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Sensors

Multifractal signatures of light-driven self-organization in acrylated epoxidized soybean oil polymers

A.E. Kiv[1,2], V.N. Soloviev[1,3*], A.O. Bielinskyi[3,4,5], M.A. Slusarenko[3], T.S. Kavetskyy[6,7*], O. Šauša[7,8], H. Švajdlenková[8,9], I.I. Donchev[1], N.K. Hoivanovych[6], L.I. Pankiv[6], O.V. Nykolaishyn[6], O.R. Mushynska[6], O.V. Zubrytska[6], A.V. Tuzhykov[1], M. Kushniyazova[10]

[1]*South Ukrainian National Pedagogical University named after K.D. Ushynsky, 65020 Odesa, Ukraine*
[2]*Ben-Gurion University of the Negev, 84105 Beer-Sheva, Israel*
[3]*Kryvyi Rih State Pedagogical University, 50086 Kryvyi Rih, Ukraine*
[4]*State University of Economics and Technology, 50005 Kryvyi Rih, Ukraine*
[5]*Kyiv National Economic University named after Vadym Hetman, 03057 Kyiv, Ukraine*
[6]*Drohobych Ivan Franko State Pedagogical University, 82100 Drohobych, Ukraine*
[7]*Institute of Physics, Slovak Academy of Sciences, 84511 Bratislava, Slovakia*
[8]*Department of Nuclear Chemistry, FNS, Comenius University, 84215 Bratislava, Slovakia*
[9]*Polymer Institute, Slovak Academy of Sciences, 84541 Bratislava, Slovakia*
[10]*Kazakh-British Technical University, 050000 Almaty, Kazakhstan*
Corresponding author(s) e-mail(s): vnsoloviev2016@gmail.com; kavetskyy@yahoo.com

Abstract. Multifractal properties of acrylated epoxidized soybean oil with and without a photoinitiator have been investigated. Using multifractal detrended fluctuation analysis, the photopolymerization process is analyzed by varying illumination duration and intensity. The research is conducted in two stages: the results concerning temperature fluctuations of the entire photoirradiation process are initially presented followed by a sliding window procedure to monitor system complexity during light cycling. Various multifractal measures including the generalized Hurst exponent $h(q)$, the multifractal Rényi exponent $\tau(q)$, the generalized fractal dimensions $D(q)$ with the left and right side curvature (ΔD_L, ΔD_R), the multifractal singularity exponents α, the multifractal spectrum $f(\alpha)$, and the multifractal heat capacity $C(q)$ with its integrated characteristic C_{area} are examined for both initial and shuffled series. The empirical results demonstrate that the moments of switching on and switching off photoirradiation are characterized by an instantaneous increase in the degree of multifractality of the system. It is hypothesized that the observed increase in multifractality at the light-switching-on/off stages is indicative of a quasi-phase transition, which may be associated with the transformation of orientational defects in the polymer network.

Keywords: acrylated epoxidized soybean oil, photopolymerization, self-organization, multifractality, multifractal detrended fluctuation analysis, phase transition.

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1. Introduction

Multifractality is one of the most promising concepts for studying complex systems phenomena that has pervaded different domains including finance [1], air traffic [2], biology [3], social psychology [4], physics [5–8], *etc*. The multiscale approach aims to provide a spectrum of fractal dimensions for a wide range of time and length scales. Using the multifractal approach, we can assess the homogeneity of a studied surface, serial long-range nonlinear correlations, and the fat tails in the distribution of fluctuations.

Polymers, being intricate macromolecules, embody the essence of complex systems, exhibiting hierarchical organization and emergent behaviors. In the realm of polymer science, photopolymerization stands as a quintessential process demonstrating such complexity. During photopolymerization, monomer molecules undergo radical polymerization upon exposure to light, resulting in formation of polymer chains with distinct properties.

At its core, photopolymerization manifests itself as a multifaceted process, with smaller-scale components like monomers and photoinitiators interacting to yield

emergent behavior at larger scales, culminating in polymerization. These emergent phenomena, such as polymerization kinetics and network formation, cannot be fully elucidated by the summation of individual molecular interactions alone, mirroring the dynamics observed in complex systems.

Moreover, the distribution of polymer chain lengths and spatial arrangements within the resulting polymer network frequently exhibits power law distribution, which is indicative of multifractal characteristics. The scaling behavior inherent in such systems, akin to that observed in multifractals, underscores the interconnectedness of polymerization processes across various length scales.

Furthermore, the absence of a characteristic length scale in photopolymerization is consistent with the concept of self-similarity, which is a defining feature of multifractals. Each constituent element, regardless of its molecular size, contributes equivalently to the global evolution of the polymer network during photopolymerization, reflecting the self-similar nature observed in multifractal systems.

Among the numerous general properties characterizing complex systems, the presence of critical thresholds appears to be a common property [9]. There are a lot of critical transitions, which represent potential hazards to various aspects of daily life [10]. Examples of such perilous transitions include spontaneous systemic failures in human health, systemic market crashes in global finance, sudden shifts in oceanic circulation or climate patterns, *etc*. Due to the inherent complexity of these systems, it is often impossible to fully predict their behaviors solely based on the behaviors of their individual components. Therefore, the characterization of the dynamical processes of complex systems in terms of macroscopic quantities, such as time series data, stands as a fundamental and crucial problem across numerous research domains [11].

The recent study [12] aimed to comprehensively outline the recent advancements in fractal theory applied to packaging materials, offering valuable insights into the practical applications of fractals in this domain. Various fractal analysis techniques were systematically reviewed for different types of materials, including inorganic materials like metal alloys and ceramics, polymers, and their composites. This review covered aspects such as fractal feature extraction methodologies and methods for calculating fractal dimensions. The studies cited in this review usually consider a single value of the fractal dimension or its spectrum only for a single scanned material surface or the entire time series reflecting the acoustic emission of the material. Their disadvantage is that they do not present the comparative dynamics of the fractal index with the evolution of the material itself, where the fractal dimension, in theory, could serve as an indicator-predecessor of the processes of complexification or degradation in the material.

In this paper, for the first time, we present a comprehensive technique of multifractal analysis for the photopolymerization process, which allows us to obtain a spectrum of different indicators or indicators-precursors of phase transformation and self-organization processes in the polymer used for sensors. Our analysis relies on the time series of temperature fluctuations of a polymer material that is subjected to cyclic photoillumination. The analysis is carried out both for the whole time series and for time-shifted window fragments, which allows us to carry out the multifractal analysis of the photopolymerization development.

2. Procedure of multifractal detrended fluctuation analysis

Multifractal detrended fluctuation analysis (MF-DFA) [13, 14], which is derived from the conventional DFA approach [15, 16], is one of the most applicable approaches for the multifractal analysis of time series data [17–19].

Since we want to estimate the multifractality of the polymer, regarding its temperature during photopolymerization, first of all, we need to consider its time series data $\{x_i \,|\, i=1,2,...,N\}$, for which we estimate cumulative profile $Y(i)=\sum_{k=1}^{i}\left[x_k-\langle x\rangle\right], i=1,2,...,N,$ where $\langle x\rangle$ is the mean value of the analyzed time series.

Then, $Y(i)$ is divided by $\mathrm{int}(N/s)$ non-overlapping segments of equal length *s*. Since *s* is not always a multiplier of the length *N*, the same dividing procedure is repeated from the opposite side of a time series to consider all segments. Thus, we obtain $2N_s$ segments.

Each segment is detrended from the difference of a segment and a local trend $\tilde{Y}^{\nu}(i)$, which is found by least-squares fitting. Polynomials of different orders can be used to detrend the series. In our case, we use a first-order polynomial $\tilde{Y}^{\nu}$. Then, for $\nu=1,...,N_s$ segments of length *s* in one direction, we find their variance $F^2(\nu,s)=s^{-1}\sum_{i=1}^{s}\left\{Y\left[(\nu-1)s+i\right]-\tilde{Y}^{\nu}(i)\right\}.$ For $\nu=N_s+1,\ N_s+2,...,2N_s$ segments in reverse direction, the variance is defined as $F^2(\nu,s)=s^{-1}\sum_{i=1}^{s}\left\{Y\left[N-(\nu-N_s)s+i\right]-\tilde{Y}^{\nu}(i)\right\}.$

As a result, the overall *q*-th order fluctuation function can be defined as

$$F_q(s)=\left\{\frac{1}{2N_s}\sum_{\nu=1}^{2N_s}\left[F^2(\nu,s)\right]^{q/2}\right\}^{1/q}, \qquad (1)$$

for $q \neq 0$ and

$$F_0(s)=\exp\left\{\frac{1}{4N_s}\sum_{\nu=1}^{2N_s}\ln\left[F^2(\nu,s)\right]\right\}, \qquad (2)$$

for $q = 0$.

Then, the log-log plot of $F_q(s)$ *versus s* should give us a power-law relation $F_q(s) \propto s^{h(q)}$, where $h(q)$ represents a generalized Hurst exponent [14]. For $q = 2$, we expect $h(q)$ to be the standard Hurst exponent.

Values of q emphasize the density of small (large) fluctuations. If these values are positive (negative), an accent is made on scaling properties of large (small) fluctuations. If the multifractal characteristics do not depend on the q values, the studied time series is presented to be monofractal.

Different types of fluctuations in a multifractal system can exhibit varying degrees of persistence and correlation. The generalized Hurst exponent can highlight such information [20]:

- If $h(q) > 0.5$, fluctuations with a particular magnitude are shown to be persistent, with an increase (or decrease) in one value followed by a subsequent increase (or decrease) in the next value.
- If $h(q) < 0.5$, fluctuations are anti-persistent, that is an increase (decrease) of one value is likely to be followed by a decrease (increase) of another value, *i.e*., it is a mean-reverting process.
- If $h(q) \approx 0.5$, the values of a series follow a random walk.
- If $h(q) > 1$, the values of a series are said to be highly correlated and non-stationary.

Apart from the generalized Hurst exponent, we can derive such multifractal measure as the multifractal Rényi (mass) exponent $\tau(q)$, which is related to $h(q)$ through the following relationship [21]:

$$\tau(q) = qh(q) - 1. \tag{3}$$

Utilizing the framework of MF-DFA, we can advance our understanding of temporal signals as complex thermodynamic systems. Within MF-DFA, key parameters such as the multifractal Rényi exponent $\tau(q)$, the Lipschitz–Hölder exponent $\alpha(q)$, the multifractal spectrum $f(\alpha)$, and the distortion exponent q can be construed as analogous to thermodynamic quantities, namely, free energy, internal energy, entropy, and temperature, respectively. Through $\tau(q)$ and q we can define multifractal "specific heat" $C(q) \equiv -\partial^2 \tau(q) / \partial q^2$ [22]. The specific heat, which serves as an indicator of the rate of energy variation, provides valuable insights into the occurrence of phase transition phenomena [23]. In thermodynamic systems, a phase denotes a state characterized by uniform physical properties, with a phase transition referring to the abrupt alteration of certain properties triggered by a critical external condition. The comprehensive measure of multifractality is expressed as the total multifractal specific heat $C_{area} = \int C(q) dq$.

The q-th order Hurst exponent is just one of several types of scaling measures used to parameterize the multifractal structure of time series. As presented earlier, we could derive a q-th order mass measure $\tau(q)$ and then use it to derive a q-th order singularity exponent $\alpha(q)$. Through a Legendre transform [24–26], we could derive the fractal dimension $f(\alpha)$ [27] of the fluctuations (regions) with the degree of singularity $\alpha(q)$:

$$f(\alpha) = q\alpha(q) - \tau(q). \tag{4}$$

The singularity exponents α for large highly concentrated fluctuations are small and located in the left tail of the spectrum, whereas for small fluctuations they are large and located in the right tail of the spectrum.

Therefore, the magnitude of multifractality is characterized by a significant deviation of the local singularity exponent from the central tendency, $\alpha(0)$. A monofractal signal is the case when alpha remains almost constant. In some cases, the multifractal spectrum is reduced to a single point at a given α.

The range of α indicates the variety of singularity exponents that describe the dynamics of the system, and $f(\alpha)$ indicates the contribution of elements with the corresponding α.

The multifractal spectrum can be characterized by different widths, which indicates the variability of processes occurring within the system. Furthermore, it can be either symmetrical or asymmetrical. The asymmetry can be either right- or left-tailed, indicating different degrees of influence of highly concentrated and low-concentrated elements (fluctuations).

Another approach considers a set of the generalized fractal dimensions $D(q)$ that can be directly calculated from Eqs. (3) and (4):

$$D(q) = \tau(q)/(q-1) \tag{5}$$

and

$$(q-1)D(q) = q\alpha(q) - f(\alpha(q)). \tag{6}$$

Let us consider the case when $q = 1$. Since $\tau(1) = 0$, it is clear from Eq. (6) that $\alpha(1) - f(\alpha(1))$. Upon differentiation of Eq. (4) with respect to q, it turns out that $\alpha(1) = D_1$, where D_1 is the information dimension [28, 29]. Therefore, it appears that $D_1 = \alpha(1) - f(\alpha(1))$. As can be seen, there is uncertainty in Eq. (5) at $q = 1$. This uncertainty can be circumvented by defining $D_1 = -\tau(1)$ [30, 31].

The information dimension can be used to describe the spatial heterogeneity of a system. The more homogeneous the attractor is, the higher this dimension should be. That is, the more configurations the elements of a given system can take, the more information we need to account for each element. With spatial homogeneity, the information entropy also increases, which links the information dimension to the concept of entropy.

Now consider the case where $q = 2$. Following Eq. (6), we obtain that $D_2 = 2\alpha(2) - f(\alpha(2))$, where D_2 is known as the correlation dimension [25, 28, 32].

Looking at $f(\alpha)$, we can derive different quantitative characteristics that describe the degree of multifractality

of a studied region and asymmetry. If the fractal indices are equal to each other, this may indicate that the system is quite simple, its elements are largely independent of each other, and its behavior approaches a normal Gaussian distribution. However, if the behavior of the system is quite complex, then the generalized fractal dimensions in the aggregate may have a sigmoid shape, indicating more complex internal relationships in the system.

The degree of this complexity may be characterized by the curvature of the right and left tails of the generalized fractal dimensions. The right side $\Delta D_R = D_0 - D_{q\max}$.

The higher is the value of this measure, the stronger is the degree of influence of the elements with the highest concentration (density or fluctuation amplitude) on the overall complexity of the system.

We can also determine the curvature of the left tail of the curve of generalized fractal dimensions, $\Delta D_L = D_{q\min} - D_0$.

This indicator will tell us how strong the influence of the fluctuated elements on the system complexity is.

3. Experimental

Examined samples. The experimental samples investigated in this study comprised sets of eight viscous solutions utilized for photopolymerization. These solutions consisted of acrylated epoxidized soybean oil (AESO) and vanillin dimethacrylate (VDM) in various molar ratios of monomers, namely AESO:VDM = 1:0, 1:0.25, 1:0.5, and 1:1, both with and without the inclusion of a photoinitiator (PI), *i.e.* 2,2-dimethoxy-2-phenylacetophenone (DMPA) [33]. The concentration of the utilized PI was 3 mol%, determined based on the total molar amount of all the monomers involved. The AESO and DMPA were from the Sigma-Aldrich, while the VDM was obtained from the Specific Polymers. The incorporation of the VDM into the AESO resulted in an elevation of the mixture viscosity [34].

Illumination procedure. Photopolymerization of the AESO/VDM samples was conducted using a Jaxman U1 LED light source emitting at 365 nm. To maintain consistent light intensity throughout the experiment, an adjustable LED power supply connected to an E36106A stabilized current source (Keysight Technologies) was employed. The light intensity was assessed and regulated utilizing an Ophir Photonics StarLite meter coupled with a PD300-UV probe.

Thermocouple temperature measurements. The temperature during the photopolymerization process was monitored using a measurement apparatus developed at the Institute of Physics of the Slovak Academy of Sciences [35]. T-type thermocouples sourced from the Omega Engineering were employed to record temperature data at 1-second intervals. More experimental details of the thermocouple temperature measurements and the results on the *in situ* polymerization monitoring by thermocouple can be found elsewhere [33].

4. Empirical results

Fig. 1 presents time-dependent temperature fluctuation of the AESO-based samples with PI and without PI (wPI). Each light switch is indicated by a text arrow with the titles “on” and “off”. Additionally, we estimate the rate of change and the average value of the autocorrelation function for the temperature time series (*T*). The rate of change is calculated as $g = [x(t+\Delta t) - x(t)]/x(t)$.

As indicated in Fig. 1b, all the fluctuations that do not belong to the regions of light on and light off remain completely independent of each other. The autocorrelation function shows that the temperature fluctuations exhibit the highest degree of correlation at the moment of turning the light on and off, pointing to the most significant self-organization processes there. The amplitude of normalized arrivals also begins to decay at the moment of illumination, but this amplitude remains practically unchanged until the light is deactivated. This may indicate that the process of polymer network organization due to continuous radical generation occurs not only at the time of switching on/off, but in principle during the entire illumination stage. As previously indicated, the most significant processes in the polymer may occur at the beginning and end of the illumination period.

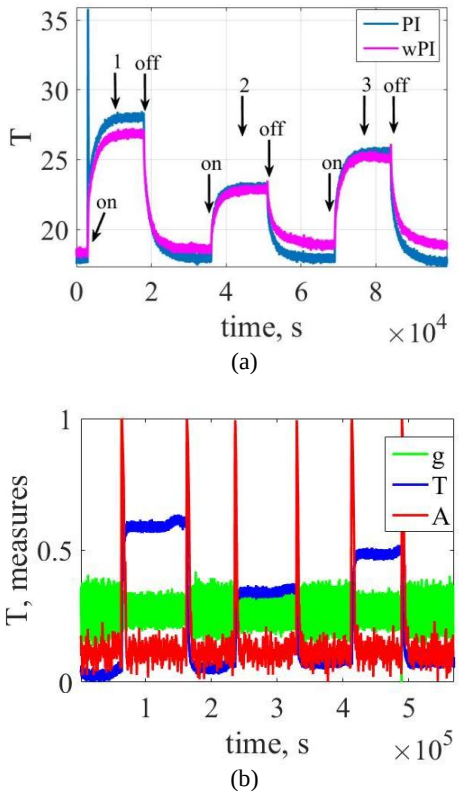
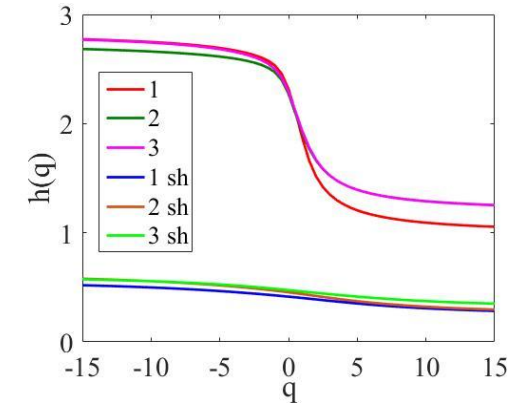


Fig. 1. Time dependence of temperature response (*T*) for a series of AESO-based samples with PI and without PI (a). Comparison of temperature fluctuations, rate of change (*g*) and autocorrelation function (*A*) (b). (Color online)

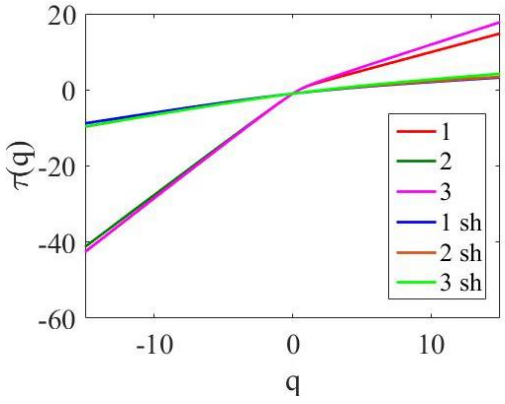
Furthermore, to measure the multifractality degree of the photopolymerization process, we present the comparative dynamics of the described indicators calculated using the sliding window approach [22, 36] along with the initial time series of temperature fluctuations. Let us formulate a sliding window algorithm. We select a fragment of the time series (window), calculate the necessary complexity measures, move the window along the time series by a predetermined value (step), and repeat the procedure until the time series is exhausted. Then, by comparing the dynamics of the actual time series and the corresponding multifractal measures, we are able to track the characteristic changes in the dynamics of the complexity measure with the change of the system under study. If a specific measure of complexity exhibits consistent behavior across all specified time periods (*e.g.*, light-switching-on/off stages) and is correlated with changes in the complexity of the system, it can serve as an indicator of those changes.

The MF-DFA calculations used the following parameters: (1) the sliding window of 2000 s and step size of 25 s; (2) polynomial order $m = 1$ for the fitted trend $\widetilde{Y}^{\nu}$ in a segment ν; (3) the values $q_w \in [-10; 10]$ with a delay $\Delta q_w = 1$ for the sliding window procedure and $q_{all} \in [-15; 15]$ with a delay $\Delta q_{all} = 0.5$ for the entire time series to consider the manifestation of fractality in the photopolymerization process across a wide range of different statistical moments; (4) the time scale s varying from 50 to 20000 for the whole time series and from 50 to 2000 for the sliding window procedure.

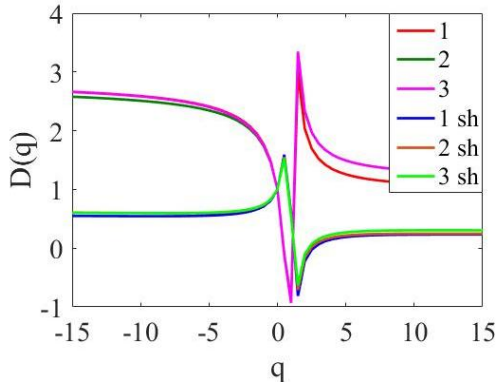
For illustrative purposes, Fig. 2 presents the generalized Hurst exponent $h(q)$, the multifractal Rényi exponent $\tau(q)$, the generalized fractal dimensions $D(q)$, the multifractal spectrum $f(\alpha)$, and the multifractal heat capacity $C(q)$ for the initial and shuffled time series of the photopolymerization stages with PI.



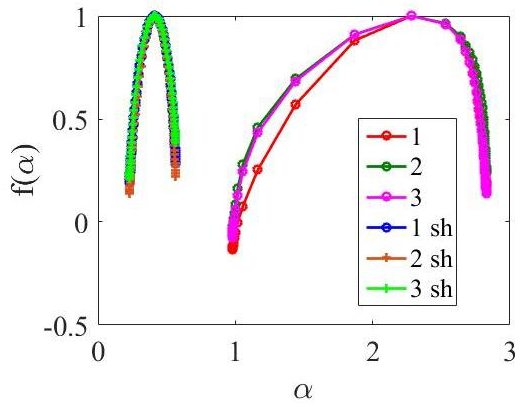
(a)



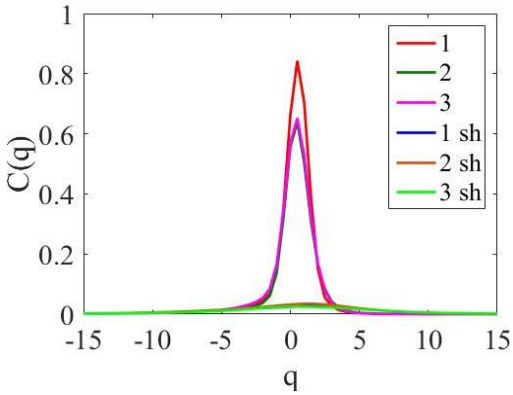
(b)



(c)



(d)



(e)

Fig. 2. The multifractal generalized Hurst exponent $h(q)$ (a), the multifractal Rényi exponent $\tau(q)$ (b), the generalized fractal dimensions $D(q)$ (c), the multifractal spectrum $f(\alpha)$ (d), and the multifractal heat capacity $C(q)$ (e) for the initial and shuffled (sh) time series of the photopolymerization process with PI. (Color online)

The following section will demonstrate that even in the absence of a photoinitiator, the studied polymer exhibits a significant degree of multifractality throughout the photopolymerization process.

As demonstrated in Fig. 2, when the light is turned on/off, the system approaches the critical region, and it is characterized by increasing multifractality. In these regions, there is a significant increase or decrease in the birth of free radicals, which affects the increasing degree of self-organization and correlation in the formation of the polymer chain. In Fig. 2a, the Hurst exponent of the q-th order for the multifractal series appears to be strongly dependent on q moments. This indicates that the source of multifractality is both short- and long-term correlation among the elements of the polymer. Since $h(q)$ is much greater than one, it may be concluded that the studied photopolymerization process reveals strong multifractal nature around the moment of instantaneous birth of free radicals at the beginning of irradiation of a material as well as at the termination of its illumination.

The shuffling procedure shows that this investigated process is indeed far from white noise since a weak dependence on q is now observed for $h(q)$. The independence of the Hurst exponent on q for the shuffled series suggests that nonlinear correlations and high-energy fluctuations lying in the heavy tail of the probability density function are the two most important manifestations of multifractality of the photopolymerization process. The same conclusions may be drawn when considering other measures of multifractality. $\tau(q)$ calculated for the whole series (Fig. 2b) is characterized by a sufficiently high degree of curvature as compared to the shuffled variant for all q. The Rényi dimension calculated for the whole series is characterized by a sufficiently high degree of curvature in comparison with the mixed variant for all values of q. The generalized dimensions $D(q)$ for the original series are characterized by sufficiently high values as compared to the shuffled one (Fig. 2c). As can be seen, $D(q)$ has a sigmoidal shape, but starts to diverge as soon as $q \to 1$. Nevertheless, this divergence does not affect the calculation of subsequent indicators.

Fig. 2d shows the multifractal spectrum for all three stages of light irradiation of the investigated AESO-based material. It can be seen from this figure that for the initial series, the multifractal spectrum shifts far to the right and is characterized by a larger range of different fractal dimensions at different α values. It is worth mentioning that the spectrum is asymmetric, and the left tail of the spectrum, which corresponds to positive q values (large-scale fluctuations), is predominant. For a series resembling white noise, $f(\alpha)$ is narrowed and close to $\alpha = 0.5$.

As can be seen from Fig. 2e, $C(q)$ is characterized by an island-shaped peak for the initial series and values close to zero for all the q values for the shuffled version. Considering the dependence $C(q)$, we may say that the most important processes that may be associated with phase transitions occur at $q \in [-5, 5]$. For statistical moments beyond this range, we do not observe multifractal dependences for the studied polymer.

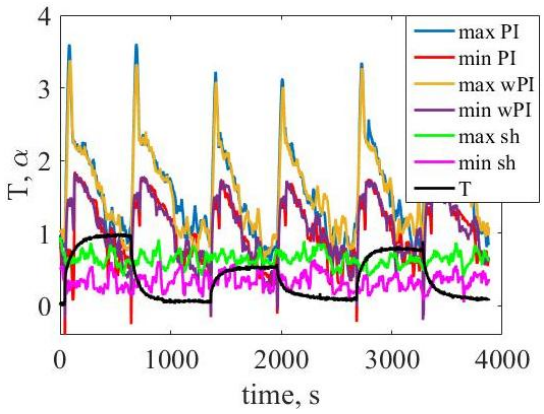


Fig. 3. Comparative dynamics of temperature measurements with PI and maximum and minimum singularity exponents calculated with PI, without PI, and shuffled series.

Next, let us present in Fig. 3 the comparative dynamics of the temperature fluctuations for the polymer with PI and the minimum and maximum singularity exponents calculated for the photopolymerization process with PI, without PI, and shuffled series.

As can be seen from Fig. 3, all the singularity indicators increase during the polymer quasi-phase transitions when the light is switched on/off. This indicates an increase in the complexity of the system: a sharp increase in the number of elements involved in the self-organized development of the system under study.

Fig. 4 illustrates the comparative dynamics of temperature fluctuations for the polymer without PI and information (D_1) with correlation (D_2) dimensions calculated for the polymerization process with PI, without PI, and shuffled series.

The information dimension is closely related to the concept of Shannon information entropy. An increase in the value of D_1 indicates a rapid rise in entropy, which serves as an indicator of the limited knowledge available regarding the current state of the system. As D_1 decreases, the entropy also decreases, which indicates an increase in spatial asymmetry, a decrease in system complexity, and an increase in our understanding of the current state of the system. This also means that we need less information to describe possible system configurations. It can be seen from Fig. 4a that D_1 is characterized by a decline during light-switching-on/off stages. This indicates an increase in the degree of regularity of the system and the collective convergence of the system's phase space trajectories towards a lower dimension.

We can see from Fig. 4b that D_2 increases during light-switching-on/off stages, indicating the increase in complex spatial organization, characterized by variations in chain lengths, branching, and spatial arrangements in the polymer. According to [25, 32, 37], $D(q)$ decreases monotonically for all the positive q values. Hence, we expect to observe the following dependence:

$D_0 \geq D_1 \geq D_2$. This relation can be flattened for non-fractal or less monofractal systems, *i.e.*, $D_0 = D_1 = D_2$. We can see from Fig. 2c that $D(q)$ is usually sigmoidal around $q = 0$ and high for multifractals. Since we observe the phenomena which arise due to the strong correlation between various components within the system, one may expect that D_2 will decrease during light-switching-on/off stages, since these states tend to a low-dimensional self-similar attractor. However, comparing Figs. 4a and 4b, we can notice that the stated monotonic decrease is not achieved for D_2. Referring back to Eq. (7) and Fig. 3, one can realize that the rapid increase of D_2 is due to rather rapid increase of the singularity exponents in the same regions of interest. Conceivably, the correlation dimension at $\alpha(q) \leq 1$ would exhibit a decline when the light is turned on/off, signifying a rapid increase in the self-organization of the system. Nevertheless, the presented dynamics do not exclude this conclusion.

Fig. 5 presents the comparative dynamics of temperature measurements for the photopolymerization process without PI and the curvature of the left (ΔD_L) and right (ΔD_R) sides of $D(q)$ calculated with PI, without PI, and shuffled series.

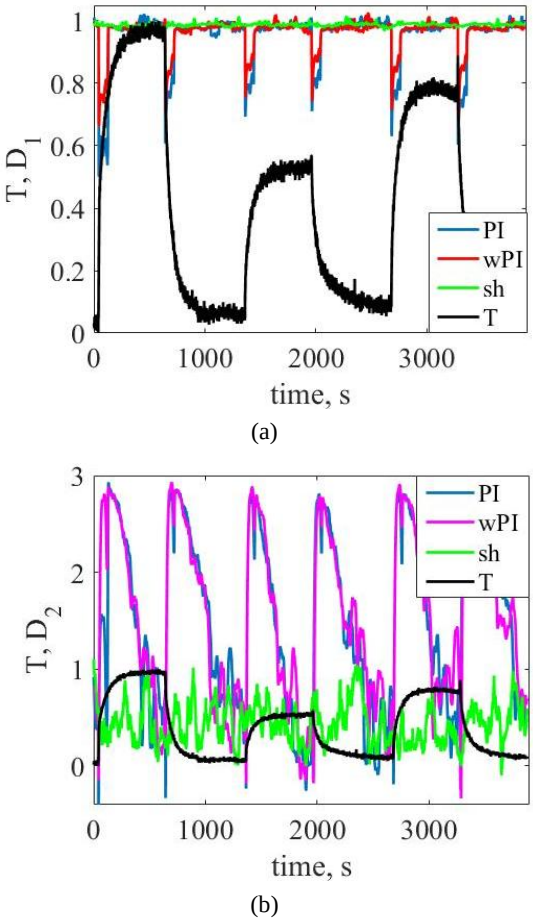


Fig. 4. Comparative dynamics of temperature measurements for the photopolymerization process without PI with D_1 (a) and D_2 (b) of the generalized fractal dimensions $D(q)$ calculated with PI, without PI, and shuffled series. (Color online)

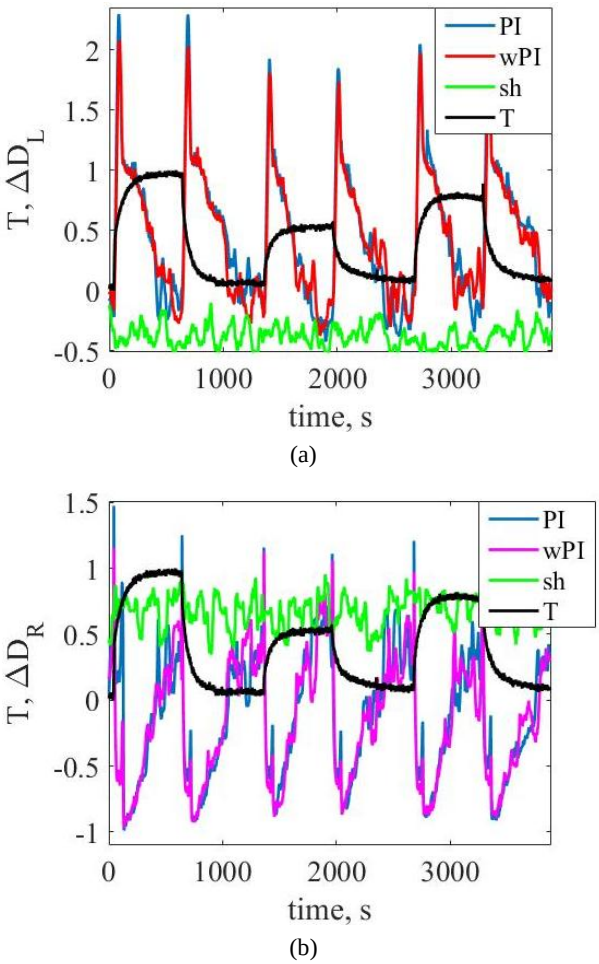
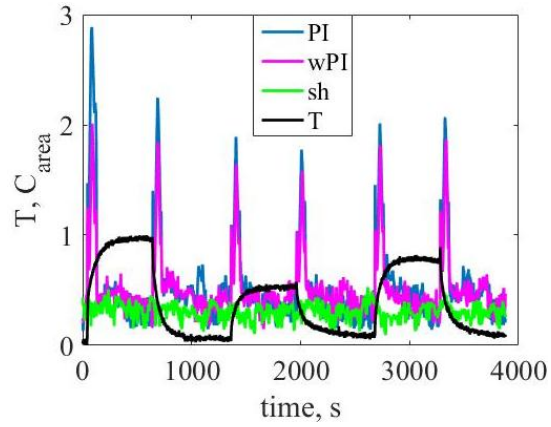


Fig. 5. Comparative dynamics of temperature measurements for the photopolymerization process without PI and the curvature of the left (a) and right (b) sides (ΔD_L, ΔD_R) of the generalized fractal dimensions $D(q)$ calculated with PI, without PI, and shuffled series. (Color online)

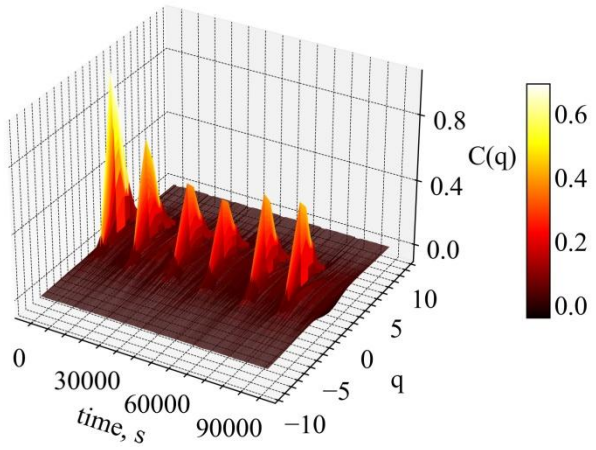
Fig. 5 demonstrates that ΔD_L and ΔD_R are asynchronous indicators of self-organized dynamics during photopolymerization. That is, both indicators react in a characteristic manner on light-switching-on/off stages, which suggests that large-scale elements play a dominant role, with few small-scale elements. This is evidences by $D(q)$, which demonstrates a higher degree of curvature for negative and positive q values.

Fig. 6 illustrates the comparative dynamics of temperature fluctuations, measured from the photopolymerization process without PI, and the integrated multifractal heat capacity C_{area} calculated for polymerization with PI, without PI, and shuffled series.

Fig. 6 shows that the dynamics of C_{area} increase at the beginning and ending of light irradiation, indicating an increase in the overall degree of complexity and periodization. It can be observed in Fig. 6b that $C(q)$ for the initial time series of photopolymerization demonstrates an increase in the system complexity at the light-switching-on/off stages. Therefore, these stages are characterized by the highest degree of transformation



(a)



(b)

Fig. 6. Comparative dynamics of temperature measurements for the photopolymerization process without PI and C_{area} calculated with PI, without PI, and shuffled measurements (a). 3D representation of the multifractal heat capacity $C(q)$ dynamics for the initial (b) time series of the photopolymerization process with PI. (Color online)

processes. It can be seen from Fig. 6a that the series shuffling procedure reflects a strong difference between C_{area} for the original and shuffled series. In other words, it becomes clear that the correlated processes of free radicals birth and their participation with monomers in the combined formation of the polymer network are of great importance over the whole investigated period of the photopolymerization. That is, it is an indicator of complexity that evidences the degree of self-organization of the physical quasi-phase transition, resulting from collective dynamics of monomers and free radicals.

5. Discussion and conclusions

In this study, the multifractal properties of AESO both with PI and without it during illumination were investigated. The change in the degree of multifractality of the investigated material was verified by varying both the duration of material illumination and the light intensity.

For the first time, MF-DFA was chosen as the main method to analyze the photopolymerization process of AESO-based material, which is a sufficiently stable and accurate procedure for studying nonlinear systems of any nature and complexity.

The calculations were carried out in two stages. First, as an example, we presented the results for the entire initial series of fluctuations in the temperature of the illuminated material. Then, in order to monitor the variation of the system complexity upon further light-switches on and off, we carried out calculations using the sliding window procedure. This involved taking a fragment of polymer temperature fluctuations, conducting MF-DFA calculations for the fragment, shifting the window by a certain step, and repeating the calculations until the complete series was exhausted.

The dependences of the generalized Hurst exponent $h(q)$, the multifractal Rényi exponent $\tau(q)$, the generalized fractal dimensions $D(q)$, the multifractal spectrum $f(\alpha)$, and the multifractal heat capacity $C(q)$ were considered for the whole initial and shuffled series. For the sliding window procedure, the minimum (α_{min}) and maximum (α_{max}) singularity exponents, the information (D_1) and correlation (D_2) dimensions, the curvature of the left (ΔD_L) and right (ΔD_R) tails of the generalized fractal dimensions spectrum, and the integral value of the multifractal heat capacity C_{area} were considered. The results of the measurements for the entire series demonstrated that the nature of the AESO-based polymer is far from random: it is apparent in a high degree of ordering of the polymer network during gradual illumination and the tails of the temperature fluctuation distribution, the extent of which deviates from the normal Gaussian distribution. Each of the indices for the shuffled series was characterized by independence from the statistical q moments, when the opposite picture was observed for the initial series. Within the sliding window procedure, it was shown that all the multifractal measures respond in a particular way (increase or decrease) at the time moments when the light is switched on/off, regardless of whether or not there is PI in the material. Such dependence of the indicators suggests that the system begins to enter a self-organization state when a sufficiently large number of small-scale and large-scale processes begin to be involved.

According to the dynamics of the proposed indicators, we may assume that in the case of the light-switching-on stage, there is an active process of bond cracking, birth of free radicals, and their participation in the processes of polymer network formation. The least studied light-switching-off stage is likely to be associated with the reduction of double bonds. Recombination and termination processes may be the dominant processes at this stage, as well as at the light-switching-on stage, where they manifest themselves in the form of phase transitions. During the photopolymerization process, the kinetics of chain growth, cross-linking, and other polymerization reactions can lead to an increase in the activation energy [38] and the formation of non-uniform

structures with varying degrees of alignment or orientation. These non-uniformities in the orientation can manifest themselves as orientational defects [39] and contribute to the observed increase in multifractality. Subsequent studies will examine the photopolymerization process of the investigated material through the lens of density functional theory, with the aim of validating the aforementioned hypothesis. Furthermore, the range of methods [36, 40–45] will be expanded to explore the multifractal, irreversible, network, and correlational characteristics of the presented physical process, as well as the range of different indicators that would allow for an expanded view of the evolution of the asymmetry of the multifractal spectrum at different moments of polymer development and even its degradation.

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Authors and CV

Arnold Kiv, DSc, Professor at the South Ukrainian National Pedagogical University named after K.D. Ushynsky and Visiting Professor at the Ben-Gurion University of the Negev. Author of more than 400 publications. The area of his research interests is physical properties of nanomaterials.
E-mail: kiv.arnold20@gmail.com,
https://orcid.org/0000-0002-0991-2343

Vladimir Soloviev, DSc, Professor at the Kryvyi Rih State Pedagogical University and Professor at the South Ukrainian National Pedagogical University named after K.D. Ushynsky. Author of more than 250 publications. The area of his research interests is physical properties of nanomaterials.
https://orcid.org/0000-0002-4945-202X

Andrii Bielinskyi, PhD student at the Kyiv National Economic University named after Vadym Hetman. Author of more than 20 publications indexed by Scopus. The area of his research interests is complex systems modeling.
E-mail: bielinskyi.andrii99@gmail.com,
https://orcid.org/0000-0002-2821-2895

Mykola Slusarenko, PhD, Head of Department at the Kryvyi Rih State Pedagogical University. Author of more than 70 publications. The area of his research interests is structural properties of biomaterials.
E-mail: nick_slusarenko@yahoo.com,
https://orcid.org/0000-0003-0288-5482

Taras Kavetskyy, PhD, Associate Professor and Senior Researcher at the Drohobych Ivan Franko State Pedagogical University and Researcher at the Institute of Physics, Slovak Academy of Sciences, Bratislava. Author of more than 400 publications. The area of his research interests is physical properties of nanomaterials.
https://orcid.org/0000-0002-4782-1602

Ondrej Šauša, PhD, Researcher at the Institute of Physics, Slovak Academy of Sciences, Bratislava, and Researcher at the Department of Nuclear Chemistry, Comenius University in Bratislava. Author of more than 100 WoS publications.
The area of his research interests is physical properties of nanomaterials. E-mail: ondrej.sausa@savba.sk,
https://orcid.org/0000-0001-9958-5966

Helena Švajdlenková, PhD, Researcher at the Polymer Institute, Slovak Academy of Sciences, Bratislava, and Researcher at the Department of Nuclear Chemistry, Comenius University in Bratislava. Author of 67 WoS publications. The area of her research interests is characterization of polymer materials.
E- mail: helena.svajdlenkova@savba.sk,
https://orcid.org/0000-0002-6439-6417

Ivan Donchev, Researcher at the South Ukrainian National Pedagogical University named after K.D. Ushynsky. Author of 10 publications. The area of his research interests is physical properties of nanomaterials.
E-mail: idonchev@gmail.com

Nataliia Hoivanovych, PhD, Associate Professor at the Drohobych Ivan Franko State Pedagogical University. Authored more than 170 publications. The area of her research interests is structural properties of biomaterials.
E-mail: natahoyvan@gmail.com;
https://orcid.org/0000-0002-3442-0674

Liudmyla Pankiv, PhD, Associate Professor and Senior Researcher at the Drohobych Ivan Franko State Pedagogical University. Author of more than 40 publications. The area of her research interests is physical properties of nanomaterials.
E-mail: lyuda_pankiv@ukr.net,
https://orcid.org/0000-0002-4918-2138

Oksana Nykolaishyn, PhD, Researcher at the Drohobych Ivan Franko State Pedagogical University. Author of more than 20 publications. The area of her research interests is structural properties of biomaterials.
E-mail: nykolaishynoksana@gmail.com

Oksana Mushynska, Senior Laboratory Assistant at the Drohobych Ivan Franko State Pedagogical University. Author of 10 publications. The area of her research interests is physical properties of nanomaterials. https://orcid.org/0000-0003-1804-2615, e-mail: nokr@ukr.net

Oksana Zubrytska, PhD student at the Drohobych Ivan Franko State Pedagogical University. Author of 7 publications. The area of her research interests is physical properties of nanomaterials. E-mail: oksanazubrytska23.02@gmail.com

Andriy Tuzhykov, PhD student at the South Ukrainian National Pedagogical University named after K.D. Ushynsky. The area of his research interests is complex systems modeling.
E-mail: andrewtuzhykov@gmail.com

Mariya Kushniyazova, PhD student at the Kazakh-British Technical University. Author of 3 publications. The area of her research interests is physical properties of nanomaterials. E-mail: kshnmariya@gmail.com

Authors' contributions

Kiv A.E.: conceptualization, writing – original draft, supervision, project administration.
Soloviev V.N.: conceptualization, investigation, writing – original draft, project administration.
Bielinskyi A.O.: methodology, investigation, writing – original draft, data curation.
Slusarenko M.A.: investigation, formal analysis.
Kavetskyy T.S.: conceptualization, investigation, writing – original draft, project administration.
Šauša O.: investigation, writing – review & editing.
Švajdlenková H.: investigation, writing – review & editing.
Donchev I.I.: investigation, formal analysis.
Hoivanovych N.K.: investigation, formal analysis.
Pankiv L.I.: investigation, formal analysis.
Nykolaishyn O.V.: investigation, formal analysis.
Mushynska O.R.: investigation, formal analysis.
Zubrytska O.V.: investigation, formal analysis.
Tuzhykov A.V.: visualization, resources.
Kushniyazova M.: validation, resources.

Мультифрактальні характеристики керованої світлом самоорганізації у полімерах на основі акрилованої епоксидованої соєвої олії

А.Ю. Ків, В.М. Соловйов, А.О. Бєлінський, М.А. Слюсаренко, Т.С. Кавецький, О. Šауša, Н. Švajdlenková, І.І. Дончев, Н.К. Гойванович, Л.І. Паньків, О.В. Николаїшин, О.Р. Мушинська, О.В. Зубрицька, А.В. Тужиков, М. Кушніязова

Анотація. У цій роботі досліджено мультифрактальні властивості акрилованої епоксидованої соєвої олії з фотоініціатором та без нього. Використовуючи мультифрактальний аналіз детрендованих флуктуацій, досліджено процес фотополімеризації при зміні тривалості та інтенсивності освітлення. Дослідження проведено в два етапи: спочатку представляються результати для температурних флуктуацій всього процесу фотоопромінення, а потім використовується процедура ковзного вікна для моніторингу складності системи в ході циклічного освітлення. Для початкових і перемішаних рядів досліджено різні мультифрактальні міри, зокрема узагальнений показник Херста $h(q)$, мультифрактальний показник Реньї $\tau(q)$, узагальнені фрактальні розміри $D(q)$ з кривизною лівої і правої частин (ΔD_L, ΔD_R), мультифрактальні показники сингулярності α, мультифрактальний спектр $f(\alpha)$ і мультифрактальну теплоємність $C(q)$ з її інтегральним показником C_{area}. Емпіричні результати демонструють, що моменти включення та виключення фотоопромінення характеризуються миттєвим зростанням ступеня мультифрактальності системи. Ми припускаємо, що зростання мультифрактальності на стадіях увімкнення/вимкнення світла є квазіфазовим переходом, пов'язаним з перетворенням орієнтаційних дефектів у полімерній сітці.

Ключові слова: акрилована епоксидована соєва олія, фотополімеризація, самоорганізація, мультифрактальність, мультифрактальний аналіз детрендованих флуктуацій, фазовий перехід.