THE IMPORTANCE OF LABORATORIES' PARTICIPATION IN INTERLABORATORY COMPARISON. CASE STUDY: INTERLABORATORY TESTS ON ADHESIVES FOR CERAMIC TILES

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The aim of this paper is to present the importance of the laboratories' participation in the interlaboratory comparison (ILC) scheme, using the case study called Interlaboratory Test for adhesive for ceramic tiles. At present, the ILC scheme organized by CEPROCIM is the only one addressed to the laboratories for testing adhesives for ceramic tiles.
The paper aims to present, by using several case studies, the influence of continuous participation at the ILC scheme on the performance improvement of the participants, affecting also the coefficient of variation, as well as the influence brought by the joining of new participants. The case studies represent both accredited laboratories according to EN ISO/IEC 17025 and non-accredited laboratories whose performance (z-score) were assessed during their participation of the all 12 editions of the Interlaboratory Test for adhesive for ceramic tiles.

Lucrarea își propune sa prezinte importanța participării laboratoarelor la scheme de competenta interlaboratoare (ILC), folosind studiul de caz denumit Încercări Interlaboratorare pe adezivi pentru plăci ceramice. În prezent, schema ILC organizată de CEPROCIM este singura adresată laboratoarelor de încercări a adezivilor pentru plăci ceramice.
Lucrarea își propune să prezinte de asemenea, prin utilizarea mai multor studii de caz, influența participării continue la schema ILC asupra îmbunătățirii performanței participanților și a coeficientului de variație, precum și influența pe care noii participanți o aduc. Studiile de caz reprezintă atât laboratoare acreditate conform EN ISO/IEC 17025, cât și laboratoare neacreditate ale căror performanțe (scorul z) au fost evaluate în timpul participării lor la toate cele 12 ediții ale Încercărilor Interlaboratoare pe adeziv pentru plăci ceramice.

Keywords: *Interlaboratory comparison scheme, adhesive for ceramic tiles, initial adhesion strength, tensile adhesion strength after water immersion, open-time, z-score.*

1.Introduction

The Interlaboratory tests on adhesives for ceramic tiles begun in 2007 as the response from the need to meet the requirements of the accreditation standard SR EN ISO/ IEC 17025 [1] to participate in PT/ ILC schemes. At that time in Europe there weren't any institution that organized such a scheme. In this context, CEPROCIM by a small team started organizing a new ILC scheme whose object was a single test - initial adhesion strength according to EN 1348/ EN 12004-2 [2] on adhesive for ceramic tiles. At the first round, only 9 laboratories participated, majority from Romania, and they worked the samples twice.

In 2014, for meeting the needs of the participants, the ILC scheme has been extended at two tests: initial adhesion strength and tensile adhesion strength after water immersion according to EN 1348/ EN 12004-2 [2]. In 2018, at the anniversary of 10 editions of ILC scheme, the scheme was extended again, at three tests: initial adhesion strength, tensile adhesion strength after water immersion and open-time according EN 12004-2.

2.Organizing the interlaboratory test program

The team that organized this scheme for adhesive for ceramic tiles has more than 30 years' experience in organizing ILC schemes.

This vast experience, as well as the participation in other international schemes, allowed the team to organize an ILC scheme for a material for which there weren't enough data at international level to establish a reproducibility of the test results obtained.

At the twelve editions (rounds) of Interlaboratory Tests on Adhesives for ceramic tiles, it's have been participated at least once 67 laboratories from 25 countries from Europe and Asia [3].

Each participant was randomly given a number which is used as laboratory code to enable confidentiality of results. Reference to each

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laboratory in all the general reports is made by its code number. More than that, from one round to the other lab code was changed and there wasn't any connection between lab name and its code allocation system over time.

Through the twelve editions (rounds) of the Interlaboratory Tests on Adhesives for ceramic tiles different types of adhesives produced by manufacturers from Romania were use (i.e. C2FTE, C2TE, etc.).

The test used in all the rounds of the Interlaboratory tests on Adhesives for ceramic tiles for establishing the homogeneity of the adhesive for ceramic tiles was determination of the residue on the 250 μm sieve [3]. For establishing the homogeneity of the adhesives used, after the homogeneity was performed, we took ten samples which were tested in the same day, by the same operator, using the same equipment, every sample have been worked in double.

The samples were considered as being homogeneous when all the results have been placed in the range: mean value of the residue on the 250 μm sieve ± 2*s* (%). The value of *"s"* represents the standard deviation of repeatability.

3.Results and discussions

For the statistical calculation, algorithm A in Annex C from the standard ISO 13528:2015 was applied. This implies an iterative calculation of the robust values for mean and standard deviation based on all the participants' results [4].

Step 1: The initial values for x* and s* were calculated:

$x^* = median\ of\ x_i$, *i = (1, 2, …, p)* (1)

$s^* = 1.483 \cdot median\ of |x_i - x^*|$, *i = (1, 2, …, p)* (2)

Step 2: The values of *x** and *s** was update:

$\delta = 1.5 \cdot s^*$ (3)

Step 3: For each x_i *(i = 1, 2, ..., p),* was calculate:

$$x_i^* = \begin{cases} x^* - \delta, & if\ x_i < x^* - \delta \\ x^* + \delta, & if x_i > x^* + \delta \\ x_i, & otherwise \end{cases} \quad (4)$$

Step 4: The value obtained after the last iteration represents the assigned value *(x_{pt})* chosen to be the consensus value. The values of x_{pt} and σ_{pt} were calculated as follows:

$x_{pt} = \sum x_i^* / p$ (5)

$\sigma_{pt} = 1.134 \cdot \sqrt{\sum (x_i^* - x_{pt})^2 / (p-1)}$ (6)

$CV = \frac{\sigma_{pt}}{x_{pt}} \cdot 100$ (7)

Step 5: The z-score is calculated as follows:

$z_i = \frac{x_i - x_{pt}}{\sigma_{pt}}$ (8)

Step 6: The evaluation of the results was made according to EN ISO/IEC 17043:

- satisfactory, when |z| ≤ 2 (9)
- questionable, when 2 < |z| <3 (10)
- unsatisfactory, when |z| ≥ 3 (11)

Where:

p the number of laboratories that took part at interlaboratory tests;

x_i the result reported by one participant laboratory *i;*

*x** robust average of the results reported by all participant laboratories, calculated according to algorithm A method;

*s** robust standard deviation of the results reported by all participant laboratories, calculated according to algorithm A method;

x_{pt} assigned value (consensus value);

σ_{pt} standard deviation for proficiency assessment;

CV coefficient of variation.

The robust estimates x_{pt} and σ_{pt} is derived by an iterative calculation, by updating the values of *x** and *s** several times using the modified data, until the process converges. Convergence was assumed when there wasn't change from one iteration to the next in the third significant figure of the robust standard deviation and of the equivalent figure in the robust average [4].

For an assessment of how the scheme helped the participating laboratories to improve the way they work and the results obtained, the coefficient of variation was selected and its evolution was followed from one round to another, depending on the number of participants, the history of participation to the scheme and sensitivity of the test. The most common use of the coefficient of variation is to assess the precision of a technique (test, in our case) [5,6].

The coefficient of variation (CV) is a statistical measure of the dispersion of data points in a data series around the mean. It is a useful statistic for comparing the degree of variation from one data series to another, even if the means are drastically different from one another. The higher the coefficient of variation, the greater the level of dispersion around the mean.

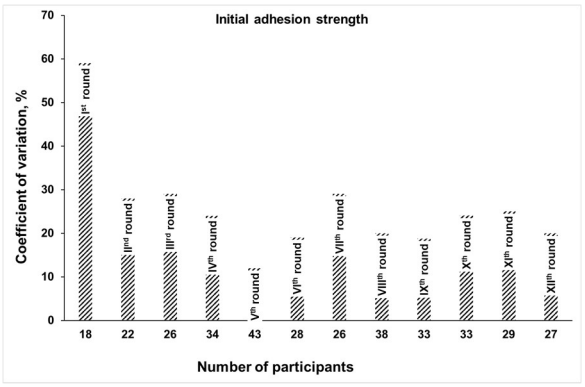
The Figure 1 shows the evolution of the coefficient of variation for the initial adhesion strength and variation for the tensile adhesion strength after water immersion.

It can be seen in Figure 1 that the values of the coefficient of variation don't exceed 50%, except for the first round, which means that the values of the tests obtained by the participants are homogeneous.

Analysing the evolution of coefficient of variation, it can be seen two trends as follow:

a) an improvement from one round to another for the initial adhesion; the biggest drop of the coefficient of variation was about 80 % compared to the first round;

b) in the case of the tensile adhesion strength after water immersion it can't be seen a big improvement of the coefficient of variation from a round to a round; the biggest drop of the coefficient of variation was about 40 % and that compare the 9th round with the 10th round [6].



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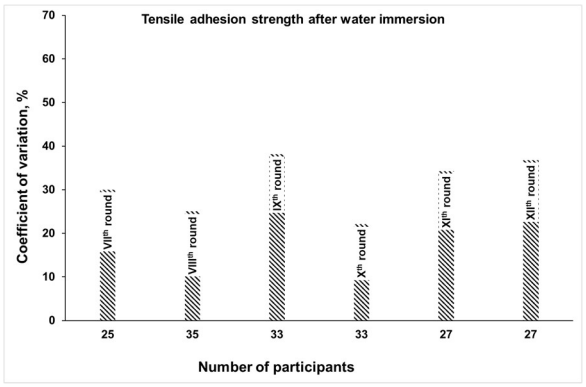

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Fig 1 - The evolution of the coefficient of variation for: a) the initial adhesion strength; b) the tensile adhesion strength after water immersion / *Evoluția coeficientului de variație a: a) aderenței inițiale prin tracțiune; b) aderenței prin tracțiune după imersare în apă*

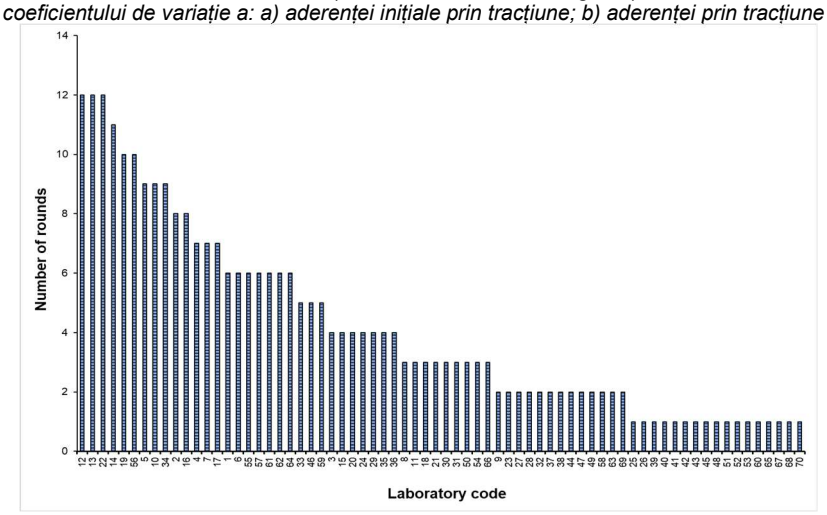

Fig 2 - The participation of the laboratories in the ILC scheme / *Participarea laboratoarelor la schema ILC*

The values of the coefficient of variation, respectively the homogeneity of the obtained results is influenced by several factors, such as: the number of participants in the scheme and the participants' experience both in domain and in the ILC participation.

For understanding the impact of the lack of constant participation of the labs, the provider made a statistical analysis using the data only from the labs that participate in 11th and 12th round of ILC. The number of labs that participated in both rounds were 19 and they obtained a coefficient of variation for initial adhesion of 18.6%, respective 18.9% and for tensile adhesion strength after water immersion the coefficient of variation was 33.5%, respective 34.2%. It can be seen that those labs have been learn from ILC experience and have been worked the samples in the same way. The difference between the coefficients of variation obtained for the two tests are influence by the sensibility of the test given by the low resistance of adhesives to the action of water.

Figure 2 shows the number of laboratories participating in the ILC scheme during the 12 rounds organized so far.

In Figure 2 it can be seen that many laboratories have participated only once or twice to the ILC scheme and only 6 laboratories participated in more than 10 organized rounds.

The reasons for which the laboratories haven't participated constantly in the ILC scheme were various: financial impediments, overcrowding of staff, obtaining a satisfying z-score after one round or on the contrary, not achieving satisfying results etc. But their management should take all measures to overcome them and use the ILC scheme in real assessment of the capability of their own lab.

The Figure 3 shows the distribution of the laboratories that participated in the ILC scheme.

It can easily be noticed form Figure 3 that 43% of all laboratories participated only in 3 rounds or less and less than a quarter participated in more than 7 rounds of the ILC scheme.

Synthetic assessment of the results obtained for each test by the participants at the all rounds of ILC Scheme, on the z-score basis is presented in Figure 4.

Evaluating the results reported for each test by the participating laboratories based on the

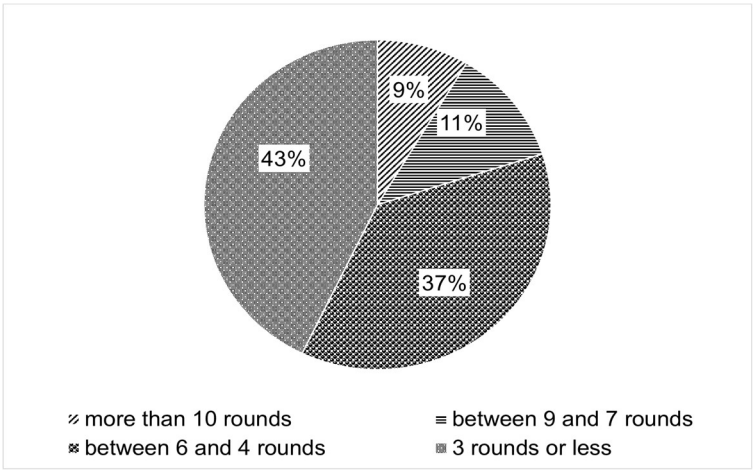

Fig 3 - The distribution of the laboratories that had participated in the ILC scheme
Distribuția laboratoarelor participante la schema ILC

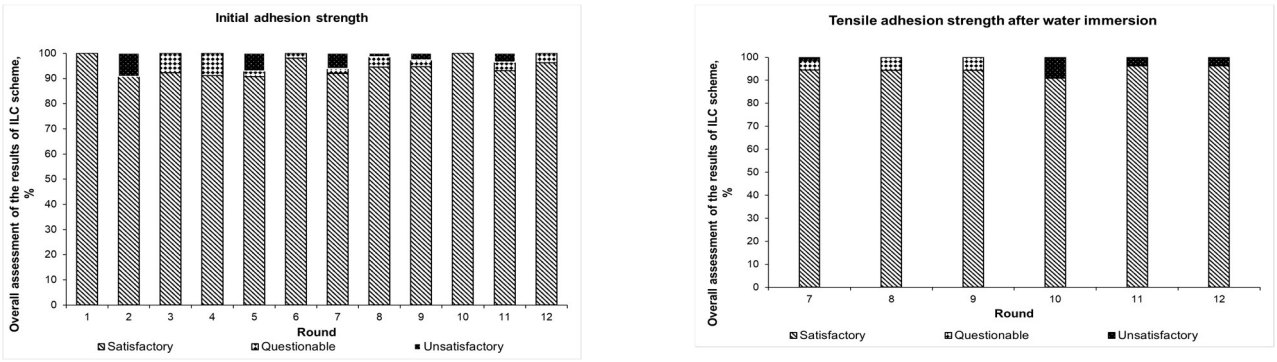

Fig. 4 - Overall assessment of the results of ILC scheme based on z score /
Evaluarea generală a rezultatelor schemei ILC pe baza scorului z

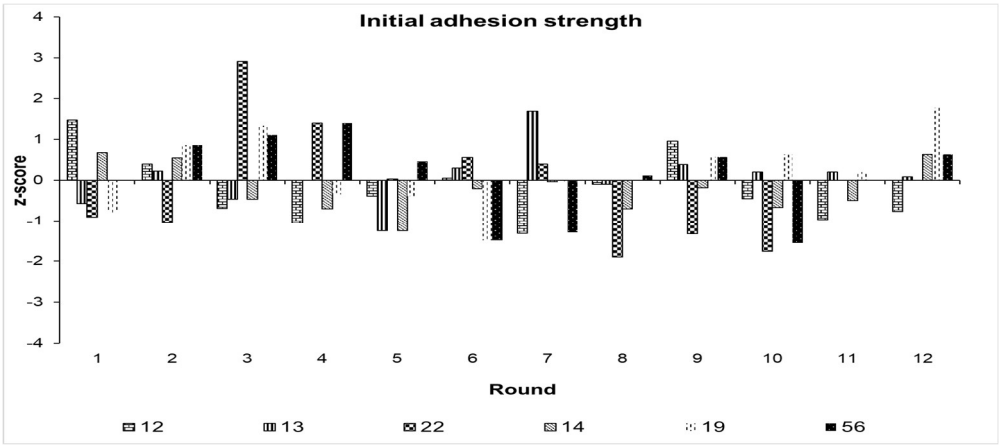

Fig. 5 - The evolution of z-score for the 6 laboratories present at more than 10 rounds of the ILC scheme /
Evoluția scorului z pentru cele 6 laboratoare prezente la mai mult de 10 runde ale schemei ILC

z-score obtained, it can be seen that 94% of them are classified as satisfactory, 3% questionable and 3% unsatisfactory.

The management of a laboratory, whether it is accredited according to EN ISO/IEC 17025 or not, must constantly evaluate its capability, and the participation in an ILC scheme is one of the routes indicated to achieve this.

Figures 5 and 6 shows the evolution of z-score of the 6 laboratories presented at more than 10 rounds of the ILC schemes organized by CEPROCIM: 12 rounds for initial adhesion strength and 6 rounds for tensile adhesion strength after water immersion.

It can be seen that the results are for the most part satisfactory, although there were situations where the value of the z-score was raised, but it wasn't recorded any unsatisfactory results ($|z|\geq 3$), although the others participants varied.

But following the trends of the 6 laboratories, it can be noticed that in the case of the laboratory no 22 (an accredited laboratory) the reported results had quite high scores, there were also recorded

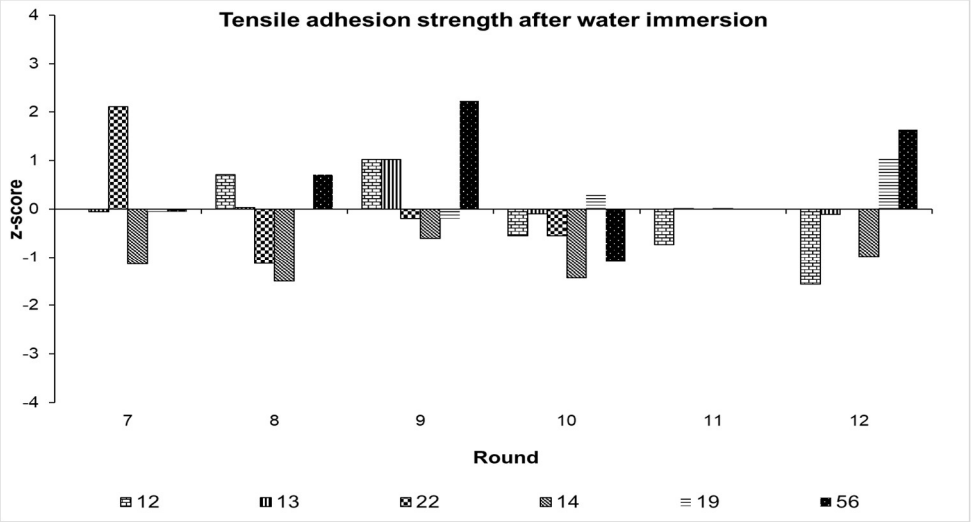

Fig. 6 - The evolution of z-score for the 6 laboratories present at more than 10 rounds of the ILC scheme/
Evoluția scorului z pentru cele 6 laboratoare prezente la mai mult de 10 runde ale schemei ILC

results as questionable. In contrast, in the cases of laboratories no 13 (accredited laboratory) and no 12 (unaccredited laboratory), the reported results were quite close to the average.

Monitoring the evolution of the z-score by the laboratory is very important because it can find the trends that persist over several rounds and it can take the necessary actions if is the case [7]. However, at the laboratory level this situation has to be investigated right from the moment when the first abnormal result was obtained, and the necessary measures should be taken to remediate the situation.

In conclusion, following the evolution of z-scores recorded for the 6 laboratories used as case studies, it can be concluded that only 4 of them, 1 accredited laboratory and 3 unaccredited laboratories, have reached a certain maturity to allow them to provide reliable results for their customers.

4.Conclusions

- Since 2007 CEPROCIM had yearly organized the ILC scheme called Interlaboratory test program on adhesives for ceramic tiles. At all the twelve rounds of Interlaboratory Tests on Adhesives for ceramic tiles, it's have been participated at least once 70 laboratories from 25 countries from Europe and Asia.
- Using the experience accumulated over time in the organization of ILC schemes, the CEPROCIM team succeeded in organizing an ILC scheme that help participants to evaluate their own capabilities and to provide reliable results to their clients.
- Over 94% of the test results provided by the participant laboratories could be framed as "satisfactory" (having $|z| \leq 2$) according to EN ISO/CEI 17043, which proves that overall, the participating laboratories performed well and tend to improve their working procedure.
- The ILC scheme prove to an important tool in evaluation of laboratories competence, showing to the laboratory's management the influence of the working mode, compliance with the microclimate conditions or equipment used.

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