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Impact of Residual Quartz on Lifetime of High Strength Porcelains

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Abstract

The target of the Study was to quantify the quantity and particle size of critical residual content. Quartz particles, as calibrated defects, were intentionally added to C-130 alumina porcelain body laboratory test bars to simulate the effect in laboratory conditions. This allowed us to quantify the influence of quartz on quartz-free porcelain composition. In the initial test series samples with 0%, 1%, 2%, and 4 % of quartz content with 200 µm and 64 µm particle size were prepared. The quartz content was confirmed by a mineralogical analysis of the fired samples. After glazing and firing of the samples, the bending strength was determined by a standard 3-point bending test to get the baseline strength. At the next step, the bars were stress-cycled on a test-bench with 50 % and 80 % of the measured baseline strength to induce cracks propagation by fatigue. Modulus of rupture of stress-cycled samples was compared to baseline strength and the influence of quartz content and quartz grain size was evaluated. The results show that already 1% of residual quartz crystals is reducing the mechanical strength and the reduction is strongly related to the size.

KEYWORDS: High voltage ceramic insulators, residual quartz, lifetime, fatigue aging

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1. Introduction

The lifetime of the high voltage insulators is of more actuality then ever as we are experiencing the biggest electrification campaign since the development of ceramic alumina porcelain insulators in 70's and 80's. The world is now moving to sustainable energy sources, which will deeply impact the energy generation and distribution in the coming years. At the same time the existing transmission lines are reaching an age where the replacement is going to be required, probably sooner than later.

The estimated remaining lifetime of installed insulators has been studied for years, but recently more and more researchers have been studying this question, mostly in close collaboration with the operating energy companies. The KEPCO Research Institute and Sungkyunkwan University in South-Korea have been actively publishing studies of different insulators aging mechanisms since 2019. (1,2,3,4) Insulators aging and replacement strategies have also been studied by an IEEE working group (5).

These studies have defined four main aging mechanisms, a) expansion of the cement, b) corrosion of the metal parts, c) mechanical and electrical stresses on insulators core and d) drying out bitumen. Resistance against the mechanical and electrical stresses is related to the microstructure of the ceramic insulators. This study will be focusing of the ceramic insulator's microstructure and resistance against mechanical stresses.

Today most ceramic high voltage insulators are made of grades of C-120 Alumina Porcelain and C-130 High Strength Alumina Porcelain as specified on the IEC 60672(6). Cristobalite and quartz porcelain are used as insulator materials on the low voltage distribution applications below 36 kV. Curiously the IEC 60672 specifies the grades only by the mechanical strength as new after production. Aging, fatigue resistance and microstructure are not even mentioned at all. This is one reason why these insulators life-time studies are so important today.

The required mechanical strength of C-120 and C-130 can be achieved by various mixes of raw materials, which leads to different microstructures and different lifetime expectations between the manufacturers. The importance of the microstructure and its impact to the lifetime has long been known, and papers often referred to were published by Dr. Johannes Lieberman (7,8). Dr. Lieberman defines the ideal microstructure for the C-130, which should contain ≥ 40 % corundum, ≤ 15 % mullite, and residual quartz content < 1%. The requirement of maximum quartz content is not included in the IEC 60672, but many OEMs have this requirement on their technical specification.

The study by A. Rawat and R. S. Gorur(9) tested 30 years old insulators and they were able to establish clear relations of the lost mechanical and dielectric strength and microstructure. The results confirm Dr. Lieberman's ideal microstructure at the Power Frequency Puncture Test: the failed samples had quartz crystals size > 50 µm and high general quartz content.

Keekeun Kim et al. (10) have demonstrated the role of corundum content as a major element increasing the resistance against aging. They were able establish a model between the corundum content and strength relationship and predicted the tensile strength of 43 years old cap & pin insulators with accuracy of 2.5%.

2. Origin of Quartz on C-130 alumina Porcelain

The quartz in the C-130 (high strength porcelain) should basically be entirely replaced by aluminium oxide (alumina, Al_2O_3). The alumina is partly coming from the fired clay and chamotte and rest is added as purified alumina. Impurified bauxite can replace the pure alumina, but the bauxite brings a lot of impurities, mainly iron and titan oxides, which give a brownish or grey colour to the porcelain which is otherwise white. Further the negative impact of impurities on the strength must then be compensated for by extra alumina oxide content in the fired product. As the

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manufacturers try to minimize the use of alumina, which is the most expensive mineral used in the recipe, there is an increased risk of the residual quartz.

The residual quartz could come from the raw material mix itself, just by the simple fact that alumina content is not high enough. Even when the alumina content is optimized residual quartz might appear when the particle size is too big. The particles don't have time to melt to the glass-matrix when they are too big. The particle size can be too big, because the milling time is short, or has been shortened for cost savings.

Another reason for big particles could be an intentionally larger percentage of coarse grain added to the body. The coarser grains fraction, up to 4 %, is needed for homogenous drying of the ceramic body in the plastic process. The internal moisture level must absolutely be below 1 % before firing, otherwise the piece might explode. The captured moisture transforms to vapour in the beginning of the process and does not have to escape so remains captured in the porosity; when the temperature rises the vapour pressure also rises and the ceramic body explodes. The isostatic process, or dry process, does not require a long drying step and the particle size is smaller than in the plastic process, which makes it easier to control the residual quartz content.

Finally, the residual quartz might come from an inapt firing curve. The curve might be too short for melting the quartz particles. On the other hand, the firing temperature should be as low as possible so the time cannot be compensated for higher temperature. Then the cooling should be fast above 1000°C and the slower at the quartz ($\beta \rightarrow \alpha$) transformation zone. All these parameters can have an impact on the presence of residual quartz and its grain size.

Liebermann has presented (7) a priority list of measures to reduce the residual quartz:

1. Reduction of quartz in the recipe
2. Formation of early eutectic melt phases
3. Increasing the alumina content to >60%
4. Low firing temperature
5. Fast cooling above 1000 °C.

Gunter Fassbinder(11) explains that the great disadvantage of the quartz is the ($\beta \rightarrow \alpha$) transition at + 573°C, which is causes about 0,7 vol% volume shrinkage during the cooling curve (12). At this temperature the glass melt resulting from the feldspar cannot follow the shrinkage of the quartz grain. The effect is a high radial tension on the quartz grain and a tangential pressure on the surrounding glassy phase (13, 14, 15). The material can support this if the grains are small enough, but the large grains will not withstand the tension and will develop microcracks (16), which then reduces the porcelain strength.

The existence of the microcracks is known to reduce the resistance against the aging especially when the insulators are subjected to cyclic stresses, both tension and bending, such as on the overhead lines, on circuit-breakers, or high insulator columns exposed to strong wind loads. During cyclic stresses the microcracks starts to propagate and the insulators and slowly loose its strength. In applications where the post insulators are only under permanent compression, there is no significant crack propagation.

3. The sample preparation and experimental method

The goal of this experiment was to prepare "calibrated defects" in the test-bar to study their impact in laboratory conditions. In this study the defect was the quartz crystals , which were added to the body in controlled size and quantity. Manufacturing "calibrated defects" is a classical Root Cause Analyses (RCA) method used in modern Quality Management. No reference of such studies with ceramic insulators was not found, but it was decided to advance by "trial and error" to find if this would work.

Pure quartz sand is available on the market in different grain sizes, and it can be sieved to separate different fractions. In this study quartz grains with a size of 200 µm and 64 µm were chosen. 200 µm was existing on the market and

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could be used as it is, further the purpose was to make sure that a measurable effect was achieved within a reasonable time frame. The 64 µm fraction was sieved and selected because this fraction is close to the A. Rawat and R. S. Gorur findings, that samples with > 50 µm quartz failed on punctuation (9). Fassbinder (11) proposes that the optimum strength is achieved when the residual quartz is about 20 µm. The normal insulators body from PPC Insulators Cab plant (Slovakia) contains about 0.5 % - 0.8 % residual quartz, which is at the limit of the measurability. Therefore, we can presume that these quartz particles are below the size of 20 µm.

The base-material was the standard industrial C-130 granulate used for the manufacturing post insulators in PPC Insulators at Cab. The basic recipe is:

- Feldspar 26 %
- Chamotte 20 %
- Clay 20 %
- Alumina 34 %

The materials are milled on the classical wet milling process down to average 5 µm particle size. After milling the slurry goes through fine sieves to remove coarse dirt and strong magnets to eliminate all potential iron impurities. The next step is the spray drying process where the slurry is sprayed directly into hot air, and instantly dry granulate drops to the transport belt where it moves to the silos for storage until loading into the iso-static press. The body used for this study was collected at this stage of the process.

The quartz particles were mixed into the base-body in the wet laboratory mill by 1 wt.%, 2 wt. % and 4 wt.%. Test-bars with 0 wt.% added quartz were made of the same batch of body to get the baseline. The body was pressed in the laboratory (Fig. 1) and extruded with the laboratory extruder (Fig. 2) to a 10 mm diameter and 150 mm long test-bars.

Fig 1. The laboratory filter-press

Fig 2. The laboratory extruder.

After drying the bars were manually glazed by dipping (Fig. 3). After glazing the samples were fired in the normal production kiln (Fig. 4). On the Figure 5 we see a ready fired Test bar.

Fig 3. Manual Glazing

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Fig 4. Samples ready on kiln for firing.

The 3 point-bending machine (Fig. 6) was used to break and submit the samples under cyclic loading. The sampling size of each test was defined as 10 pieces. First the base-strength was tested at 3-point bending machine, then new samples were cycled with 50 % of the measured breakage strength and after cycling they were loaded to breakage. As there was no references of how many cycles the bars could support it was decided to start with 100 and 500 cycles. (Fig. 7)

After the breakage the test-bars were inspected to be sure that the breakage was not caused by an external defect on the glazing or impurities on the body.

Fig 5. Ready fired test bar.

Fig 6. 3-point bending machine.

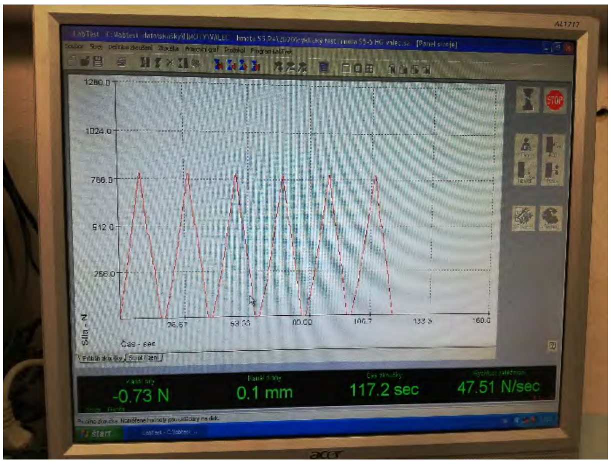

Fig 7. Cycling curve

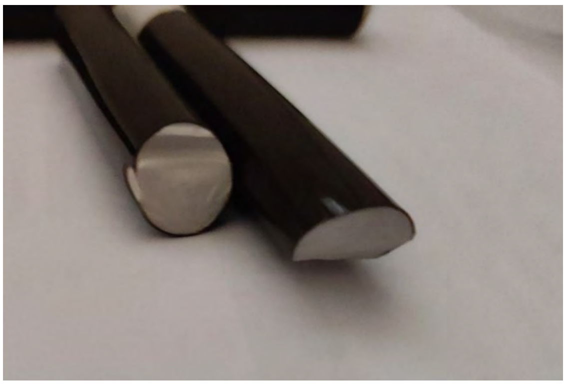

Fig 8. Broken test bar at 3-point bending test.

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4. Results

10 samples of two test series with 64 µm and 200 µm quartz particles were broken at the 3-point bending test-machine without cycling. The results are on the table 1.

Quartz		Rmo (MPa)	Standard deviation	Weibull modulus
64 µm	0%	194.45	7.06	30.7
	1%	184.56	10.00	20.6
	2%	182.70	5.32	37.4
	4%	179.07	4.10	48.1
200 µm	0%	192.44	4.74	44.7
	1%	135.01	13.67	10.4
	2%	132.25	12.41	11.5
	4%	113.57	3.16	36.2

Table 1: Base line results

The first observation here was that the drop of the mechanical strength was visible already at 1 wt.% of added quartz. Increasing the quartz further didn't have the same impact, apart from the samples of 4 wt.% of 200 µm quartz particles, where another drop was observed. The Weibull modulus was constant as it should be with this type of ceramic material.

There was no published data or internal indications regarding what combination of force and cycle quantity would be the optimal to simulate the crack propagation on C-130 ceramic test bars. The limitation factor is that the 3-point bending cycling does not allow high-speed cycling the maximum cycles would be c. 200 cycles per day on a normal working day, as the equipment requires an operator presence during the testing. Thus, to be able to calculate the Weibull modulus and have confidence in the repeatability it was decided that a set of 10 bars would be used for every test.

The first cycling was selected to be 50 % and 80 % of the measured average breakage force for 100 cycles for the 200 µm quartz particle size test series. The reasoning was that the observed loss of strength of 30 % caused by the quartz suggested that the crack propagation, could be measurable already with such low forces and low cycle quantity.

These were the arguments for why it was not decided to start with the approach of cycling to the failure.

On the figure 9 we can see the results of the first two cycling series.

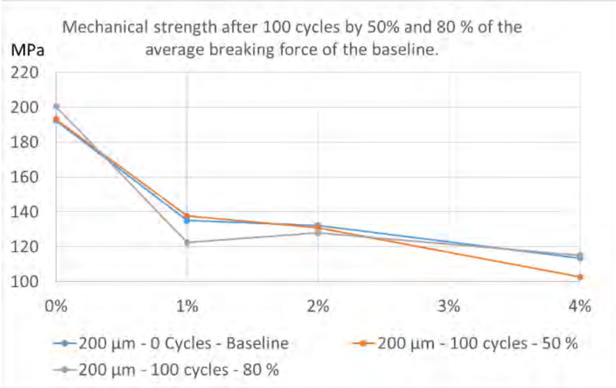

Fig. 9: Mechanical strength after 100 cycles by 50% and 80 % of the average breaking force of the baseline.

We can see from the figure 9, that there is not significantly difference between graphs. The conclusion was that the quantity of cycles was too low to cause measurable crack-growth. Then the similitude of the curves with high Weibull modulus is a clear indication that there was only one failure mode at the breakage. This means the base material was homogenous, which allows the elimination of failures caused by impurities or non-homogeneous structure. Therefore, when there is a measurable difference, it would be caused only by the added quartz particles.

The cycling was increased to 500 cycles. The results are on the figure 10.

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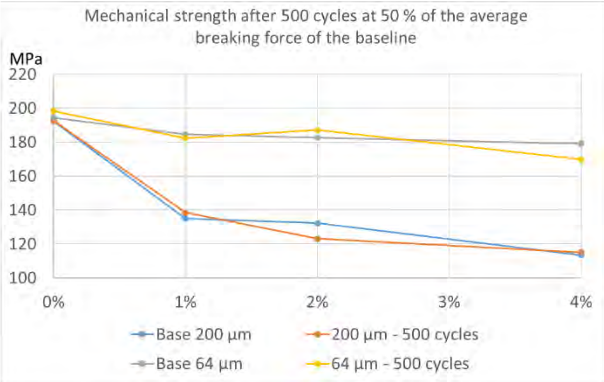

Fig. 10: Mechanical strength after 500 cycles at 50 % of the average base-line breaking force.

The increased cycling was not enough to cause measurable crack-growth with the applied force.

Table 2. The mineralogical analysis was executed in an external laboratory by RFA/XRD-Method

Quartz		Corun-dum (%)	Quartz (%)	Mullite (%)	Glass Phase SiO_2 (%)
64 µm	0%	32.56	0.89	19.7	46.8
	1%	32.73	0.63	19.5	47.1
	2%	31.00	0.71	19.3	49.0
	4%	32.16	1.68	20.0	46.2
200 µm	0%	34.19	0.86	19.6	45.3
	1%	33.17	1.17	19.2	46.4
	2%	31.84	1.75	19.0	47.4
	4%	32.09	3.11	18.9	45.9

Table 2: The mineralogical analyses.

The breakage surface was observed by a light microscope. The quartz grains were easy to identify on an otherwise smooth surface.

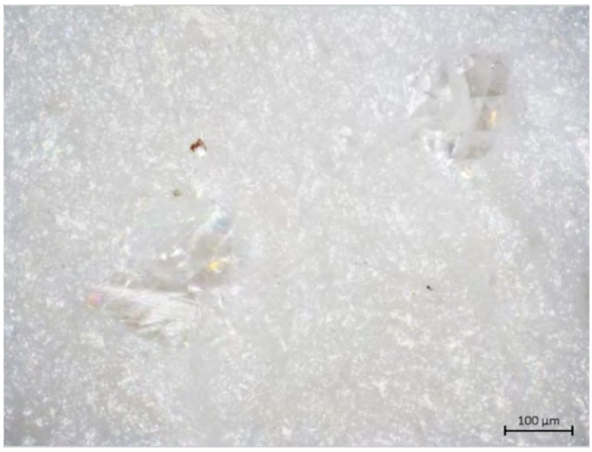

Fig 11: two 200 µm quartz crystals on

Fig.12: one 64 µm quartz crystals on breakage surface breakage surface.

It was decided to increase the cycling quantity to 30’000 with 4 % - 200 µm sample at 80 % of the nominal breaking force to see if clear crack propagation could be observed.

The sample did not break during the cycling. After 30’000 cycles the mechanical breaking force was same level then it was at after 100 and 500 cycles.

A dye penetration test was executed to reveal the potential cracks on the brakeage surface.

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Fig.13: Breakage surface after 30’000 cycles. The dye penetration test reveals the quartz particles clearly as the liquid penetrates the microcracks of the crystal. Anyhow there is no visible crack propagation on the base material.

5. Discussion and Conclusions

The baseline of the 3-point bending tests results allows us to immediately to confirm the important impact of quartz on the mechanical strength caused by as little as 1 % of quartz in the mineralogical structure. This is fully in line with the Lieberman $_{(7,\ 8)}$ studies on the impact of the quartz content.

Further we can see the importance difference between 64 µm and 200 µm particle size. The size was known as a critical factor, but the critical limit is less evident as there are only a few papers referring to the quartz particle size. A. Rawat and R. S. Gorur $_{(9)}$ showed that large size > 50 µm quartz particles did reduce the strength, while Fassbinder$_{(11)}$ proposes that the size should be below 20 µm.

The D_{50} particle size of the C-130 body is directly related to raw-materials and the manufacturers milling process. It is clear that, 200 µm particles are rare on the plastic ceramic body, but they are not excluded as a residual fraction of the grain size distribution. This risk of 200 µm particles is basically eliminated on the isostatic process where the spray drying allows excellent grain size distribution. The 64 µm particles are already omnipresent in any manufacturing process.

There is an interesting observation on the mineralogical analyses of the samples. The 200 µm size particles were increasing the quartz content proportionally to the added quartz content. The 64 µm samples didn’t show such a direct relationship. The samples 1 wt.% and 2 wt.% quartz showed similar quartz content to the base-material with 0 wt.% added quartz. Only 4 wt.% quartz addition was enough to show an increase of quartz above 1 wt.% after firing. This suggest that some of the 64 µm particles were melted to the glaze-phase during the firing process. In any case, the residual quartz particles were big enough to cause a loss of strength compared to the base material.

The test bars with 0 wt.% added quartz show about 0.8 % quartz on the XRD analyses. This is the real residual quartz. Knowing that the particle size D50 after milling in isostatic process is 5 µm we can presume that the residual quartz is systematically below 20 µm and therefore harmless.

One major conclusion is that the preparation of “calibrated defects” to study their impact on C-130 ceramic for insulators does work. The results are repetitive, and the base-material is sufficiently homogenous to eliminate multiple failure mechanisms at the 3-point bending test. This is an important finding and opens doors for follow-up studies.

At the beginning of the study there was no indication what the optimal cycling frequency and force would be. The cycling of the test bars by 100 and 500 times with 50 % and 80 % of the average strength was not enough to cause a measurable crack propagation. On real life the stress cycles counts from tenths of thousands to hundreds of thousands or more. But most of the cycling forces are probably below 50 % of the average strength and 80 % would present extreme conditions, for example in a tropical storm.

This underlines the role of residual quartz, when the particles are big enough, as a latent defect,

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not a defect causing early childhood failures, but a defect which can shorten the excepted lifetime by many years. A. Rawat and R. S. Gorur (9) showed in their work how the deterioration of mechanical and electrical strength occurs at a later stage - after 10 years in service.

In this study we were not able to observe systematic crack propagation on the base material. Jan Schulte-Fischedick et al. (17) analysed in their LeKI project in 2019 220 old insulators, which were been service from 40 to 55 years. Jan Schulte-Fischedick is focusing on the crack propagation and aging mechanism of the C-130 long rods insulators. He explains that fracture mechanism describes the super- critical crack growth causing immediate failure, but in contrast sub- critical crack growth lends to gradual growth and this is thus the mechanism of mechanical aging of brittle materials.

With the use of stress intensity factors K_I the principal course of the sub-critical crack growth can be visualised by drawing the crack growth speed as function K_I in double logarithmic diagram, see Fig. 14.

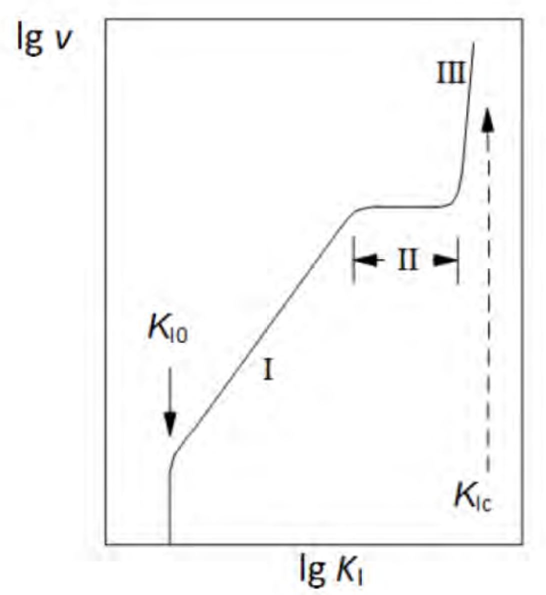

Fig.14: Schematic sub- critical crack growth development.

In most cases K_I must pass a threshold K_{I0} to initiate crack growth, in the region I the crack speed obeys exponential equation, in the cases where region II exist, a constant crack speed can be observed. Finally in the region III the transition to super- critical crack growth and complete failure occurs. In the case of ceramics regions II and III takes only a negligible time, so region I is relevant for the lifetime prediction.

In the conclusion Jan Schulte-Fischedick et al. state that alumina-based porcelain is mechanically aging, however, in a non-linear way. Some insulators didn't show any aging at all, while others exhibited severe aging. This demonstrates the effect of the initial crack length to the excepted lifetime.

This statement is aligned with Fassbinder's and A. Rawat and R. S. Gorur papers about the critical size of the quartz grain. The grain size, as a non-homogenous zone, is more critical to the crack propagation than the quartz quantity is self. This can be applied to other impurities: any foreign particles reaching the sub-critical size will impact the crack propagation and excepted lifetime by mechanical aging.

BIBLIOGRAPHY

1. Taeyong Kim, Youn-Jung Lee, Simpy Sanyal, Jung-Wook Woo, In-Hyuk Choi and Junsin Yi: "Mechanism of Corrosion in Porcelain Insulators and Its Effect on the Lifetime". Applied Science January 2020.
2. Simpy Sanyal, Taeyong Kim, Chang-Sung Seok, Junsin Yi, Ja-Bin Koo, Ju-Am Son and In-Hyuk Choi: "Replacement Strategy of Insulators Established by Probability of Failure". Energies, Volume 13, Issue 8, April 2020.
3. Simpy Sanyal, Fawad Aslam, Taeyong Kim1, Seongho Jeon, Youn�Jung Lee, Junsin Yi, In�Hyuk Choi, Ju�Am Son, Ja�Bin Koo: "Deterioration of Porcelain Insulators Utilized in Overhead Transmission Lines: A Review", Transactions on Electrical and Electronic

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Materials volume 21, pages16–21, December 2019.

4. Taeyong Kim, Simpy Sanyal, Ja-Bin Koo, Ju-Am Son, In-Hyuk Choi and Junsin Yi: “Analysis of Long-Term Deterioration Characteristics of High Voltage Insulators”, Applied Science, Volume 10, Issue 1, December 2019.

5. IEEE Overhead Lines Subcommittee, WG on Insulators performance and Applications; E. A Cherney, A. C. Baker, J. Kuffel. Z. Lodi, A. Phillips, D. G. Powell, and G. A. Stewart: “Evaluation of and Replacement Strategies for Aged High Voltage Porcelain Suspension-Type Insulators.”; IEEE Transactions on Power Delivery, Vol 29, No 1, February 2014

6. IEC 60672:1995 “Ceramic and glass insulating materials”

7. J. Lieberman, “Avoiding Quartz in Alumina Porcelain for High Voltage Insulators. American Ceramic Society Bulletin, Vol. 80 No 6, July 2001

8. J. Lieberman, “Reliability of materials for High Voltage Insulators. American Ceramic Society Bulletin, Vol. 79, No 5, May 2000.

9. A. Rawat and R. S. Gorur: “Microstructure Based Evaluation of Field Aged and New Porcelain Suspension Insulators”. IEEE Transactions on Dielectrics and Electrical Insulation (Volume: 16, Issue: 1, February 2009)

10. Keekeun Kim, Byungwoo Moon, Damhyun Kim, Kibum Park, Chang-Sung Seok, Taeyong Kim, Junsin Yi, In-Hyuk Choi: “Mechanical property evaluation according to alumina content of aged porcelain insulator”. Journal of Material Science and Technology, Volume 9, Issue 5, October 2020.

11. Gunter Fassbinder: “A new Ceramic Body Concept for High Strength HV Insulators”, Materials Science, August 2002.

12. ŠTUBŇA, Igor – TRNÍK, Anton – VOZÁR, Libor: “Thermomechanical analysis of quartz porcelain in temperature cycles”. Ceramics International. 2007, 33(7):1287-1291.

13. Weyl, D.: ”Über den Einfluss innerer Spannungen auf das Gefüge und die mechanische Festigkeit des Porzellans“, Ber DKG 36, 319-324 (1959)

14. Schüller, k.-H.: “Hochfeste Porzellane auf Quarzt- und Cristobalibasis, Teil 1-3“, Ber DKG 44, (1967)

15. ŠTUBŇA, Igor – MÁNIK, Marek – HÚLAN, Tomáš – TRNÍK, Anton: “Development of Stress on Quartz Grain in Illite Ceramics During Cooling Stage of Firing”. Journal of the Ceramic Society of Japan. 2020, 128(3):117–123.

16. CHMELÍK, František – TRNÍK, Anton – ŠTUBŇA, Igor – PEŠIČKA, Josef: “Creation of microcracks in porcelain during firing.” Journal of the European Ceramics Society. 2011, 31(13):2205–2209

17. Jan Schulte-Fischedick, Prashant Siingh, Fabian Lehretz, Pascal Hettich, Claudia Bucharsky, Günter Schell, Michael J. Hoffmann, Wolfgang Marthen, Heinrich Wekenborg, Heinrich Pohlmann: “Materials based lifetime assessment of porcelain insulators.” Proceedings of the 21st International Symposium of High Voltage Engineering (ISH 2019), LNEE 599, pp, 1-12, 2020 Springer Nature Switzerland AG.