Leonardo Gariboldi *

Developing a technique for researches in cosmic-ray physics: Nuclear emulsions from Bristol to Europe

(1) Introduction

A historical reconstruction of cosmic ray and elementary particle physics in the 40–50's more sensitive to the technical aspects of the physical research put in evidence the role played by the studies made with nuclear emulsion technique. For instance, in *Image and Logic* [1], Peter Galison gives an interesting analysis of the studies made on nuclear emulsions by the group of Bristol after the first suggestion by Heitler to Powell, following the first expositions made by Blau and Wambacher [2] on the Austrian Alps. With nuclear emulsions, Powell's group was able to discover the pion.[3] Another interesting result obtained with this technique applied to cosmic rays was the study of K-mesons in the 50's. We can frame in this period the researches on nuclear emulsions and on cosmic rays made by the physicists led by Giuseppe ("Beppo") Occhialini who exported the Bristol technique to Belgium and to Italy.

Occhialini left Powell's group in 1948 and went, with his future wife Connie Dilworth, to Brussels to the *Centre de Physique Nucléaire*. He had been called there by Max Cosyns to start a new laboratory working with nuclear emulsions. Occhialini was the scientific leader of the Brussels group up to 1959. In the same years, Occhialini was appointed university professor in Genoa from 1950 to 1952, and in Milan from 1952 on. In Brussels, Occhialini's group studied the new emulsions: the Kodak NT2 and NT4 by Berriman, and the Ilford G5 by Waller. Actually, the NT4 and G5 emulsions were devices of fundamental importance for cosmic ray physicists since they were used to detect also relativistic electrons.

Occhialini played also a fundamental role in the development of other groups of research on nuclear emulsions in Italy and their belonging to a European network of laboratories devoted to the study of cosmic radiation. We can just remember, for example, the "pilgrimage" of Italian researchers to Bristol or Brussels to learn the technique of nuclear emulsions, such as: Carlo Franzinetti, Marcello Ceccarelli, Michelangelo Merlin, Alberto Bonetti, Giulio Cortini, Giulio Levi-Setti, Giovanna Tomasini, and Livio Scarsi.

(2) Classical methods to load nuclear emulsions

Nuclear emulsions were loaded with particular elements to study the *Z*-dependence of some cosmic ray interactions: disintegration stars, meson production, pair production, There were three classical methods to load nuclear emulsions.

With the first method, the emulsion was soaked with the loading element by placing it in a saline solution. This method could cause problems during the processing of the emulsions, obliging the observers to a statistical comparison of the results obtained with normal emulsions.

The second method (usually called the "sandwich" method) consisted in placing a thin sheet of the loading material between two emulsion plates. A grid was then produced on the emulsions with X-

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* Università degli Studi di Milano, Istituto di Fisica Generale Applicata, Milano, Italy; email: leonardo.gariboldi@unimi.it.

[1] Galison, Peter (1997): *Image and Logic. A Material Culture of Microphysics* (Chicago: The University of Chicago Press, 1997).

[2] Blau, Marietta; Wambacher, Herta (1937): "Disintegration Processes by Cosmic Rays with the Simultaneous Emission of Several Heavy Particles" *Nature* 1937, *140*: 585.

[3] Lattes, Cesare Mansueto Giulio; Muirhead, Hugh; Occhialini, Giuseppe Paolo Stanislao; Powell, Cecil Frank (1947): "Processes Involving Charged Mesons", *Nature* 1947, *159*: 694.

rays. This method had the disadvantage of a high loss in the number of identifiable tracks. It was but possible to assign a track to a grain of loading material.

The third method consisted in the diffusion of the loading material into the emulsion as microscopic grains.

(3) The wire method to load emulsions

The wire method to load nuclear emulsions was suggested by Beppo Occhialini, Gilberte Meulemans, and A.M. Vincent [4] in 1951. They used wires of the loading material, a few microns thick, to form a grid inside the emulsion. The results obtained with the wire method were better than those obtained with the sandwich method, but still worse than those obtained with the grain-diffusion method. The scanning of the emulsions was but much faster, especially if conducted along the wires.

To produce this kind of emulsions, they poured the emulsion on a grid of nylon and platinum wires. The wires were covered by a layer of thorium, with a diameter of 30 µm, at 1 mm of distance one another, and where covered by a 200 µm thick emulsion.

With this kind of emulsions, they could get interesting results in the study of cosmic rays, provided that they made an analysis of the possible distortions due to the presence of wires inside the emulsions. The distortion could be ignored at 30 µm from the wire.

Since the wire emulsions seemed to be a powerful device, Beppo Occhialini and Alberto Bonetti reconsidered the function of the wire .[5] The wire was now thought as the support of a cylindrical layer of emulsion. The wire could also be substituted by a thin tube filled with a loading liquid. In 1951, Occhialini and Bonetti thus suggested to produce cylindrical plates that had to be carefully analysed with respect to the distortion of the tracks.

The shadow produced by the wire or the thin tube permitted to use cylindrical emulsions as localising detectors, with an advantage in directionality with respect to other wire detectors, such as the Geiger-Müller counters. A system of two joined cylindrical emulsions was used to detect radioactive sources in biological tissues.

(4) The clearing method

As for the processing methods of emulsions exposed to cosmic rays, we must observe that during the processing they met a difficulty. The emulsions became more and more non-transparent, mainly because of the accumulation of colloidal silver grains in the emulsion. This problem affected most of all thick emulsions (from 100 µm on).

> Although the progress in application of photographic plates to nuclear physics in recent years has been very great, *little attention has been paid until recently to the processing. [...] The way in which we process nuclear plates determines what we see*, [...].[6]

Beppo Occhialini, Connie Dilworth, and Eric Samuel found a simple way to avoid the accumulation of colloidal silver.[7] This method was based on the different activity of the processing materials with the variation of temperature. After a normal development, the emulsion was alternatively soaked in baths of HCl solution (1:1200), distilled water, hypo, and distilled water again. The soaking time ranged from 15 minutes to an hour depending on the thickness of the emulsion. The process was repeated until a sufficiently clean emulsion was obtained. The emulsions were tested with γ-rays, at different temperatures, to measure the quantitative effect of the temperature on the diminution of the

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[4] Meulemans, Gilberte; Occhialini, Giuseppe Paolo Stanislao; Vincent, A.M. (1951): “The Wire Method of Loading Nuclear Emulsions”, *Il Nuovo Cimento* 1951, *8*: 341.

[5] Bonetti, Alberto; Occhialini, Giuseppe Paolo Stanislao (1951): “Cylindrical Emulsions” *Il Nuovo Cimento* 1951, *8*: 725.

[6] Dilworth, Constance Charlotte; Occhialini, Giuseppe Paolo Stanislao; Vermaesen, L. (1950): “On Processing Nuclear Emulsions. Part 1. Concerning Temperature Development” *Bulletin du Centre de Physique Nucléaire de l’Université Libre de Bruxelles* 13°, Février 1950. Quotation on page 1. Evidence is mine.

[7] Dilworth, Constance Charlotte; Occhialini, Giuseppe Paolo Stanislao; Samuel, Eric (1948): “Eclaircissement des plaques photographiques nucléaires”, *Bulletin du Centre de Physique Nucléaire de l'Université Libre de Bruxelles*, 2, Août 1948.

sensibility of the emulsions. The sensibility was defined as $S = (N_T - N)/(N_{20} - N)$ where N was the number of grains impressed before the exposition; N_T was the number of grains impressed after exposition at temperature T; and N_{20} was the number of grains impressed after exposition at 20°C.

It was not easy to determinate the dependency of the sensibility on temperature, since γ-rays of different energy actually produced β-rays with different ionising power. It was thus not possible to determinate the variations of sensibility for any particle of a given specific ionisation. The sensibility depended furthermore on the way of processing the emulsions and on the very kind of the emulsions themselves:

> Le problème de la désensibilisation a donc été résolu pour les plaques Ilford C_2 et Kodak NT2a par l'emploi d'un mélange de glace carbonique et d'acétone qu'il est facile de se procurer. Pour des plaques plus sensibles, des temperatures plus basses seront necessaries, come celle de l'oxygène liquide, ou mieux de l'azote liquide pour des raisons de sécurité du transport.[8]

(5) The temperature development method

The chemicals used for the development of nuclear emulsions had to satisfy a series of characteristics: resistance to oxidation in air; low alkalinity, to avoid the emulsion swelling and deformation in the warm phases of the development; low concentration, to avoid the precipitation in the cold phases; molecular simplicity, to have a constant rate of penetration; low solving action on silver halides.

Occhialini's group used an organic, highly toxic developer: amydol (2:4-diaminophenol dihydro-chlorid). Amydol is active also in lightly acid solutions used to prevent oxidation in air. Amydol could be used in low concentration and has a simple molecular structure. With the temperature development method [9], amydol was used to process emulsions thick up to 1,2 mm. Occhialini thought they could even bring the thickness up to 3 mm.

The first step of the temperature development — the immersion in the developing bath — had the aim to fill the emulsion with the developer at low temperature, for a time depending on the thickness of the emulsion. The thickest emulsions were before soaked in distilled water in order to reduce the immersion time in the developer. Very thick emulsions, once processed, could have been studied with the new technique of reflecting microscopes, invented by W.J. Bates and Occhialini [10] in 1948. At very low temperatures, the penetration of the developer diminished, but its activity diminished even more. For G5 and NT4 plates, they usually worked with developing baths at 5°C[11].

After having soaked the emulsion with the developer, the second step — the warm developing — started. They heated the emulsions dry. Once taken from the developing bath, the emulsions were dried with paper and put in a vessel at the optimal temperature with an inert gas to prevent oxidation. The temperature depended on the kind of particles they wanted to observe:

> Where discrimination is required between highly ionising particles e.g. fission fragments and alpha particles, soft development is needed […]. On the other hand, when particles of very low ionising power e.g. electrons, mesons and protons at relativistic energies, are to be observed, the problem becomes one of discrimination between the tracks and the

[8] Cosyns, Max G.E.; Dilworth, Constance Charlotte; Occhialini, Giuseppe Paolo Stanislao (1949): "Obturateur thermique pour plaques nucléaires", *Bulletin du Centre de Physique Nucléaire de l'Université Libre de Bruxelles*, 6, Janvier 1949. "The problem of the desensibilation has thus been resolved for Ilford C2 and Kodak NT2a plates by the use of a mix of dry ice and acetone which was easy to get. For more sensible plates, lower temperatures would be necessary, such as that of liquid Oxygen, or better of liquid Nitrogen because of security of transport". Quotation on page 3.

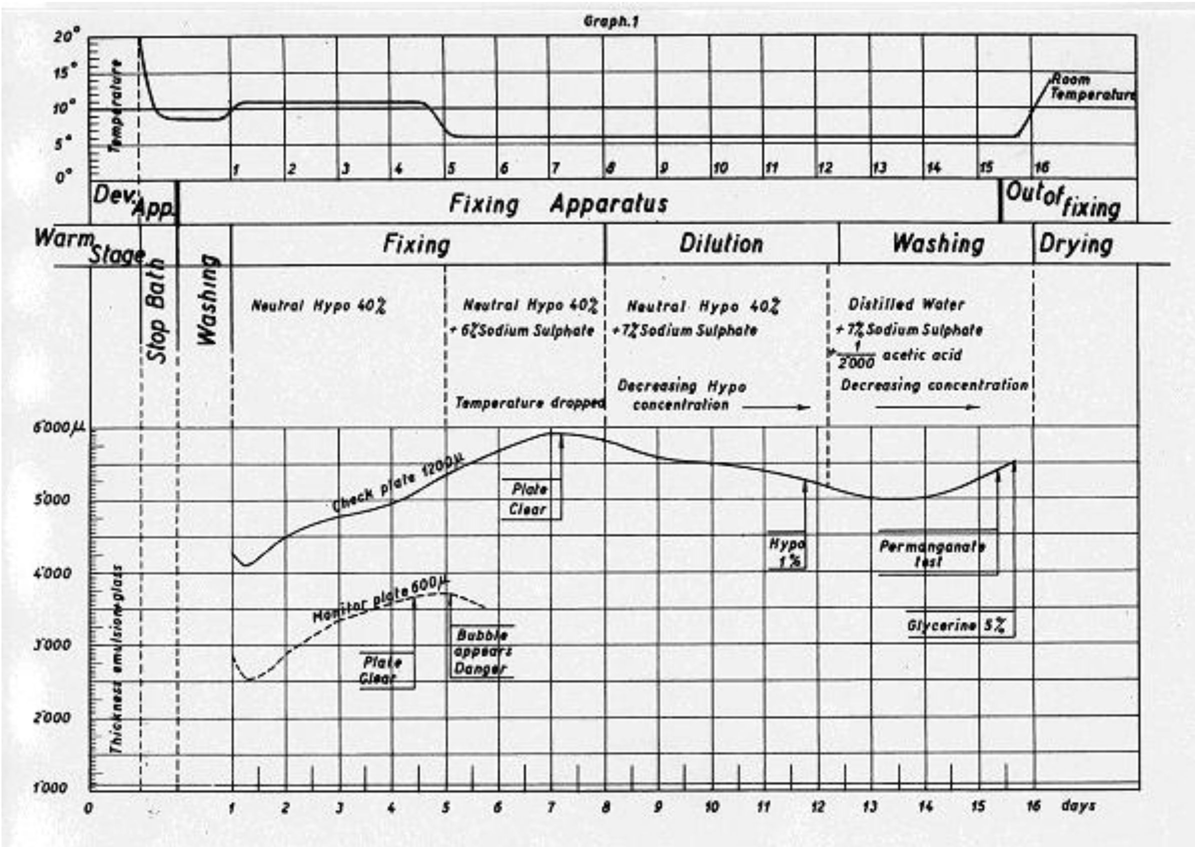
[9] Dilworth, Occhialini, Vermaesen (1950), *op. cit.*, Bonetti, Alberto; Dilworth, Constance Charlotte; Occhialini, Giuseppe Paolo Stanislao (1951): "On Processing Nuclear Emulsions. Part II. After Development Techniques", *Bulletin du Centre de Physique Nucléaire de l'Université Libre de Bruxelles*, 13b, Mars 1951.

[10] Bates, W.J.; Occhialini, Giuseppe Paolo Stanislao (1948): "Applications of the Reflecting Microscope to the Nuclear Plates Technique", *Nature* 1948, *161*: 473.

[11] Herz, A.J. (1952): "The Development of Ilford G5 Emulsion with ID19 Developer", *Journal of Scientific Instrument* 1952, *29*: 15.

background, and while full development is required, ever development leads to high background and consequent difficulties in recognition. In Ilford G5 plates it has been found that 1° difference of temperature will decide between under, full, and over development.[12]

With the third step — the final washing and fixing —, the development was stopped with a gradual decrease in temperature. At about 13°C, the plates and the vessel were washed with distilled water to remove the developer. After the washing, they put a diluted solution (0,2% – 2%) of cold acetic acid (6 – 9°C). At the end, the plates were washed under running water and fixed.



“Steps of Occhialini's Temperature Method of Development”

In the development of thick emulsions they had to prevent the distortion of the tracks, that is the slipping of the upper layers of the emulsion on the lower ones. In the case of a regular deformation, they measured the distortion of the plate with a “distortion vector” **K**. The curvature of the track was proportional to $\mathbf{k} = \mathbf{K}/T^2$, where T was the thickness of the emulsion. The vector curvature **k** was measured in Covans (after Cosyns and Vanderhaeghe) defined by the formula with **K** in micron and T in millimetres. The temperature developing method by Occhialini could not cause distortion of the tracks, if properly used.

Possible causes of distortion of the tracks were: 1) general distortions in the development: caused by too high temperature and alkalinity in the warm step, or by sudden variations of temperature and pH; 2) general distortions in fixing and washing: caused by sudden variations of temperature, pH or saline concentration, too fast or vorticous movements of the liquids on the surface of the plates, non horizontal position of the plates, presence of bacteria; 3) general distortions in drying: caused by too fast drying, too high temperature, inhomogeneous conditions of temperature and humidity inside the plates, non horizontal position of the plates; 4) local distortions: caused by dust and impurities penetrated into the emulsion, or by touching the plates with one’s hands; 5) distortions preceding the

[12] Dilworth, Occhialini, Vermaesen, *op. cit.* Quotation on pages 6 –7.

developing. A careful application of the development method permitted to obtain tracks with a distortion of about **k** = 15 Covan, whereas the previous results were of about **k** = 200 Covan.

The reduction of the distortion of the tracks was the most important problem in those years, followed by the one concerning the transparency of the emulsions. The velocity of the development was sacrificed in favour of the conservation of constant conditions in the fixing devices.

(6) The study of relativistic particles and the β-decay of muons

The improvements in the production of more sensitive nuclear emulsions [13] and in the technique of development and examination of tracks, permitted the study of relativistic phenomena [14], and the observation of the β-decay of the muon.[15]

> The possibility of processing plates of great thickness (up to 600 microns) afforded by temperature development has brought into use plates in which it is possible to follow to the end even highly scattered particles with a range of several hundred microns in the emulsion.
>
> In a group of Kodak NT4 plates 400 microns thick, exposed at the Pic du Midi, we have observed phenomena which could not be detected in the old plates, and have also obtained more accurate information on phenomena already observed.[16]

Furthermore, analyses on the disintegration stars put in evidence the existence of kinds of particles not yet studied.[17] Occhialini's group, with the C2 plates, could detect the existence of δ-rays, of heavy fragments such as He^6 and C^{10}, and relativistic particles. They also suggested three possible explanations of the fast electrons observed in the disintegrations stars: a) as decay product of the mesons captured by the atoms but not yet fallen on the nuclei (improbable for π and μ mesons); b) as high energy Auger electrons emitted during the atomic capture of a meson, without the production of a disintegration star; c) as β-rays emitted by the daughter nucleus after the meson capture and after the evaporation of one or more neutrons.

Researches on mesons made using nuclear emulsions showed that 62% of the mesons decayed inside the emulsion, while 38% was captured by nuclei. Only 2,5% of the latter were negative pions captured by nuclei without detectable stars, while 97,5% were muons. With no direct evidence, they supposed that the muons were equally divided among positive and negative ones. The captured negative muons were suggested to be about 80%, since the probability to stop a muon is proportional to Z, while the probability to capture it is proportional to Z^4.

The emission of slow electrons was mainly attributed to an Auger effect accompanying the capture of muons, or maybe also of negative pions capture without producing a star. It was very difficult to determinate the real number of slow electrons and their origin, because of the high scattering. Slow electrons with $E < 20$ keV were not identified, since they had a too short track. With $20 < E < 30$ keV they could be easily obscured by the mesons tracks, and with $50 < E < 150$ keV they were easily identified only at the beginning of their track.

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[13] Berriman, Robert W. (1948): "Electron Tracks in Photographic Emulsions" *Nature* 1948. Berriman "Recording of Charged Particles of Minimum Ionizing Power in Photographic Emulsions" *Nature* 1948, *162*: 992.

[14] Cosyns, Max G.E.; Dilworth, Constance Charlotte; Occhialini, Giuseppe Stanislao; Schönberg, Mario (1949): "Double Stars with Relativistic Particles from Cosmic Rays", *Nature* 1949, *164*: 129. Cosyns, Max G.E.; Dilworth, Constance Charlotte; Occhialini, Giuseppe Paolo Stanislao; Schönberg, Mario; Page, N. (1949): "The Decay and Capture of μ-Mesons in Photographic Emulsions", *Proceedings of the Physical Society* 1949, *62*: 801.

[15] Brown, R.H.; Camerini, Ugo; Fowler, Peter Howard; Muirhead, Hugh; Powell, Cecil Frank; Ritson, D.M. (1949a): "Observations with Electron-sensitive Plates Exposed to Cosmic Radiation. I. Decay of μ-mesons", *Nature* 1949, *163*: 47; *idem*, (1949b): "Observations with Electron Sensitive Plates Exposed to Cosmic Radiation. II. Further Evidence for the Existence of Unstable Charged Particles of Mass ~ 1000 m_e and Observations on their Mode of Decay", *Nature* 1949, *163*: 82.

[16] Cosyns, Dilworth, Occhialini, Schönberg (1949), *op. cit.* Quotation on page 129.

[17] Bonetti, Alberto; Dilworth, Constance Charlotte (1949): "Heavy Splinters in Cosmic Ray Stars", *Philosophical Magazine* 1949, *40*: 585. Hodgson, P.E.; Perkins, Donald Hill (1949): "Heavy Nuclear Splinters", *Nature* 1949, *163*: 439.

(7) The use of electron-sensitive emulsions

The identification of high energy particles with electron sensitive emulsions was based on the standard technique of measuring range, grain density, scattering, number of δ-rays along the track, and deflection in a magnetic field. From these data, they could calculate energy, velocity, mass, and charge sign of the particle.

The range-energy relation used in the study of the pions was based on a relation found by Cesare M.G. Lattes, Peter H. Fowler, and Pierre Cüer[18] for protons and α-particles: $R = E^{1.73}$ valid up to 13 MeV. From the relation $R = f(\nu)\ M/Z^2$ they could obtain the range-energy relation for any kind of particles, once it was known for one. In the case of multiply charged particles, the trend to reduce their charge by electron capture invalidated a simple use of these relations. Lattes, Fowler, and Cüer's relations could be used also with the new emulsions that had the same stopping power of the normal emulsions.

The multiple scattering angle $\alpha = \varphi\ (\nu, Z)\ Z/E$, where $\varphi(\nu, Z)$ was a theoretically known function, was also used to determinate the mesons mass, with a background error of 0,5° per 50 μm. A reduction of the background was obtained in Bristol and Brussels, and permitted to measure energies up to 1 GeV. The mean scattering angle gave important information on the nature of the particles if the determination of the range was not possible. They could use, for instance, the energy loss-scattering relation $\alpha = f(dE/dR)/M$ that was different for each kind of particle and permitted to determinate the nature and mass of the particle.

(8) Further developments

An improvement in the researches with nuclear emulsions was possible in Milan thanks to the application of a principle already known to microscopic technique. The problem concerned the possibility to make short movements between the observed object and the ocular micrometer. The solution was the measure of the rotation around an axis perpendicular to the optical axis of one or more plane, parallel blades, introduced along the optical tube of the microscope. This method had already been used in optical instruments, such as Klausen's blade micrometer, and permitted to shift the image of the object in the plane of the micrometer, and measure its shift. An instrument of this kind was the MS2 Koristka microscope, built by Cantù for Occhialini's group.

> With a device of this kind, we made a series of preliminary measurements both of diameters of single grains, and of distances between the grains. The results have been so satisfactorily both for rapidity and precision, that we have been induced to test the method on one of the most delicate measurements on nuclear plates, that of the thickness of the tracks left by singly charged particles at the end of their range.[19]

The diameter of the single grains, the distance between the grains, and the density of δ-rays, were useful to determinate the velocity of the particles, and to contribute to the determination of the mass and charge of particles whose ionisation was three times the minimum.

Since the measurement of the track thickness was made with respect to a fixed straight line parallel to the track itself (the rotation axis of the blade), from the profile of the track they could determinate also the sagitta of the multiple diffusion. With very thick tracks, they could so determinate the barycentre of the track from the coordinates of very single grain.

At the 1954 Varenna School, Occhialini described the limits and the possible improvements of the nuclear emulsions technique.

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[18] Lattes, Cesare Mansueto Giulio; Fowler, Peter Howard; Cüer, Pierre (1947): "Range-Energy Relation for Protons and α-Particles in the New Ilford 'Nuclear Research' Emulsions", *Nature* 1947, *159*: 301.

[19] Bonetti, Alberto; Dilworth, Constance Charlotte; Ladu, Mario; Occhialini, Giuseppe Paolo Stanislao (1954): "Misure in lastre nucleari", *Rendiconti dell'Accademia Nazionale dei Lincei* 1954, *17*: 311.

"Con un dispositivo di questo genere abbiamo eseguito una serie di misure preliminari sia di diametri di grani isolati, sia di distanze tra i grani. I risultati sono stati così soddisfacenti per rapidità e precisione, che siamo stati indotti a provare il metodo su una delle misure più delicate in lastre nucleari, quella della lunghezza delle tracce lasciate da particelle di carica unitaria a fine percorso." Quotation on page 311.

It is well to note the importance of knowing the instrumental limitations of the nuclear emulsions, and to accept them scrupulously when one is making refined measurements, such as those relative to the velocity and the mass of fundamental particles. These measurements are concerned, not only with the number and the distribution, but also with the shape and the density of the silver grains along the tracks of the ionizing particles; the fundamental requirements relative to the physical and photographic properties of emulsions and the methods of manipulation are derived from them:

1) Sensitivity to ionizing particles and to their variations in velocity (stopping power, discriminating power, sensibility to the development);
2) Uniformity and flexibility of the development;
3) Reproducibility of the geometrical conditions of the exposure.[20]

Nuclear emulsions were a satisfying device for many of their properties, but much less for some as the discriminating power. The uniformity and flexibility of the development had their effect on the results one could obtain for the developed tracks. The quantity of developer caused a significant colouring of the tracks, the loss of transparency, and the loss of discrimination. The aim of the second step was to guarantee the conservation of the structure of the grains and the uniformity of the optical conditions. A typical problem of the fixing was the corrosion of the grains, beginning from the surface layers of the plates, with the possibility to get the total destruction of the tracks. The tracks distortion could be avoided only partially by careful elimination of water and by soaking the emulsion with plasticizing materials such as glycerine or sorbitol. At the end of the fixing, it was better to protect the emulsions with a thin layer of oil or with a thin sheet of glass or mica.

One of the aims of Occhialini's group was the possibility to get useful information from the profile of short tracks to find the sensibility of the parameters with the variation of ionisation in few microns.[21] The measurements were made with the Koristka MS2 microscope. With two blades, rotating in perpendicular planes, they could measure the shifts along two perpendicular directions, avoiding the little shifts of the plate. They drawn the projected profile of the track by measuring the distance of the two extreme points from a fiducial line in the ocular shifting the line (filar micrometer) of the image (Klausen micrometer). From such information they could determinate the variation in the mean thickness of the dark parts of the track, the length of the gaps, and the blacked area per 100 µm.

Measurements were made with monochromatic light of constant intensity in the ocular, measured by a photometre in the ocular itself. Because of the decreasing efficiency of the scanner, the resting times were fixed at regular and predetermined times. The dependence of the thickness of the track on the velocity of the incident particle was an important method to distinguish particles with the same charge but of different mass. This technique was applied to tracks at most a few hundred microns long, but "it is evident that the usefulness of thickness measurements will extend to longer ranges on tracks of higher mass and charge".[22]

As a conclusion, we can observe that in the late 40's – early 50's the nuclear emulsions technique evolved as an impressive tool for researches in nuclear and particles physics. Occhialini's contribution in those years were recognised to be of the utmost importance in the last attempts of the *cosmiciens* to obtain information on elementary particles of cosmic origin before the definitive advent of the accelerators era. A technique used first on continental Europe (Blau and Wambacher) was soon exported to Bristol where it exploited its potentialities (Powell's group) and was again re-exported back to continental Europe (Occhialini's group and others) as an exemplar case of spreading of a technique from local to global research.

[20] Bonetti, Alberto; Occhialini, Giuseppe Paolo Stanislao (1954): "Technique of Nuclear Emulsions", *Supplementi al Nuovo Cimento* 1954, *2*: 222. Quotation on page 222.

[21] Alvial Cáceres, Gabriel; Bonetti, Alberto; Dilworth, Constance Charlotte; Ladu, Mario; Morgan, J.; Occhialini, Giuseppe Paolo Stanislao (1956): "Measurements of Ionization", *Supplementi al Nuovo Cimento* 1956, *2*: 244.

[22] *Ibid*. Quotation on page 252.