Inhomogeneous Beam Brightness Intensity Converter (IBBIC) as support software for cathodoluminescence microscopy studies and images pre-processing

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Article

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ABSTRACT

Cathodoluminescence (CL) has gained over time a growing interest in science and technology. CL microscopy has one of its most common applications in geological research, however, such analysis is affected by brightness values not dependent on the CL response of analysed samples, but on the nature of instrumentation currently adopted.

In this work a software has been developed, IBBIC, which is able to convert the brightness intensity of inhomogeneously distributed beams to simulate the response that the investigated materials would have had under the theoretically optimal conditions. Furthermore, it can reduce the contribution of a particular kind of noise, which remarkably manifests values of brightness unrelated to any actual luminescence effect. Moreover, it is equipped with tools capable of supporting the operator in choosing the appropriate parameters for image processing minimizing artefacts, which would otherwise be demanding, laborious and difficult to apply on a large collection of images.

To assess the IBBIC software, it was run on 412 images provided by literature and acquired using CL microscopy in widely varying conditions. The results show that it can be applied to microscopy CL studies, as it does not significantly distort the CL response of the analysed samples, but it makes it possible to study correlations between image portions, which are far apart from each other.

The software may be extremely helpful in CL studies and other similarly error-prone instruments, as such images pre-processing can pave the way for hitherto challenging and not widespread quantitative/semi-quantitative image analysis.

KEY-WORDS: Brightness correction, Heterogeneous Beam, Software, Cathodoluminescence, Image Analysis.

INTRODUCTION

The cathodoluminescence (CL) phenomenon was first described in 1879 by Crookes as a result of an experiment in which a glass was bombarded by converged electron rays (Crookes, 1879). In 1953, Adirowitsch (Adirowitsch, 1953) gave an explanation of that on the basis of the energy band model. As reported by him, CL may be briefly explicated by the excitation of electrons from the valence bands to the conduction bands due to the cathode beam radiation, and their subsequent recombination. During this last step, photons, having wavelengths corresponding to their own material band gap, were emitted, thus generating the observed colour.

CL has been an aid to geological studies since the second half of the 20th century, when in 1965, with the studies of Smith & Stenstrom (Smith & Stenstrom, 1965) on quartz, feldspar and carbonates, and those of Long & Agrell (1965), and Sippel (1965), on thin sections, the use of this instrumentation was started in geomaterials field.

CL studies are performable with the aid of an electron microprobe, a scanning electron microscope (SEM), a transmission electron microscope (TEM) or some special optical cathodoluminescence microscopes. The latter approach, thanks to the capability of observing relatively large rock sections, and possibly comparing them with the observations under petrographic optical microscope, has aroused much interest in the study of geomaterials (Boggs & Krinsley, 2006; Marshall, 1988). This attention is due to the fact that data obtained using CL highlight further information on the nature of the mineralogical phases. That is commonly possible thanks to the interpretation of their different colours acquirable under the effect of the electron beam (Matter & Ramseyer, 1985). The multitude of factors on which minerals CL response depends, makes it difficult to identify the causes that generate the observed colours. Generally speaking, they depend on activating ions or defect centers present inside the object generating luminescence, but if the latters are absent or exiguous, a non-luminescence of the investigated material may be found (Pagel et al., 2000). This last phenomenon can occur even if there is a concentration of elements (e.g., Fe^{3+}, Fe^{2+}, Ni^{2+} and Co^{2+}) to act as a quencher of CL (Marfunin, 1979).

Although CL results assessment might be an extremely delicate practice, it is widely employed since it is considered an outstanding method to detect both structural and chemical information about solids. The following are a few examples of the CL application in various fields of study: the investigation of building materials such as natural stone, cement, mortar and concrete (materials science) (Götze, 2009); the distinction between authigenic quartz precipitated as cement in sandstones and detrital quartz grains (petrography) (Sippel, 1968); the discrimination of ceramic groups, concurrently with information related to provenance or manufacturing processes (cultural heritage) (Odelli et al., 2022); the identification of rare earth element (REE)-bearing minerals in ore (mining sector) (Imashuku & Wagatsuma, 2021); the classification of zircons by combining them with deeps (mineralogy) (Zheng et al., 2022); the examination of the disparity in CL of quaternary carbonate deposits (sedimentary geology) (Braithwaite, 2016); the identification of meteorite impact microstructures and their demarcation from tectonic deformations ones (planetary sciences) (Hamers & Drury, 2011); the analysis of raw pharmaceutical materials and formulated products (pharmaceutical industry) (Nichols, 2012). Notwithstanding these cases are only an exiguous part among the countless bibliographic papers and they highlight the huge impact that CL has on solid materials studies.

The interest shown in the geomaterials field is largely linked to optical interpretations based on observations under microscope. The commonly adopted equipment requires an electron gun placed at an angle to the incident surface while the acquisition instrument acquires information perpendicularly to it. Due to instrumentation geometry and the intensity of the electron beam, higher in its centre and gradually less intense moving away from it, analysed samples are subjected to different intensity of the ray incident. As a consequence, theoretically identical parts of the same sample would have a different response in terms of CL depending on the intensity of irradiation (Odelli et al., 2022). Although data assessments related to these typologies of study are often qualitative and this aspect is not significant, for a more quantitative approach it might be an obstacle to the interpretation. For example, regarding image analysis it is not possible to carry out thresholds to separate uninteresting portions because of the intrinsic removal of meaningful ones. Threshold is a basic and very common operation in imaging in which a binary operation is applied to the image to isolate regions of the image comparing a pixel's characteristic (colour, saturation, luminosity or combinations thereof) with a threshold value. The resulting binary image can be used as a filter to remove the contribution of undesired pixels. However, in Fig. 1, in which some examples of image collected and analysed in this work are showed (Fig. 1A and Fig. 1C), it is possible observe how, by using images obtained from CL microscopy, it is often impossible to make adequate thresholds, due to the different contribute of the irradiation intensity in different image portions (Fig. 1B and Fig. 1D).

The aim of this work was therefore to develop a software able to support operators in mitigating brightness gradient of the images acquired with the CL optical microscope, in order to bring the information obtained as close as possible to that which should be obtained with a homogeneous beam. For this purpose, a converter able to provide the hypothetical result of a homogeneously distributed beam, having the same intensity across the analysed sample, was projected and applied. Furthermore, with the same intent, it was tried to remove, from the acquired images, brightness errors not dependent on the investigated material, but on instrumental acquisition limits.

This paper work starts from Odelli et al. (2022), but it focuses mainly on the operator user experience, providing the operator with a set of tools capable of simplifying the choice of optimal parameters for the correction algorithm, thanks to the immediate, clear and precise feedbacks they provide. Thus, the IBBIC software speeds up the whole correction process and can also be used by operators who lack a wide proficiency in image processing theory and tools. Using the IBBIC software is possible to carry out brightness corrections in the specific context of CL analysis. The context reduction permits to improve the software usability, because the user is not overwhelmed by a great variety of image processing algorithms and parameters, but he has to tune only a few parameters. Also, the correction results are previewed in real-time while the parameters are tuned, allowing the operator to assess accurately its choice basing not only on a few sparse results, but on the continuous variation of many results, directing the parameters choice in the direction which provides the best improvement. Finally, the software permits to cycle on a whole image collection, previewing the results of the same correction, and it allows the user to correct the whole collection at once using the same parameters.

IBBIC software is downloadable for free from the link https://github.com/Volpintesta/IBBIC or contacting authors.

The software performances have been compared with the ones of an equivalent MATLAB script (MathWorks, n.d.-a) which uses the Image Processing Toolbox (MathWorks, n.d.-b). The MATLAB script, as expected, has proven to be consistently better (with an average image conversion speed that is 3 times faster than the IBBIC software's one). The performance difference, even if the software uses at its core the OpenCV (OpenCV, n.d.) libraries, which are well known for their flexibility in image processing and for their performances, is justifiable with the choice to develop in Java (Java, n.d.) to be able to develop a product that could easily be used in a wide range of operating systems without requiring neither the user to compile the code by himself, neither the authors to distribute a specific software versions for each operating system used by researchers, a task which would be affordable by a software house, not by some lone researchers.

So, the target of this software is not an image processing expert, who would be able to write its own script in MATLAB, using a much more flexible and powerful tool, but a not-technician researcher or geologist with no expertise in image processing or in MATLAB, who is willing to sacrifice some performances to have a task-specific, easy to use tool with immediate feedbacks.

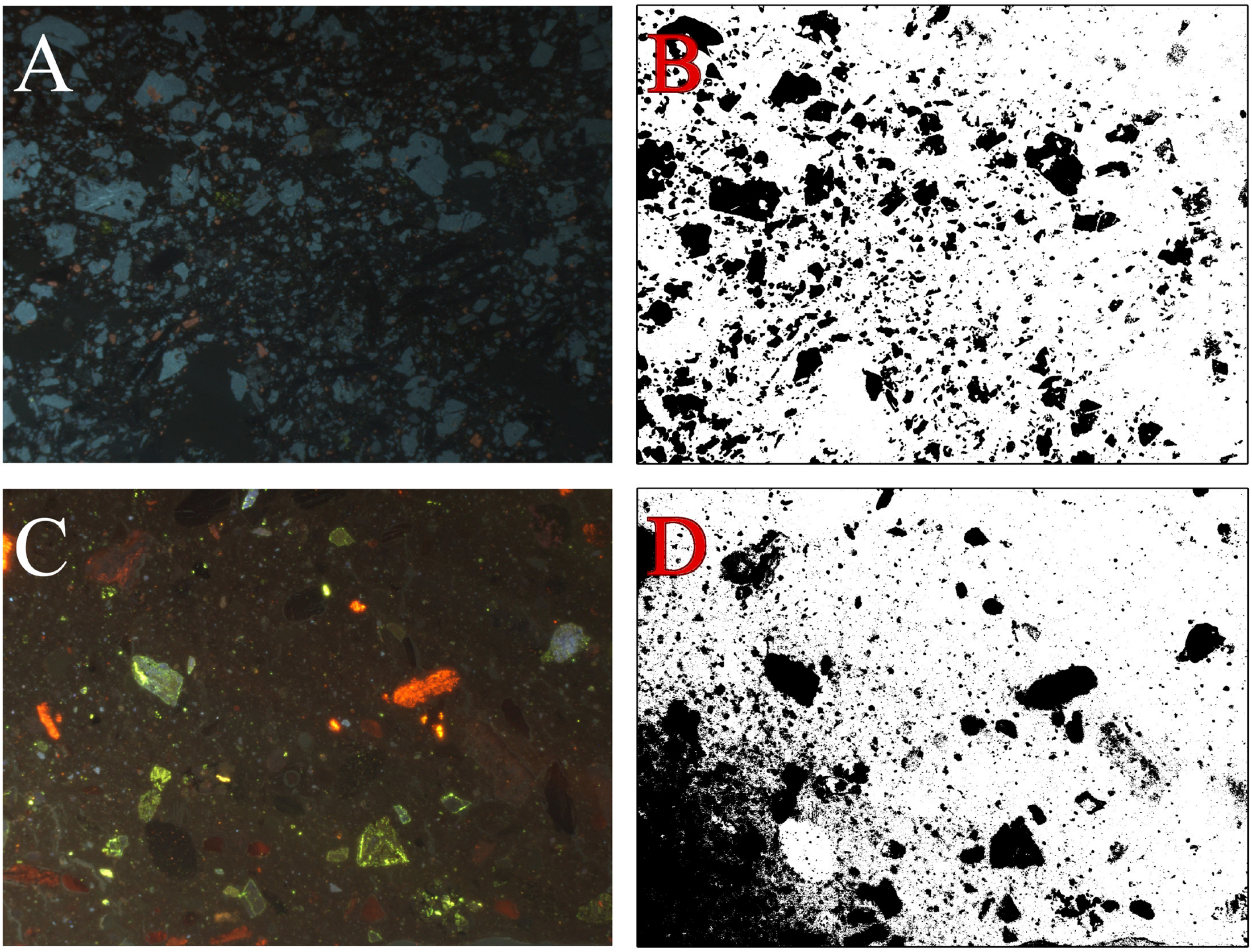

Fig. 1 - Examples of heterogeneous beams in CL images. A and C are as-received images from Odelli et al. (2022). B and D represent attempts to perform thresholds to separate the mineral inclusions of A and C pictures, respectively.

SAMPLES

The developed software has been tested on 412 images from scientific articles (Del Sole et al., 2020; Emami et al., 2021; Odelli et al., 2022). The pictures were acquired using CL microscopes with variable instrumental setting (100-300 µA, 7.5-15 kV, 15-72 s exposure time), and different typologies of surface treatment (polished and unpolished), therefore providing a method assessment related to a wide range of case studies.

METHOD

The images obtained from CL microscopy and collected for this work have been subjected to the developed software. Software operations, which implement the aforementioned corrections and the reasons for their execution will be explained below.

The first operation carried out is the splitting of the image into its components of the HSB colour references system (Joblove & Greenberg, 1978): Hue (H); Saturation (S); Brightness (B). From here on, the program works only with the B channel, except in the last step, in which it is combined again with the two other channels to obtain the starting image with a corrected brightness. The benefit in using B, rather than other parameters for the luminosity description (e.g., lightness L or luminance Y), was found since it corresponds to the light intensity that an observer estimates coming from a given area (Oleari, 2008). Therefore, it was considered the most suitable parameter for an evaluation of instrumental conditions.

Having isolated B, the next step is to apply a Gaussian blur with a sigma value high enough to obliterate all the sample details, to reproduce a representation of the heterogeneous intensity of the incident beam with sufficiently acceptable approximation. For the calculation of this filter, the software uses a sigma computed from a user-defined percentage applied on the minimum image dimension, with a default value of 20%. The starting B channel (Fig. 2A) is then divided by the calculated filter (Fig. 2B), to obtain an image in which the original beam intensity heterogeneity has been mitigated, producing a resulting image in which the beam intensity is homogeneously distributed across the analysed surface (Fig. 2C).

Consecutively, a second correction is applied to the image by default, but giving to the operator the choice to disable it. The purpose of this step is to filter out Poisson noise, a kind of noise present in every image, whose contribution is dominant in the darkest areas of the image, where the measurement error fluctuation is high because of the few numbers of photons acquired by the

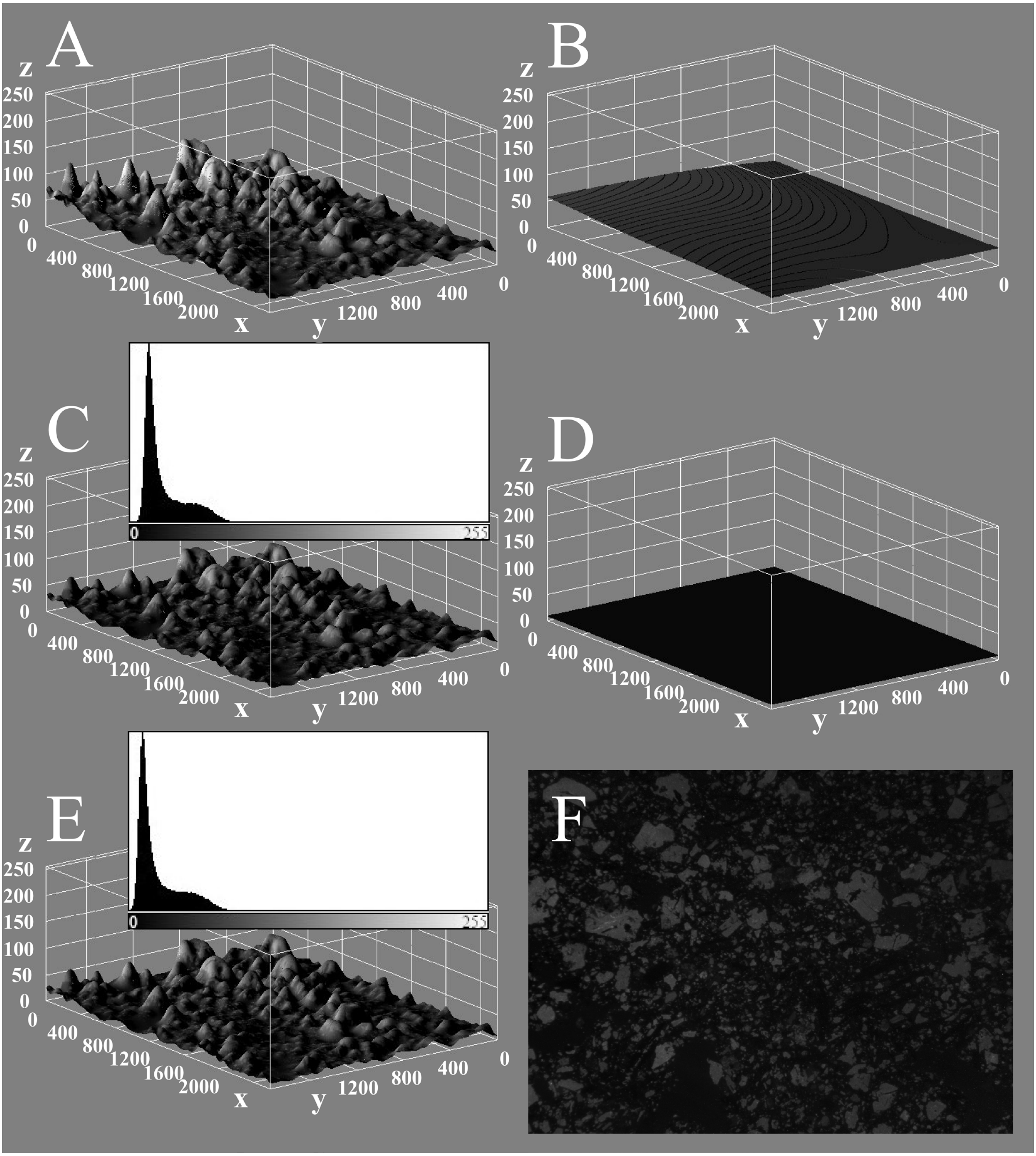

Fig. 2 - Brightness representations of an image from Odelli et al. (2022) collection (also showed in Fig. 1A) processed by IBBIC software. A-E figures show 3D plots in which "x" and "y" are its coordinates (pixel) and "z" represents the brightness values; in particular figures show: A, starting brightness values; B, filter applied for the heterogeneous beam correction; C, result of the first correction and detail of its histogram (0-255); D, filter applied for the correction of overvalued brightness concerning non-luminescent components (the image is nearly-black and flat since it is the constant minimum brightness value to subtract); E, result of the second correction and detail of its histogram (0-255) (notice the histogram is slightly moved to the left, in comparison to C, as D has been subtracted from C). F shows the brightness channel after correction (notice the homogeneous brightness distribution, in comparison with Fig. 1A, where a heterogeneous beam brightness is clearly visible).

instrument. This noise manifests as non-zero brightness values in areas of the image that should theoretically be completely black because they do not respond to the CL stimulation (Marfunin, 1979; Pagel et al., 2000) (Fig. 2C histogram). To correct this behaviour, a baseline zeroing operation is applied, subtracting a very low value (Fig. 2D) from each pixel of the image and clamping the result to 0 (Fig. 2E). To avoid an extra configuration from the user, which would reduce the possibility to apply the transform to a big count of images, this value is computed by blurring each image using a very low sigma and taking the minimum value of the whole resulting image. Ultimately, following this operation, the image correction is finished and, following the recalculation of the image with the new B (Fig. 2F), the approximation of what would have been obtained with a homogeneous electron beam and a zero baseline is returned (Fig. 3).

RESULTS AND DISCUSSION

The purpose of the IBBIC software is to correct the heterogeneously distributed irradiating beam taking into careful consideration the physical context to simulate the results that should be obtained if there were no instrumental error.

Experimentally, it is difficult to compare CL response in sample portions far from each other, especially when attempting to carry out quantitative or semi-quantitative analysis (Odelli et al., 2022). Figure 1 shows the impossibility of performing brightness thresholds for the separation of mineral inclusions due to the heterogeneously distributed electron beam. In the portions of the images where the beam is more concentrated (bottom left) the amount of inclusions is overestimated since the matrix is also counted (Fig. 1D), while in the portions where the electron beam is less intense (upper right), the image is so dark that almost nothing is selected by the threshold (Fig. 1B). Furthermore, even by modifying the threshold parameters, it is impossible to ensure that all image inclusions are taken with a sufficiently acceptable result. As an example, the specimen shown in Fig.1A is relatively homogeneous in its components, therefore all their inclusions should present similar CL results. This problem is resolved by the IBBIC software, as it takes only a few seconds to apply the correction on a large image, simulating its response in the condition of a homogeneously distributed beam (Fig. 3A and Fig. 3C), therefore allowing to set adequate thresholds (Fig. 3B and Fig. 3D).

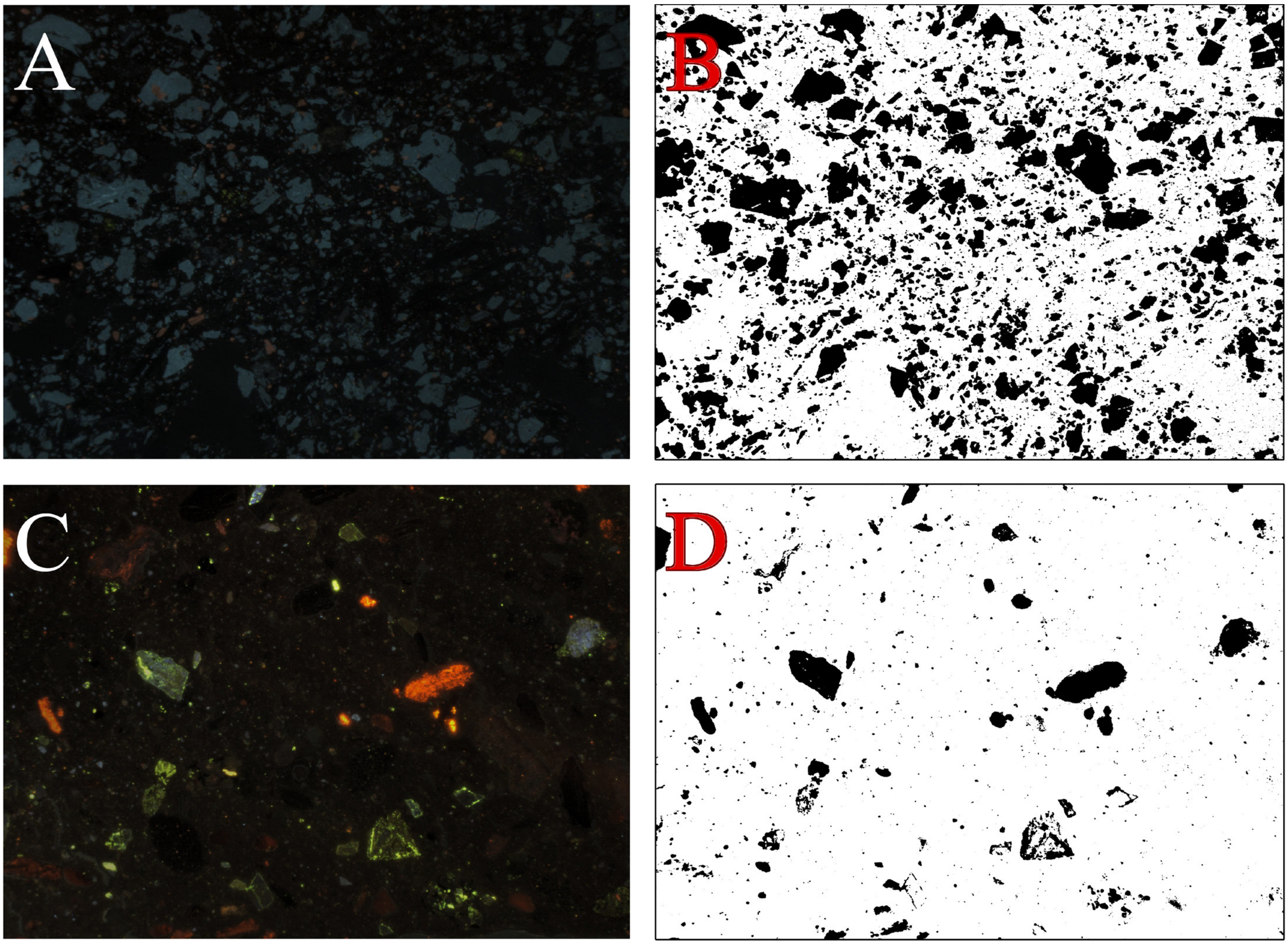

Fig. 3 - Final results of IBBIC software correction. A and C represent how Fig. 1A and Fig. 1C appear at the end of the conversion, respectively. B and D represent attempts to perform thresholds to separate the mineral inclusions of A and C pictures, respectively.

Generally speaking, the quality of the result is highly dependent on the quality of the approximation of the incident beam heterogeneous intensity, which is dependent on a fair compromise in the choice of the sigma value, which should be high enough to dissolve all the sample details, but also small enough to minimise the incident beam intensity gradient flattening. This aspect is one of the most important improvements in methods compared to the version applied in Odelli et al. (2022). In that work the sigma of the Gaussian blur applied was chosen on the basis of each image dimension. Although brightness correction did not alter the images significantly (Odelli et al., 2022), the approach used did not take into consideration their components. As figure 4A shows, big inclusions are able to affect the beam intensity approximation, therefore the choice of a sigma on the basis of the mere image dimensions could lead to artefacts in the areas wherein there are large anomalous portions that can enhance a certain type of contribution. The software developed here allows the observation of both a preview of the beam intensity approximation (Fig. 4B), and of the application of a possible threshold (Fig. 4C), thus evaluating the goodness of the result based on the image investigated. The powerful tool here is that the preview is continuously updated, so the user, while dragging the blur radius percentage slider, can simultaneously observe in real-time how the effect of the inclusions fades away increasing the radius size, and decide to stop when the obtained filter sticks enough to the desired result. Besides, switching in threshold preview mode, it is possible to both continue to tune the blur radius slider as always, and change the threshold value and see how the preview reacts before and after the correction. Dragging the threshold slider, the user can assess in real-time if there is a specific pattern in how the inclusions appear in the image or not. In fact, if the inhomogeneous beam effect is present, while dragging the threshold, the apparition trend will be from one side to another of the image (Fig. 4C on the left). On the contrary, after the correction, the inclusions will appear without a recognizable trend, depending just on their own brightness (image on the right). The possibility to observe in real-time how the inclusions appear while changing the threshold is much more clarifying on the goodness of the chosen filter than a few snapshots taken at specific threshold values, especially when the brightness gradient caused by the inhomogeneous beam has a low variation and the trend is only recognizable in the variation between previews. Consider that figure 4C is an extreme example provided to clearly highlight the problems in threshold operation, but in many samples (e.g., Fig. 1A and Fig. 1B), the problem is still there, but it is not as much as clearly visible in a single snapshot. These tools are extremely useful to avoid accidental formation of artefacts after filter application, and as a consequence greater errors in a quantitative image analysis. With the same purpose, other useful tools added in the IBBIC software are the rescaling of the brightness values between 0 and 255 (enhancing contrasts) and the preview of the final correction result (Fig. 4D).

In order to validate the IBBIC software, the images from three literature works (Del Sole et al., 2020; Emami et al., 2021; Odelli et al., 2022) were processed and their results were compared, evaluating whether the image analysis correction could somehow alter some of the results. The first one was Odelli et al. (2022). From it was possible to process 107 low magnification images from polished and unpolished samples. The aim of that work was to classify ancient pottery based on CL observations regarding petro-fabric and mineral inclusions. For this purpose, with the aid

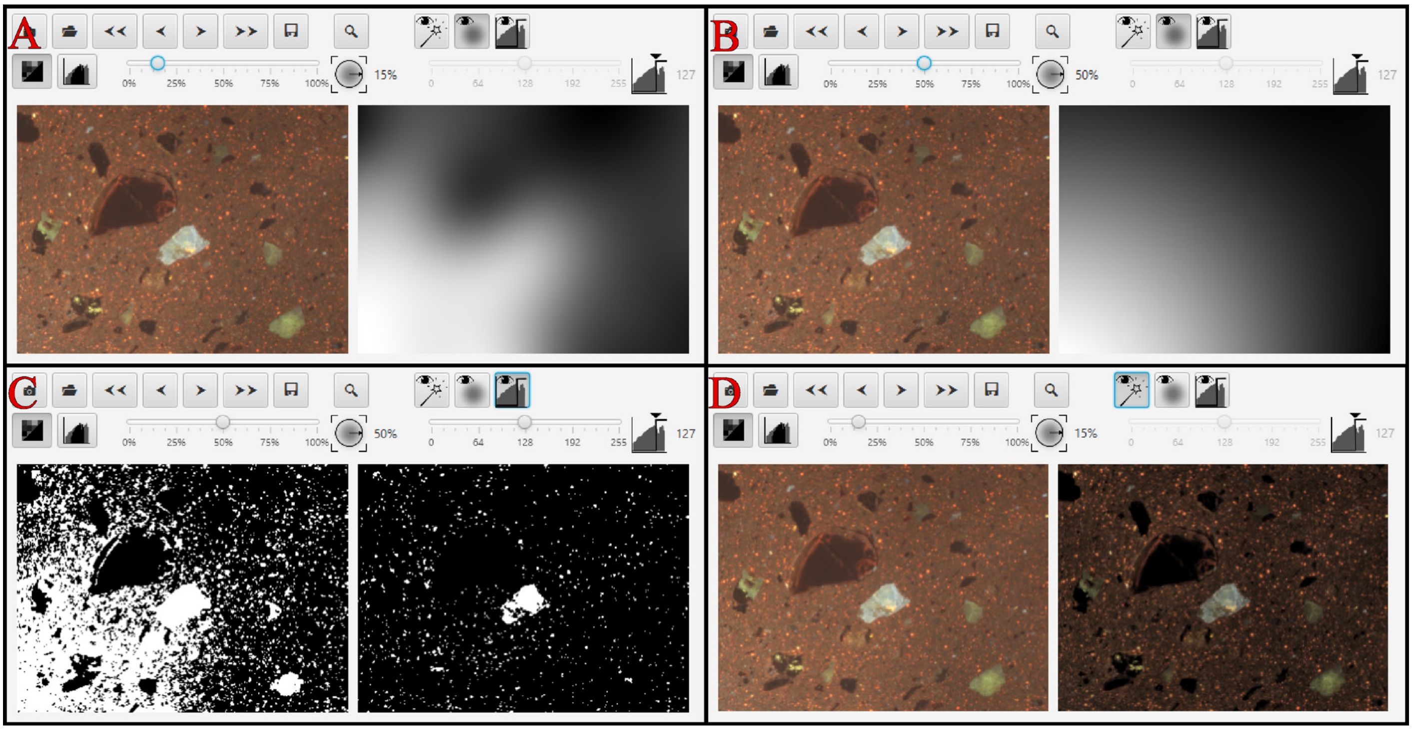

Fig. 4 - IBBIC's previews: A and B shows the analysed image on the left and the preview of the beam intensity approximation; C represents an example of threshold before (left) and after (right) IBBIC execution; D shows on the right the final result of correction on the basis of the chosen parameters.

of an image analysis routine (Cathodoluminescence Image Analysis Color Classification – CIACC) developed by the authors, the images were sorted based on their dominant colouring according to a standard observer.

In this work, the CIACC routine was run on the images before and after IBBIC correction, both with the second correction enabled and disabled, then the percent discrepancy in the images assignment of the dominant colour was evaluated. It has been observed that, following the first correction, the percent discrepancy was 7.5 % and following the second one it was 14 %. This is a consequence of the fact that brightness corrections alter the way in which portions of the image are perceived. The human eye perceives equal brightness changes differently as the hue varies (Smith & Guild, 1931), therefore the deviation between the results is an expected effect and not an error to be resolved. In any case, a minimum of 86% of the images were catalogued in the same way regardless of the type of correction applied. The pictures that were catalogued differently come from samples containing different components or more generally borderline cases (i.e., images straddling the limits of values assigned for classification).

The second work used for the validation of the method was Emami et al. (2021), in which ancient ceramic finds from the Kur River Basin has been investigated. 60 images of 10 different samples produced over a period of approximately 6000 years (seventh–second millennium BCE) were obtained from it. One of the main goals of this latter work was the application of CL as a complementary tool for the study of ancient ceramics and their manufacturing processes. In particular, the interpretation of the CL data was performed on the basis of the emitted colours and their variations, thus being able to recognize the mineralogical phases, the concentrations of trace elements and to interpret the heat-transfer related to manufacturing process.

The application of the software has not made any change able to provide significant deviation from the interpretations made in Emami et al. (2021). For example, in Fig. 5 it is possible to observe the sample Mushki pottery (Fig. 5A) investigated in that work as well as the result of the IBBIC software conversion (Fig. 5B). The elements investigated by the authors were described as: homogeneous matrix with dominant orange CL carbonates; red quartz, yellow unreacted carbonates inclusions and black iron-

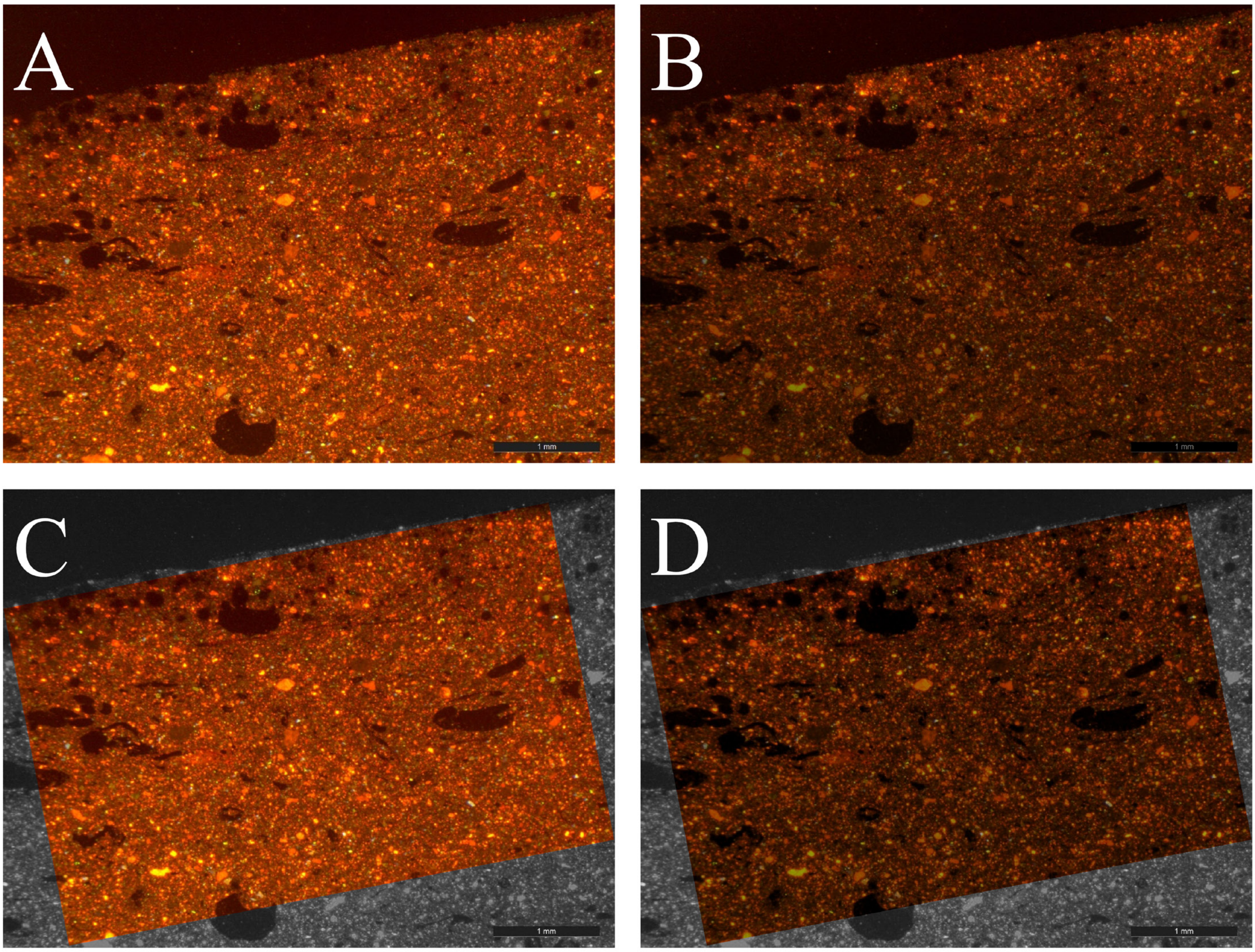

Fig. 5 - A: Mushki pottery sample from Emami et al. (2021). B: A picture result following IBBIC processing. C: crop of a representative portion of Mushki pottery. D: C result following IBBIC processing. Gray scale portions of C and D images have not been taken into consideration for the brightness correction.

rich aggregate (hematite); absence of stratification effects. All these factors are still present following the correction thus making possible an adequate interpretation of the CL results.

However, special attention must be paid to the processed images: large portions not belonging to the analysed sample may affect the Gaussian blur for the correction of the brightness gradient creating brightness artefacts. Therefore, it is important that the user properly sets the radius of the Gaussian blur and eventually removes these portions before processing the image. Another element to take into consideration is that, when the second correction is applied bringing the non-luminescent portions back to brightness values equal to 0, it should be fundamental to analyse the images without scale bars or other added dark elements that can be taken into account for the detection of the minimum. Depending on the type of analysis, an effective choice could be to crop this type of portions that affect errors, carrying out the analysis exclusively on representative portions of the sample, and as a consequence obtaining better results from the brightness correction (Fig. 5D, in which non-luminescent inclusions take on values closer to 0, with respect to what is shown in Fig. 5B).

It should be noted that the two works taken into consideration above use low magnitude images. This is an important aspect since the brightness gradient given by the heterogeneous electron beam is clearly visible in relatively large portions of the analysed surface. As observed in Del Sole et al. (2020), using high magnifications, no appreciable brightness gradients could be found and any brightness corrections might partially obliterate detailed information. It is essential to underline that the IBBIC software is designed as an aid for the study of materials and in no way it can replace the operator. In fact, the latter plays a key role for the correct use of the software, as only its critical spirit is able to avoid the creation of artefacts due to the brightness correction, therefore discerning when and how to use the software. For this reason, during the software-programming phase, it has been voluntarily decided to leave maximum autonomy to the user by choosing which software steps to carry out and how.

Ultimately, it should be claimed that the software, although it was designed to facilitate the study of geomaterials with the CL microscope, is applicable in other fields in which there is the need to correct brightness distributions, thus making it helpful and promising for images pre-processing in many applications.

CONCLUSIONS

CL is extremely helpful in the study of geomaterials, however, with the purpose of performing image analysis, it is afflicted by brightness issues that make it difficult to correctly interpret distant portions of images acquired at low magnitude. The IBBIC software developed in this study can mitigate such artefacts providing images closer to what would have been obtained in their absence. Its application, tested on a large collection of images acquired in CL microscopy using variable instrument settings, did not significantly alter the nature of the images, thus respecting their characteristics. However, following the conversion it was possible to better correlate different portions of images, paving the way for an easier image analysis with quantitative and/or semi-quantitative approaches. The IBBIC software can therefore play an important role in the images pre-processing and it provides maximum autonomy to users in choosing how to apply the parameters. This easy-to-use software also provides multiple tools and previews designed to give real-time evaluation parameters for an adequate brightness correction.

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