.3 – Neutron Radiography acquisition

The Neutron Radiographies of the Quality Indicators and metallic samples were performed in the irradiation channel installed in the collimator of the Neutron Irradiator. The NR were obtained by direct exposure method, using for image recording both, a CR-39 sheet and an AA-400 Industrex radiographic film, in close contact with a gadolinium metal converter foil, with 50 μm thickness, placed inside an aluminium Kodak radiographic cassette, closed mechanically by spring pressure. The PM355 SUPERGRADE type, CR-39 sheets acquired from the British company Page Moldings with 625µm thickness, were carefully cut with sharpened stiletto, obtaining enough squares of 50 mm side to record the neutron radiography images of the test specimens. The test specimens fixed to the CR-39 plate were submitted to the fast neutron beam from the Irradiator, without interruption, maintaining the statics of the image acquisition system: test specimens and CR-39 sheet. After some irradiations, the exposure time that provided the best quality of the neutron radiography images was 72 hours. After irradiation, the CR-39 plate was submitted to chemical process to develop the neutron radiography image recorded. To transform the latent tracks into developed tracks, the plastic sheets were immersed in a sodium hydroxide (NaOH) aqueous solution, in the concentration of 250 g of NaOH in 1 liter of distilled water. The developing time used for the CR-39 sheet was 6 hours. This time may be varied. The detector is immersed in the NaOH solution and kept at 90 °C in a Quimis thermostatic water bath. The chemical process is interrupted washing the CR-39 sheet with tap water, until all the NaOH solution is removed from the surface of the CR-39 plastic sheet. A second washing is carried out with distilled water to remove impurities that may interfere with the quality of the images recorded in the detector. Drying must be quick and uniform with hot air, preventing spots in the CR-39 sheets. Drying with paper or similar should be avoided to prevent scratching the CR-39 sheets, i.e., causing spurious defects that impair the image quality.

The direct method of neutron radiography exposition was used to record the images with silver radiographic film in close contact with the converter foil. Regarding the neutron beam, the test specimens were fixed to the aluminium Kodak radiographic cassette, mechanically closed by spring pressure, fed with a gadolinium conversion screen, with 50 μm of thickness and the Kodak Industrex AA-400 film. The Neutron Radiography was obtained after 66 hours of uninterrupted exposure to the neutron beam extracted from the Neutron Irradiator. This time can be optimized, drawing the system’s characteristic curve. The radiographic film was developed at the LNRTR, following the instructions of the film manufacturer. The radiographic process was conducted under the following conditions: temperature of the developer and fixing solutions of 21°C; developing time of 4.5 minutes and fixing time of 7 minutes. The development was interrupted by washing with tap water before immersing the radiographic film in the fixing solution. The radiographic plate was washed with tap water for 20 minutes and then left to dry naturally in place ventilated and free from dust.

2.4 – Characterization of the Neutron Radiography Image

Three parameters are usually determined, besides visual acuity, to assess the imaging capability of the radiographic systems: the densitometry measures of the Beam Purity Indicator – BPI, the resolution and the Modulation Transfer Function – MTF.
According to ASTM-E 545-91 norm, the percent contents of the parameters that influence the quality of the neutron beam can be estimated based on the densitometry measures of the BPI image. They allow to quantitatively determine the radiographic contrast, the contribution of $\gamma$ rays, the image sharpness and the information on the film and development quality. The BPI recommended by this norm should be positioned in the neutron radiography cassette parallel and as close as possible to the film.

The contents of thermal neutrons, fast neutrons, low and high energy $\gamma$ rays must be determined, according to that norm, by densitometry analysis of the BPI image, with each calculation expressed, respectively by:

1) Effective thermal neutron percent content, $C$, or neutron radiography contrast, from film background exposure due to non-scattered thermal neutrons.

\[
C = \frac{(D_\Phi - [sD_{BN} + \Delta D_{Pb}])}{100\cdot D_\Phi}
\]  

(1)

2) Effective scattered neutron content, $S$, which is the darkening percent of the film background caused by the scattered neutrons.

\[
S = \frac{\Delta D_{BN}}{100\cdot D_\Phi}
\]  

(2)

3) Effective $\gamma$ ray content, which is the percent of film background darkening caused by low energy gamma radiation (absorbed by 2 mm of lead).

\[
\gamma = \frac{(D_T - sD_{Pb})}{100\cdot D_\Phi}
\]  

(3)

4) Effective high energy gamma content, $P$, which is the background exposure percent by $\gamma$ rays from the process of pair production (absorbed by 2 mm of lead).

\[
P = \frac{\Delta D_{Pb}}{100\cdot D_\Phi}
\]  

(4)

Where:

1) $D_{BN1}$ and $D_{BN2}$ are the film mean optical densities measured through the images of the boron nitride disks;
2) $D_{Pb1}$ e $D_{Pb2}$ are the film mean optical densities measured through the images of the lead disks;
3) $D_\Phi$ is the film mean optical density measured at the center of the hole in the BPI.
4) $D_T$ is the film mean optical density measured through the image of the Teflon;
5) $\Delta D_{BN}$ is the difference between $D_{BN1}$ and $D_{BN2}$;
6) $\Delta D_{Pb}$ is the difference between $D_{Pb1}$ and $D_{Pb2}$.

The calculations of the above parameters can be performed through the gray levels of the digitized images, because they have a linear proportion relation to the optical density values [5].

Image resolution is defined as the shortest distance that separates two objects so that they can be distinguished. The evaluation of the total resolution of the system is done adjusting a function to the distribution obtained by scanning the gray levels in the interface region of an image of a material highly absorbent of neutrons and direct beam. A function normally used is the Edge Spread Function – ESF, given by:

\[
ESF = A + B\arctan(Cx+D)
\]  

(5)
where, $A$, $B$, $C$ and $D$ are free parameters in the adjustment and $x$ is the scanning coordinate. Figure 8 shows the distribution curve of gray levels – GL in the interface region of a gadolinium foil image.

![Distribution curve of gray levels in the interface region of a gadolinium foil image](image)

**Figure 8 – Distribution of gray levels in the interface region of a gadolinium foil image[6].**

In this method, the resolution $U_T$, is related to the full width at half maximum (FWHM) of the differentiated ESF function – Lorentz distribution, and is given by [9]:

$$U_T = \frac{2}{C} \quad (6)$$

The Modulation Transfer Function – MTF, for an electronic imaging system is defined as the ratio between the contrast recorded and the ideal contrast that should have been produced, as a function of the spatial frequency of the radiographed object [9]. It is a graphic representation of the ability of the system to produce the spatial frequency present in the image. Unlike the resolution that represents a discreet function of the spatial frequency that the system can discern, MTF is a continuous function, where greater amounts of information on system performance and limitations are obtained.

The modulation of a response signal of an imaging system is a fundamental factor in its spatial resolution determination. Modulation can be defined as a function of the Gray Levels – GL, by:

$$\text{Modulation} = \frac{N_{C_{\text{max}}} - N_{C_{\text{mid}}}}{N_{C_{\text{mid}}}} \quad (7)$$

To visualize the contrast of a radiography image, a modulation of up to 90% of the signal amplitude [10] is accepted, an image with values below this limit does not have acceptable quality.

2.5 – Neutron Radiography Images

The CR-39 sheets containing neutron radiography images were scanned using a ScanWizard Pro: 4800 AF equipment manufactured by Microtek, installed at the Laboratory of Neutron Radiography in Real Time – LNRTR/PEN/COPPE/UFRJ. Two scanning methods are available: light reflection and light transmission. The reflection mode resulted in a better image visualization. The neutron radiography images are shown in figures 9 (a), (b), (c), and (d).
Figure 9 – (a) CR-39 Neutron Radiography without digital processing; (b) after contrast enhancing; (c) using a green transparent polymeric filter; (d) image submitted to contrast enhancing

Figure 9 (a) is the scanned image without digital processing. The image observed in figure (b) is the previous image submitted to contrast enhancing, through the Image Pro-Plus computer program for image treatment. The image in figure (c) resulted from the digitization of the image in (a) using a green, transparent polymeric filter. Following the recommendations of Domanus [11], the green filter resulted in the best image. Figure (d) shows the image in figure (c), submitted to contrast enhancing using the Image Pro-Plus program.

The neutron radiography images using radiographic films and gadolinium converter screen are shown in figure 10. In this case, the Neutron Radiographies were digitized in two ways, at the Laboratory of Neutron Radiography in Real – LNRTR/PEN/COPPE/UFRJ: 1) using a ScanWizard Pro: 4800 AF scanner from Microtek, and 2) illuminating the NR in a negatoscope, and then capturing the image with a Sony Cyber-shot digital camera of 10.1 mega-pixels. Figure 10 (a) shows the Illumination of the NR in the negatoscope; figure 10 (b) shows the NR captured by a digital camera. The images are of the test specimens: two steel pieces (ball bearings) one with defect and the other without defect; BPI and IVR.

Figure 11 shows the NR obtained by radiographic film, captured by the ScanWizard scanner, by reflection, of the three test specimens: two ball bearings (with and without defect), BPI and IVR.
Figure 10 – Neutron Radiography of the steel ball bearings (with and without defect), BPI and IVR. (a) under negatoscope illumination and (b) image captured by digital camera.

Figure 11 – Neutron Radiography of the same test specimens scanned in the ScanWizard Pro: 4800 AF Microtek.

3. RESULTS

The analysis of the neutron radiography images of the BPI provides information on the quality of the neutron beam. The parameters $C$, $S$, $\gamma$ and $P$ define the effective contents of: thermal neutrons ($C$), $\gamma$ radiation ($\gamma$), scattered neutrons ($S$) and high energy $\gamma$ rays ($P$), respectively. They are calculated based on the mean gray levels measured [5], and are listed in Table 1. The content of thermal neutrons is given by the relation of gray levels of the images of BN disks one on top of the other, hidden by the Teflon. The $\gamma$-ray content represented by the relation of gray levels of the images of Pb disks, one on top of the other, hidden by the Teflon. Beam alignment is given by the parallelism observed among the Cd wires, positioned in the same way, in relation to Teflon. We can observe that the neutron radiography setup obtained with the Neutron Irradiator has a small contribution of $\gamma$-rays, similar to that emitted by the Argonauta reactor, although the imaging system is near the $^{241}$Am-Be source. The ratio $n/\gamma$ is also a very important parameter to obtain high quality Neutron Radiographies, small $n/\gamma$ ratios result in images of greater sharpness, once the radiographic films are sensible to $\gamma$ rays. The $n/\gamma$ ratio can be improved using a lead or bismuth filter that captures $\gamma$ radiation. Those parameters are calculated and valid [10], only for images obtained with the system of radiographic film coupled to a neutron converter screen.
Table 1 – Parameters C, S, γ, and P.

<table>
<thead>
<tr>
<th>Neutron Radiographic</th>
<th>C (%)</th>
<th>S (%)</th>
<th>γ (%)</th>
<th>P (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Argonauta Reactor</td>
<td>43.62±0.28</td>
<td>6.71±0.02</td>
<td>2.69±0.05</td>
<td>4.02±0.06</td>
</tr>
<tr>
<td>Neutron Irradiator</td>
<td>39.81±0.32</td>
<td>4.46±0.02</td>
<td>1.82±0.12</td>
<td>1.93±0.02</td>
</tr>
</tbody>
</table>

Figure 12. a shows the ESF function of the Neutron Radiography with CR-39 of the system using the CR-39 sheet and Figure 12.b shows the ESF function of the Neutron Radiography using radiographic film and converter.

The curve represent the Edge Spread Function, ESF, adjusted to the gray levels distributed along the spatial scanning of the neutron radiography image by the least square method. For the described conditions, we estimated that the system resolution using a CR-39 sheet is $U_T = 444.4\, \mu m$, figure 12, and using the radiographic film and converter is $U_T = 363.6\, \mu m$, which, in practice means that the NR Image is capable of differentiating different objects separated by a distance equal to or higher than 444.4 μm for the CR-39 and 363.6 μm for the system with radiographic film and converter foil. This resolution is higher than desirable for the intended use.

The maximum and mean values of gray levels of those images and the correspondent MTF values as a function of the spatial frequency of the penetrameter in the neutron radiography images obtained in the Argonauta/IEN/CNEN using radiographic film and converter and in the Neutron Irradiator using CR-39 and the radiographic film and converter are shown in table 2.

Signal modulation was calculated for three scanning spatial frequencies along the image of the penetrameter for the images obtained in the Neutron Irradiator using the CR-39 and the radiographic film, the gray level measures were obtained using the Image-Pro Plus digital image processing program, the values are the mean of ten scans in the region of interest. For comparison the signal modulation was calculated for two spatial scan frequencies present in the penetrameter for the images obtained in the Argonauta Reactor using radiographic film, optical density measures were obtained using a microdensitometer, the values result from a mean of 10 scans in the region of interest [6].

When we analyze the MTF values of the CR-39 images obtained in the Neutron Irradiator we observe very low values. The recorded contrast as a function of the ideal contrast in the representations of the spatial frequencies 2 lines/mm and 4 lines/mm shown by the parameters calculated are well below desirable. In the spatial frequency 1 line/mm, the parameter is a
little better, although still below, as can also be visualized by visual acuity in figure 9 (d). On the other hand, the parameters that define contrast on the images obtained in the Neutron Irradiator using radiographic film and the converter are excellent. Indirectly comparing with the parameters found on the imaging system using radiographic film of the Argonauta Reactor, which are acknowledged in literature as of good quality, we can assume that the NRs are as expected and within a contrast range acceptable for the intended use.

Table 2 – MTF values calculated for neutron radiography assays in Argonauta / IEN/CNEN Reactor and Neutron Irradiator

<table>
<thead>
<tr>
<th>NEUTRON RADIOGRAPHY</th>
<th>SPATIAL FREQUENCY (line/mm)</th>
<th>MAXIMUM VALUE (GRAY LEVEL)</th>
<th>MEAN VALUE (GRAY LEVEL)</th>
<th>CALCULATED MTF VALUE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Argonauta Reactor</td>
<td>1</td>
<td>1.49±0.02*</td>
<td>0.85±0.01*</td>
<td>0.75±0.03</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>1.49±0.02*</td>
<td>1.00±0.02*</td>
<td>0.49±0.02</td>
</tr>
<tr>
<td>CR-39</td>
<td>1</td>
<td>204±1</td>
<td>166±1</td>
<td>0.23±0.01</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>183±1</td>
<td>154±1</td>
<td>0.18±0.01</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>165±1</td>
<td>147±1</td>
<td>0.13±0.01</td>
</tr>
<tr>
<td>Radiographic film</td>
<td>1</td>
<td>241±1</td>
<td>129±1</td>
<td>0.87±0.02</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>245±1</td>
<td>134±1</td>
<td>0.83±0.02</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>245±1</td>
<td>159±1</td>
<td>0.54±0.02</td>
</tr>
</tbody>
</table>

* Optical density values measured using a microdensitometer.

The gray level magnitude can be compared to optical density, once they are related and generate a dimensionless comparative parameter [5].

The comparison between the images obtained with the Neutron Irradiator and the Argonauta Reactor serve only as reference, for a direct and quantitative comparison other parameters such as the exposure time, Reactor operating power, and the experimental conditions of both systems should be taken into consideration.

The analysis of the images by visual acuity allows concluding that the image of Teflon of the BPI digitizing with digital camera is more visible than the same image digitized with a scanner, however the image of the scanner offers a better contrast and resolution in the penetrator visualization. The parallelism of the BPI wires shows the neutron beam alignment.

As for the metal structure of the ball bearings, we can observe that the FNR using radiographic film, figure 10, reveals more details than the NR using CR-39 with the boron foil. The test specimens, BPI, penetrator that were examined are in accordance with the parameters in the literature [11] and allowed qualitatively and quantitatively characterizing the imaging systems used.

Before an image is evaluated, we must specify the objective of this image and only then we can determine which is the more adequate detection and imaging system to be used. For less refined inspections, the CR-39 or the radiographic film can be used or both of them, because one can complement the other in detail visualization. We can conclude that the NRs performed in the Neutron Irradiator channel are acceptable for the intended inspection because they present good resolution, contrast and visual acuity to detect impurities, different materials, inclusions and voids.
4. CONCLUSIONS

The NR technique using fast neutrons is still little used, but it can be a powerful tool for the inspection of bulky samples, mainly those composed of high density materials. Thus, the development of this subject represents an important breakthrough in the research of Non-Destructive Tests of the LNRTR/PEN/COPPE/UFRJ.

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