

Turkish Journal of Electrical Engineering & Computer Sciences

http://journals.tubitak.gov.tr/elektrik/

Turk J Elec Eng & Comp Sci (2022) 30: 1492 - 1505© TÜBİTAK doi:10.55730/1300-0632.3861

Research Article

Evaluating the role of carbon quantum dots covered silica nanofillers on the partial discharge performance of transformer insulation

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Received: 08.11.2021	•	Accepted/Published Online: 11.01.2022	•	Final Version: 31.05.2022
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Abstract: The article presents the experimental results on the role of carbon quantum dots (CQD) covered silica nanofillers on the partial discharge (PD) properties of transformer oil insulation. The improvement in PD performance of nanofiller blend oil is tested with increased voltage gradient and nanofiller concentration. PD of nanoblend oils for various concentrations of modified silica ranging from 0 to 0.1% wt was measured. PD activity of the test samples is simulated in the laboratory with needle, rod and plane electrode geometry combinations. The facets of PD signals such as PD magnitude, PD inception and time duration of PD extracted from phase-resolved partial discharge patterns were analyzed. Detailed time-frequency domain evaluation of PD pulse is carried out. The enclosure of CQD- SiO_2 nanostructures has considerably improved the inception voltage up to 23% than base transformer oil and SiO_2 nanofluid. Magnitude of discharge is noticeably reduced by the influence of CQD-treated SiO_2 nanoparticles in mineral oil irrespective of the electrode geometry.

Key words: Partial discharge, mineral oil, inception voltage, carbon quantum dots, Weibull distribution

1. Introduction

Partial discharge is vital in encouraging the aging and deterioration of insulation systems, which ultimately ends in power transformer failures [1]. PD detection in high voltage components is a powerful diagnostic tool to estimate the insulation life of transformers. Liable on the cause involved, PD can exist in a preexisting gas phase (bubbles, cavities), or in the insulating liquid medium [2]. This bubble theory is referred to explain the mechanism of breakdown in insulating liquids by Krasucki [3]. Phase-resolved partial discharge (PRPD) patterns, partial discharge inception voltage (PDIV) and PD magnitude are most important criterion for the primary identification of degraded insulation systems and the valuation of industrial product quality [4].

Research works have demonstrated that the significance of PD by streamer initiation and propagation in transformer oil can be examined by ultrahigh frequency (300 MHz to 3 GHz) oscilloscope [5]. The time-frequency analysis of PD measurement system can disclose PD pulse shape relating to charge carrier movement. The PD pulse shape is inferred by the equivalent time length of PD pulse and its magnitude. Electric utility research works have shown that nanofluids of mineral oil are performing better with improved dielectric characteristics [6–12]. Meanwhile, the new findings about the properties of carbon quantum dots (CQD) [13–15] make further interest in using it for electrical insulation applications. Carbon quantum dots (CQD) particles (size < 10 nm)

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and have surface passivation to inorganic substances. CQD can be effortlessly manageable by their particle size and functional group. Only a few research reports are available about using CQD on the surface of silica nanostructures resulting in governable hydrophilic-hydrophobic surfaces of silica nanostructures. Research works have reported that the surface-modified silica nanostructures with carbon quantum dots can improve the dielectric properties of transformer oil such as AC breakdown strength, dissipation factor, dielectric constant and lightning impulse withstand strength [13]. CQD treated nanofillers not only in mineral oil but also in biodegradable oils have influenced the insulation properties [15]. These results suggested that usage of CQD over the surface of silica nanostructures can improve the uniform dispersion of nanomaterials in the transformer oil which eventually results in improved insulation characteristics. Hence, the present work recommends the use of inorganic SiO_2 nanostructures with organic carbon quantum material to ease uniform dispersion of nanomaterials in insulating transformer oil. The results are presented to show the effect of CQD modified SiO_2 nanostructures on partial discharge characteristics of transformer oil nanofluid at various mass fractions.

Some research reports say that the characteristic features of PD intensely rely on experimental test condition such as electrode geometry, magnitude of applied electric potential, nanofluid properties and its purity [16]. The test electrode structure and shape have a very significant role for PD inception voltage assessment. It is related with the existing applied electric potential around the electrode surface/tip. As the highest electric field is found at the tip of needle electrode, it is generally accepted for PD inception voltage evaluation [17]. As of now, there is no detailed report regarding the effect of CQD modified silica nanofillers on PD activity with respect to different electrode configurations such as needle and plane electrodes. Hence, in the work reported, the outcome of different electrode and mass fraction of nanofiller has been analyzed. The nanofluid preparation and test procedures are discussed in Section 2. The result outcomes and the effect of adding CQD treated silica nanofillers for PD characteristic improvements are discussed in Section 3.

2. Experimental procedure

The purification of base oil and nanofluid preparation is carried out as discussed below.

2.1. Nanofluid preparation

Charcoal is weighed in a round bottom flask, the quantified solvent mixer is added to the flask and heated at 75– 90 °C with constant stirring. The obtained solution is diluted in the mixer of water and solvent, which results in two layers in the sample flask. The organic layer was collected and washed 2–3 times in the solvent, which gives carbon quantum dots solution. Surface treatment of SiO_2 is done by adding nanospheres of SiO_2 nanoparticle (10–30 nm size, 2.5 g/mL density, 3.9 relative permittivity and 99.5% purity) and CQD solution in a flask. The solvent is added to the sample and stirred for about 3 h at room temperature. The sample is filtered and dried at room temperature at normal atmospheric conditions. After the solvent gets evaporated, the CQD coated SiO_2 will settle down which is the required nanoparticles. Figures 1a and 1b portray field-emission scanning electron microscopic (FESEM) pictures of SiO_2 and CQD modified SiO_2 materials respectively. Recognizing any discrete differences in size and morphology of silica nanostructures and the same without CQD coverage is a difficult process (because of the infinitesimally small CQD size which is 2.5–5.0 nm). Presence of CQD coverage around the SiO_2 nanostructure surface is confirmed by the elemental mapping in FESEM as shown in Figure 1c. The identification of carbon in along with silicon, oxygen elements endorses the presence of CQD in SiO_2 nanostructures. The moisture level of transformer oil and nanofluid were recorded at room temperature of 27–32 °C with a Karl Fischer moisture measurement and was to be in the range of 10–15 ppm. The test samples and their representation are given in Table 1. Mineral oil-LN 70 is procured from a local retailer of Indian Oil Corporation Ltd, and this is used as the base oil in this work. Oil is firstly filtered with microfilter material as it can retain substances above 1mm size. The prepared nanostructures are mixed at 0.01%wt, 0.05%wt and 0.1%wt mass fraction in transformer oil to get nanofluid of SiO_2 and CQD covered SiO_2 . The blend of oil and nanomaterials is done in a magnetic mixer and this stirrer process is continued for 60 min. After the stirrer process is completed, the nanofluid is exposed to ultrasonication in a high frequency sonicator for about 60 min. The nanofluid is thermally exposed in an oven for about 48 h at 75 °C temperature. The prepared nanofluid and base fluid are left untouched for about 96 h to confirm that there is no agglomeration in the nanofluids.



Figure 1. (a) FESEM image of SiO_2 , (b) FESEM image of SiO_2/C , (c) elemental mapping of Si atoms.

Sample identity	Type of nanofiller	Mass fraction
MO	-	-
S1	SiO_2	$0.1 \mathrm{g/L}$
S2	SiO_2	$0.5 \mathrm{g/L}$
S3	SiO_2	1.0 g/L
C1	$CQD-SiO_2$	$0.1 \mathrm{g/L}$
C2	$CQD-SiO_2$	$0.5 \mathrm{g/L}$
C3	$CQD-SiO_2$	1.0 g/L

Table . Test sample representation.

2.2. Partial discharge test

Partial discharge is evaluated for the experimental studies simulating sharp imperfections (needle and plane electrode configuration) and quasi-uniform field circumstances (rod and plane electrodes as well as plane to plane electrode configuration). The graphic diagram of the test arrangement used to record partial discharge is shown in Figure 2. The setup consists of a 180ml vessel where the needle to plane electrodes (needle curvature radius of $1.5 \ \mu$ m) of 5mm gap distance between them is used to simulate corona discharge. In addition to needle-plane electrode geometry, rod to plane and plane to plane electrode geometry were used for the comprehensive study of partial discharge. The needle electrode of the PD vessel is connected to a 100kV transformer and the plane electrode is linked to the ground cable as shown in Figure 3. Partial discharge signals were measured using a high-frequency current transformer (HFCT) device, connected across the ground cable of the plane conductor and a PD analyser capable with 50MHz bandwidth, which is possible to record a discharge pulse with sampling rate of 100 MS/s as well as sensitivity of 2 mV/div to 5 V/div. The same experimental setup of needle and plane electrode geometry is used for rod to plane and plane to plane electrode geometry by swapping the electrodes in the sample test vessel.



Figure 2. Electrode configurations (a) needle-plane electrode, (b) rod-plane electrode, (c) plane-plane electrode.



Figure 3. Experimental setup of partial discharge test.

3. Results and discussions

3.1. Effect of nanofiller mass fraction and electrode geometry on PRPD pattern

PD activity is analyzed with respect to applied electric field strength across the test cell. A needle and plane electrode geometry is used to examine the corona discharge characteristics of partial discharge classification. The PDIV values for all the test specimen such as pure mineral oil, SiO_2 nanofluid, and CQD- SiO_2 nanofluid were measured and the comparison of the results is reported in Figure 4. The initiation of PD pulse takes place at 13 kV in the case of base transformer oil, this result is in accordance with the inception voltages observed by other research reports like Tsuchie et al. [18]. The inclusion of 0.01%wt mass fraction of SiO_2 nanostructures in transformer oil has improved the PD inception voltage to 16kV and it is 3kV greater than that of base transformer oil.



Figure 4. Partial discharge inception voltage of test samples.

The nanostructure level of SiO_2 nanoparticles is further increased to 0.05% wt and 0.1% wt and it is observed that the addition of SiO_2 nanoparticles has raised the PD inception voltage to 18kV. Improvement in PDIV due to the addition of SiO_2 is reported in several research papers [19–22]. The PDIV results observed for C1 nanofluid are found to be 18kV, which is 2kV higher than that of SiO_2 nanofluid of the same concentration. CQD nanoparticles concentration is raised to 0.05% wt and further to 0.1% wt and in both cases the PDIV is found to be 20kV, which are 23% higher than pure transformer oil and 11% greater than SiO_2 nanofluid of the same concentration. The PDIV results obtained for other electrode configurations like plane-plane and rod-plane configurations were also in agreement with PDIV values of needle to plane electrode geometry in which CQD treated nanofluids perform well with higher PDIV values.

Phase-resolved partial discharge pattern recorded at 24 kV test voltage for every samples in needle-plane test setup are reported in Figure 5. The inclusion of CQD modified SiO_2 nanostructures influences the partial discharge performance of mineral oil, since it is possible to notice the PD magnitude of C2 and C3 samples are lower with minimal dispersion. Figure 6 shows the correlation between PD magnitudes, applied electric potential variation and nanostructure mass fraction in needle-plane electrode geometry. It is noticed that, PD magnitude increases when the system voltage increases beyond the inception voltage. The sample MO recorded the maximum magnitude in all test voltages and the nanofluid samples showed lower PD magnitude than the MO sample. Nanofluids S2 and S3 of SiO_2 nanofluids showed better improvement than the MO sample. Meanwhile sample C2 and C3 of CQD nanofluid showed a much-reduced PD magnitude than MO and SiO_2 nanofluids.

Figure 7 shows the effect of CQD- SiO_2 nanoparticles in PD magnitude for three different electrode configurations. CQD treated nanofillers effectively reduced the streamer development in nanofluid and results



Figure 5. PRPD pattern of samples (a) needle-plane, (b) rod-plane, (c) plane-plane electrode configuration, 24 kV test voltage.

in improved PD magnitude reduction. In plane-plane electrode configuration, similar kind of results with lower PD magnitude is observed as like in needle-plane test. Rod-plane electrode configuration results were also in relevance with other configurations with better performance of C1 and C2 nanofluids. The presence of sharp tip in needle-plane setup results in higher PD magnitude than rod-plane electrode without sharp tips. The dependency of PD magnitude on electrode configuration is well observed in plane-plane electrodes with minimal PD activity.

Under applying test voltage to needle electrode, free electrons (produced due to corona discharge) are absorbed by silica nanoparticles resulting in the production of negative ions near the needle electrode which restrain the weakening of the external electric field between the needle and plane geometry [21,23]. Therefore, it is hard to expand corona discharge further, so that the PD activity reduces. The increased shallow trap density of liquid insulating medium converts the fast mobile electrons to slower ones and hinder the streamer propagation



Figure 6. PD magnitude variation concerning nanofiller concentration and applied voltage.



Figure 7. PD magnitude variation concerning nanofiller concentration and electrode configuration, 24 kV test voltage.

hence reduces the propagation of PD [24]. The surface-treated silica in mineral oil reduces the partial discharge tendency to produce molecular chain scission and degradation of insulating oil. The hydrophilic properties of insulating silica particles in transformer oil may lead to absorption of moisture from the oil medium, thus decrease the creation of partial discharges at the early phase and it may be the cause for the enhancement in the PDIV of nanofluids.

Since both CQD and dispersion medium, which is insulating oil, are hydrophobic in nature, hence surface energy and interfacial tension among the nanofillers/liquid insulation interfaces can extremely reduce when put in interaction [13]. Hence, the attractive force between oil/CQD– SiO_2 nanofillers will be predominant, thus favoring better dispersion of CQD-covered silica when compared with pure silica. Surface modification of nanoparticles with CQD introduces further steric and electrostatic repulsive force, due to the presence of surface functional groups such as hydroxyl, epoxide and carboxylic acids [24,25]. This can increase the electrical doublelayer repulsive force between CQD modified silica nanofillers, which is superior to Van der Waals attraction force, which reduces the agglomeration of nano additives in the liquid insulating medium. The lower relaxation time constant of SiO_2 nanofillers leads to the quick absorption of free electrons on the surface of nanoparticles [26]. The 2.5–5nm size of CQD provides surface passivation towards inorganic materials. Partial coverage of CQD over the surface of the SiO_2 nanoparticles does not affect the silica nanoparticle's hydrophilic properties and, hence, it may results in moisture absorption from the liquid insulating medium, thus increase the PDIV and reduce PD magnitude.

3.2. Equivalent time length of PD pulse

PD time length and the bandwidth of recorded PD are analyzed based on Equations 1 and 2 where the time domain partial discharge signal is s (t).

$$\sigma_T = \sqrt{\int_0^T (t - t_0)^2 \bar{s}(t)^2 dt} \tag{1}$$

$$\sigma_T = \sqrt{\int_0^T f^2 \bar{s}(f)^2 df} \tag{2}$$

f denotes the frequency of discharge pulse and s(f) denotes Fourier transform of the normalized pulse. The

dots in the time-frequency plot shows the real values recorded from Equations 1 and 2. Figure 8 depicts T-F map of MO and nanofluids of SiO_2 and CQD at 24kV for needle, rod and plane electrode combinations. The equivalent time length of base mineral oil without any nanofiller is heading nearly 300ns but when nanofillers of SiO_2 and CQD are added, it reduces to 150ns which is almost half the equivalent time length of base mineral oil. SiO_2 nanofluids have PD time lengths up to 130ns with an average value of 100ns. The PD time length of CQD nanofluids is observed to be a maximum of 120ns with an average value between 60 to 70ns. The effect of active reduction in the equivalent time length of PD is studied in different aspects such as nanofiller concentration and variation in test voltages. In needle plane configuration, the time length variation is analyzed for varying nanofiller concentrations along with the increase in test voltage, the result observations from the test are summarized in Figure 9.



Figure 8. PRPD t-f mapping of samples, test voltage 24 kV.

Partial discharge time length tends to increase when there is an increase in applied voltage. For an increase of 8kV in applied voltage there is an average increase of nearly 40 to 50ns in time length of PD regardless of the kind of filler and concentration. In the aspect of nanofiller concentration to the PD time length, there is a noticeable decrease in time length. With an increase in filler concentration, it can be noticed that S1 and S2 have lesser time length when SiO_2 nanofluids are concerned with MO and the time length is further reduced with addition of CQD nanofillers, the reduction can be observed in the results of C2 nanofluid with a minimum time length in most of the cases. Even with an increase in voltage, the time length of CQD nanofluids remains lesser than MO and SiO_2 nanofluids. The results in Figure 10 show that MO has maximum pulse time length in plane-plane and rod-plane electrode configurations. The equivalent time length of CQD- SiO_2 nanofluids is lesser than that of MO and silica nanofluids in all the electrode configurations.



Figure 9. Equivalent time length under increasing test voltage.



Figure 10. Equivalent time length of test samples. 24 kV test voltage.

3.3. PD waveform observations

The time-frequency plot of PD observations in needle-plane electrode geometry exhibited that MO has the maximum pulse amplitude and CQD nanofluid has the least pulse amplitude than MO and SiO_2 nanofluid. It is understood from Figure 11, that discharge pulse peak decrease for C2 sample than S2 sample and MO sample. The frequency spectrum of MO, S2 and C2 indicated a peak at 19, 20 and 20.5 MHz and the frequency spectrum observations showed peak value in the range of 19 to 21MHz for all the test samples in Figure 12. The duration of the sequence of discharge pulses is slightly different for the three samples. The CQD nanofluids have a lower duration than MO and silica nanofluid, sample C2 showed much-reduced pulse duration. The discharge pulse period mostly rely on the number of discharge pulses in a single PD sequence (repetition rate). Numerous research works have stated that the PD sequence repetition rate is more delicate to the existence of moisture and scums such as dissolved gasses, dust, fibrous and ionic impurities [22–24].

Hence, the lower repetition rate can be possibly achieved by the absorption of these impurities by the inclusion of SiO_2 and CQD- SiO_2 nanostructures and in this concern, CQD- SiO_2 nanoparticles perform better than SiO_2 nanoparticles. Repetition rate with respect to varying electric potential applied and mass fraction of nanofillers in needle to plane electrode geometry is shown in Figure 13. The repetition rate of pure mineral oil is higher than nanofluids; the value varies from 3 to 4.9. The repetition rate of SiO_2 nanofluids is lesser than MO, S3 has a much lesser value and has a maximum of 3.9 which is 25% lesser than MO. The repetition rate of CQD nanofluids in most of the cases is lesser than MO and SiO_2 nanofluids with C2 having a minimum value of 2.9 which is 68% lesser than MO and 34% lesser than the S3 sample. The repetition rate of the test samples are evaluated under dissimilar electrode configuration and the results are summarized in the graph reported in Figure 14. The test results are reported for a test voltage of 24kV and the reputation rate of CQD nanofluids is much lower than MO and silica nanofluids in different electrode configurations.

3.4. Statistical observation of partial discharge waveform

Statistical examination of partial discharge waveform in random probability distribution can produce real-time evidence and the condition of dielectric medium. Skewness is evaluated to understand the distribution of data in a stated range, while the shape parameter is to measure the dispersion in the height of PD pulse. PD software evaluation is done to analyze the skewness, alpha and beta values of the PRPD pattern of oil specimen recorded at 24kV test voltage. The skewness parameter is analyzed to determine the asymmetry or degree of dissimilarity (tilt) in the data. Statistical evaluation is done and the results outcomes of the needle to plane electrode for



Figure 11. PD pulse (left) and frequency spectrum (right) (a) MO, (b) S2, (c) C2. 24 kV test voltage.



Figure 12. PD pulse peak in frequency spectrum, 24 kV test voltage.

the sample are compared and reported in Figures 15–17. The value of skewness is obtained with Equation (3).

$$Skewness(S_k) = \frac{\sum_{i=1}^{N} (x_i - \mu)^2 f(x_i)}{\sigma^3 \sum_{i=1}^{N} f(x_i)}$$
(3)

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Figure 13. Repetition rate variation w.r.t. nanofiller concentration and test voltage.



Figure 15. Mean of PD skewness distribution for test samples (a) skewness positive, (b) skewness negative.



Figure 14. PD repetition rate for test samples, 24 kV test voltage.



Figure 16. Mean value of scale parameter (a) α positive, (b) α negative.

f(x) denotes the partial discharge magnitude q, μ is the mean value of q, σ denoted the variance of q.Skewness is evaluated to a reference Weibull distribution. The skewness of the test samples are depicted in Figure 15 under needle plane electrode geometry at test voltage of 24kV. The skewness numbers of nanofluids were observed to be lesser than the sample MO. Skewness is lesser for SiO_2 nanofluid and C2 nanofluid has a significantly lower distortion than MO and SiO_2 samples. It can be understood from the illustration that the skewness tends to increase beyond certain concentration levels for both SiO_2 and CQD nanofluid.

The variation of scale parameters for positive and negative pulse sequence of mineral oil and nanofluids for needle to plane electrode geometry at 24kV electric potential is expressed in Figure 16. The scale parameter for all the samples tends to increase for an increase in voltage. The scale parameter of the MO sample is higher in all the test voltages and for nanofluids, the value remains less with a small degree of variation for voltage rise. The α value is lower for the S2 sample when considering MO and silica nanofluids and the samples C1 and C2 showed a much lower α value than base mineral and silica nanofluids. Figure 17 depicts the variation of shape



Figure 17. Mean value of shape parameter (a) β positive, (b) β negative.

parameter (both positive and negative phase values). It can be observed from the comparison that the shape parameter tends to be higher for nanofluids than for transformer oil without nanofillers. Shape parameter is higher for C2 nanofluid concentration than that of other concentrations. The shape parameter value is higher for all the CQD concentration levels when compared with SiO_2 nanofluids of the same concentration. It is possible to conclude from the results that while comparing the shape parameter of nanofluids the CQD nanofluid is having better values of than that of SiO_2 nanofluids. The inclusion of a lower %wt mass fraction of nanofillers demonstrated lower scale parameter values, which authorizes less PD data dispersion. Reduced scale parameter value alongside with high β value shows minimal discharge with minimum dispersion. These observations demonstrates that the inclusion of SiO_2 modified CQD nanoparticles may increase the shape parameter and tend to decrease the skewness parameter by which it is possible to confirm that the inclusion of CQD nanostructures does well than SiO_2 nanoparticles in dipping the partial discharges.

4. Conclusion

The inclusion of CQD modified SiO_2 nanostructures to transformer oil has influenced the dielectric characteristics of the transformer oil, this consequences in the influence of a reduction in the PD activity. The inclusion of CQD nanostructures has enhanced the PD inception voltage up to 23% and nearly 6V greater than that of pure transformer oil and SiO_2 nanofluid. Magnitude of PD is decreased from 0.1v to 0.02v due to the influence of CQD-modified SiO_2 nanofluies in transformer oil. Lower PD activity results in strong dielectric characteristics, which ensures reliable and safe operation of power system. Further detailed research in the application of CQD nanoparticles with other nanoinsulating materials can improve the insulation condition in power apparatus.

Acknowledgment

Author (C.S) would like to thank the Department of Science and Technology (Grant No. SR/NM/NT-1051/2016 (G) dt.15.03.2018), Govt. of India, for funding this research under Nano Mission Scheme. Author S.C gave the idea and evaluation. Authors K.P. and B.S. did the experiments and results, P.K. wrote the paper.

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