DOI 10.26724/2079-8334-2022-4-82-112-117
UDC 616.314-089.27-085.467:620.179.17

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ESTIMATION OF FRACTURE RESISTANCE OF NANOCOMPOSITE FILLING MATERIALS USING ACOUSTIC EMISSION METHOD

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The strength characteristics and features of the fracture of nanocomposite filling materials under bending using the acoustic emission method were analyzed. Nanocomposites are one of the widely used filling materials. To ensure the long-term operation of such restorations, it is important to know the dynamics of the fracture propagation in them under the influence of load. Four commercial nanocomposites were ranked by flexural strength, and modulus of elasticity using the acoustic emission method. Their fracture resistance was estimated by the analysis of acoustic emission signal parameters. Based on a matrix with triethylene glycol dimethacrylate monomer nanocomposite from a foreign manufacturer has the highest flexural strength (164.44±20.42 MPa) and resistance to fracture. Based on a matrix with ethoxylated bisphenol-A dimethacrylate monomer composite has the highest modulus of elasticity (107.46±2.95 GPa). The results of the performed studies will help to make an effective choice of filling materials to ensure the long-term operation of the corresponding restorations.

Key words: dental restorative materials, nanocomposite, fracture strength, composite resin, acoustic emission.

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ОЦІНКА СТІЙКОСТІ ДО РУЙНУВАННЯ НАНОКОМПОЗИТНИХ НАПОВНЮВАЛЬНИХ МАТЕРІАЛІВ ЗА ДОПОМОГОЮ МЕТОДУ АКУСТИЧНОЇ ЕМІСІЇ

У статті проаналізовано міцнісні характеристики та особливості руйнування нанокомпозитних пломбувальних матеріалів за згину за допомогою методу акустичної емісії. Нанокомпозити є одними з найпоширеніших пломбувальних матеріалів. Для забезпечення тривалої експлуатації таких реставрацій важливо знати динаміку розвитку руйнування в них під дією навантаження. За допомогою методу акустичної емісії ранжували чотири комерційних нанокомпозити за міцністю на згин та модулем пружності. За аналізом параметрів сигналів акустичної емісії оцінили їх опір руйнуванню. Найвищу міцність на згин (164,44±20,42 МПа) та опір руйнуванню має нанокомпозит на основі матриці з мономером триетиленглікольдиметакрилат зарубіжного виробника. Найвищий модуль пружності (107,46±2,95 ГПа) має композит на основі матриці з мономером етоксильований бісфенол-А диметакрилат. Результати виконаних досліджень допоможуть здійснювати ефективний вибір пломбувальних матеріалів для забезпечення довготривалої експлуатації відповідних реставрацій.

Ключові слова: стоматологічні матеріали, нанокомпозит, міцність руйнування, композитний полімер, акустична емісія

This study is a fragment of the research project "Improvement and development of new methods of diagnosis and treatment of patients with defects, deformities and functional disorders of the dento-maxillary system", state registration No.0119U002102.

Among the large number of materials used for tooth restoration, dental composites are one of the most widely used. They best satisfy the basic principles of biomimetics in dentistry – have balanced luminosity, translucency, opacity, and wear resistance of artificial restorations of teeth [13]. The range of composite materials presented in the dental market is very wide: hybrid, nano-hybrid, micro-filled, packable, ormocer-based, and flowable composite as well as compomers and flowable compomers [5]. Composite resin restorations are influenced by mechanical properties, such as flexural strength and flexural modulus of elasticity. When choosing one or another material, it is important to know its behavior during the exploitation of the restoration, namely the initiation and propagation of fracture under the action of bending loads.

The acoustic emission (AE) method, which is based on the registration of elastic waves arising because of the formation, change, and failure of the structure of various materials, is currently the most effective for studying the processes and stages of the development of defects in their structure [11]. It can detect the origin of fracture, the initial location of defects, and their propagation, accurately determine the maximum strength of materials, and identify the mechanisms of fracture. The advantages of the method include the possibility of obtaining information about the fracture already in the early stages, as well as its high sensitivity, as this method makes it possible to detect even small defects.

The AE method was usually used to study the fracture resistance of various composite materials [3], to study the tooth/composite interface [2, 8, 12], and to analyze polymerization stresses and the degree of shrinkage [4, 7, 9].

Thus, as the analysis of literary sources shows, the AE method serves as an effective tool for real-time monitoring of the stages of fracture of dental composites.

To date, we are not aware of any publications devoted to the study of the initiation and fracture propagation in nanocomposites for dental fillings under the action of bending loads.

The purpose of the study was to determine the strength characteristics (flexural strength and modulus of elasticity) and features of fracture of light-cured nanocomposite materials for teeth restoration under three-point loading using the acoustic emission method.

Materials and methods. To perform this study, four different nano-hybrid dental composites for dental fillings were used: Latelux (Latus, Ukraine; resin matrix: BisGMA, UDMA, TEGDMA; inorganic filler parties of (0.02–3 μm)); Tetric N-Ceram (Ivoclar Vivadent, Liechtenstein; resin matrix: BisGMA, UDMA, TEGDMA; filler: Ba glass, Ba-Al-F-Si glass, YbF_3, SiO_2, MO (0.04–3 μm)); Charisma Classic (Kulzer, Germany; resin matrix: BisGMA, TEGDMA; filler: Ba-Al-Fe glass, SiO_2 (0.01–2 μm)); FILTEK Z250 (3M ESPE, USA; resin matrix: BisGMA, BisEMA, UDMA; filler: ZrO_2/SiO_2 (0.01–3.5 μm)).

For the three-point flexural strength test, 10 bar-shaped specimens with a length of 25 mm, a width of 2 mm, and a height of 2 mm were fabricated from each composite resin, following the manufacturers' instructions and ISO4049 [6]. The material was packed and shaped into a special mold under laboratory conditions at an air temperature of 18–21 C. In the cells of the mold, an appropriate light-curing composite material was introduced in small portions with subsequent condensation to the bottom and walls of the cell with the use of a dental filling plugger for composites (type Ladmore, TNBBL2, manufacturer: Hu-Friedy).

Next, polymerization of the corresponding material was carried out using ultraviolet radiation with a wavelength of 420–480 nm and a light intensity of 2.0–2.2 mV/cm^2 (TURBO Program), using an LED wireless photopolymer lamp Bluephase 20i (G2) (Invoclar Vivadent).

After polymerising the material in the cells, the samples were taken out of the cuvette. Within the following 20 seconds, each of the surfaces was exposed to light once again.

The samples were carefully sanded with abrasive paper (320 grit abrasive) until the appearance of a dry surface shine, after which each of them was visually inspected to identify possible defects in the thickness and defects polymerization and structure, which could lead to false results during the tests [6]. Before the tests, the samples were stored in a physiological solution at a temperature of 37^0 C for 24 h.

Specimens were submitted to the three-point bending test in a SVR-5 testing machine (Karpenko Physico-Mechanical Institute of the NAS of Ukraine) at a crosshead speed of 0.5 mm/min until fracture.

The two-channel AE detection system SKOP-8 (Karpenko Physico-Mechanical Institute of the NAS of Ukraine [11]) was used to record the AE activity while testing the specimens. The recordings of AE and mechanical load were synchronized in time. AE measurement conditions of 40 dB pre-amplifier, the threshold of discrimination within 30 %, and a 4 MHz sampling rate were adopted. Band-pass filtering of 75–600 kHz was performed. The relative error of AE signals amplitude registration is below ±10 %.

The maximum fracture load (*F*, in N) of each specimen was recorded, and the flexural strength (σ_f), in MPa, was calculated as follows:

$$\sigma_f=3Fl/2bh^2 \quad (1)$$

and the modulus of elasticity (*E*), in MPa, was calculated as follows:

$$E=3Fl^3/2bh^3\mathrm{D} \quad (2)$$

where *l* is the distance between the supporting rollers (20 mm); *b* is the specimen width (~2 mm); *h* is the specimen height (~2 mm); *D* is the deformation of the sample for *F*.

To evaluate AE signals during the fracture of composite resins used for teeth restoration, the method proposed in [10, 11] was used, which involves the analysis of the energy E_{WT} of local maxima of the continuous wavelet transform (CWT) of AE signals characterizing each elementary act of fracture.

Results were statistically analyzed using analysis of variance (ANOVA). Post hoc Tukey's HSD multiple comparison tests (p=0.05) were used to find statistically significant differences between the different composites.

Results of the study and their discussion. Fig. 1 shows the typical dependencies between load change and distribution of AE signal amplitudes over time.

It was determined that during the loading of samples of restorative composites, the fracture progresses differently in different materials. The highest AE activity was observed in Latelux and Charisma Classic, which indicates that destruction in such composites starts already at the early stages of loading.

Filtek Z250 also demonstrated insignificant AE activity. On the contrary, in Tetric N-Ceram, AE signals were generated when the failure load was reached indicating its high structural homogeneity and fracture resistance.

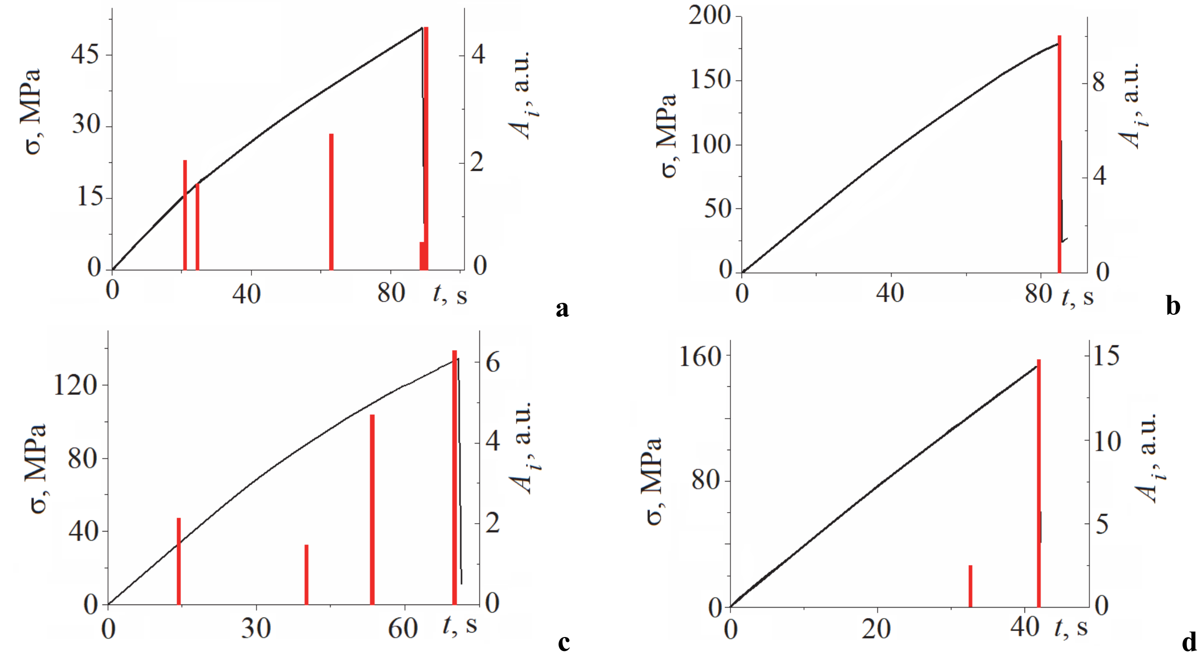

Fig. 1. Typical dependencies between load change and distribution of AE signal amplitudes over time (1 a.u. = 1 mV): *a* – Latelux; *b* – Tetric N-Ceram; *c* – Charisma Classic; *d* – Filtek Z250.

We can see that the time it takes to fracture the samples completely is different. In particular, the average time it takes to completely fracture Tetric N-Ceram samples with the highest strength and Latelux samples with the lowest strength differs insignificantly and is 85±15 s. For other composites, it is as follows: Charisma Classic – 75±12 s, FiltekZ250 – 42±10 s.

The flexural strength and modulus of elasticity of the materials were determined by Eq. 1 and 2, respectively (Table 1).

Table 1

Mean and standard deviation values (SD) of flexural strength σ_f and modulus of elasticity *E* of restorative nanocomposites

Material	σ_f, MPa mean±SD	*E*, GPa mean±SD
Latelux	60.75±9.24	19.80±5.26
Tetric N-Ceram	164.44±20.42	57.59±2.88
Charisma Classic	141.19±9.81	60.10±17.19
Filtek Z250	129.38±33.94	107.46±2.95

The flexural strength of Latelux is statistically significantly different from other materials ($p<0.01$). At the same time, between the flexural strength of the composites Tetric N-Ceram and Charisma Classic (p>0.2), Charisma Classic and Filtek Z250 (p>0.09), Tetric N-Ceram and Filtek Z250 (p>0.8), this difference not significant.

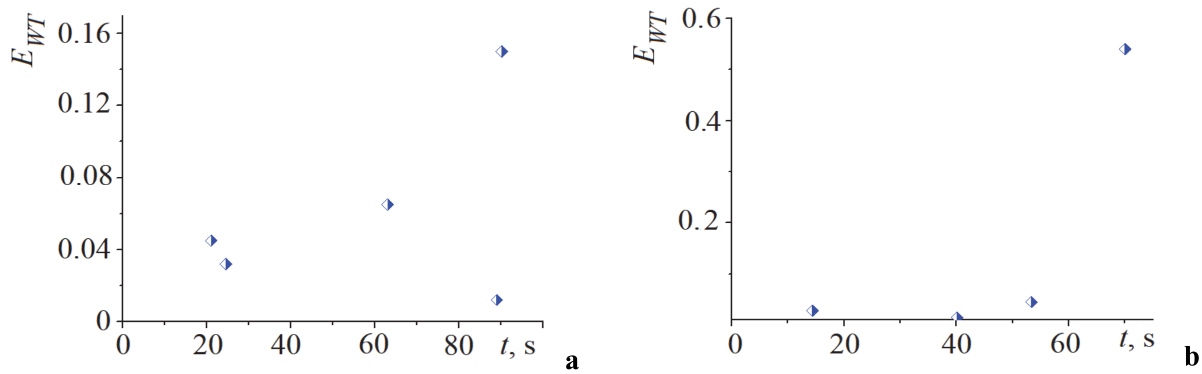

Fig. 2. Typical time distributions of AE signals by energy parameter during three-point bending of samples of restorative composites: *a* – Latelux; *b* – Tetric N-Ceram.

The modulus of elasticity of Filtek Z250 is statistically significantly different from Latelux (p<0.01) and Charisma Classic and Tetric N-Ceram (p<0.05). At the same time, this difference is not significant between the elastic modulus of Latelux and Tetric N-Ceram (p>0.1), Latelux and Charisma Classic (p>0.5), Tetric N-Ceram and Charisma Classic (p>0.7).

To study the dynamics of fracture of dental composites, the AE signals recorded during the experiments were analyzed according to the energy parameter E_{WT}. Fig. 2 shows the typical time distributions of AE signals by energy parameter for Latelux and Charisma Classic.

We can see that as the load increases, AE signals are mainly generated due to the propagation of micro-cracks, and as it approaches its maximum value, a big macro-crack forms, which eventually causes the sample to split into two parts.

The ranges of numerical values of the energy parameter E_{WT} for different types of fracture in restorative nanocomposites were determined (Table 2).

Table 2

The range of values of the energy parameter E_{WT} for nanocomposites

Material	Type of fracture	
	propagation of micro-cracks	propagation of macro-cracks
Latelux	$0.012 \leq E_{WT} \leq 0.045$	$0.1 \leq E_{WT} \leq 0.18$
Tetric N-Ceram	–	$0.1 \leq E_{WT} \leq 1.5$
Charisma Classic	$0.015 \leq E_{WT} \leq 0.065$	$0.1 \leq E_{WT} \leq 0.6$
Filtek Z250	$0.011 \leq E_{WT} \leq 0.052$	$0.1 \leq E_{WT} \leq 2.0$

We would like to note that during the propagation of micro-cracks in Charisma Classic, AE signals with greater energy – and therefore amplitude – were generated than in Latelux and Filtek Z250. It is known that the amplitude of an AE signal is directly proportional to the area of the newly formed fracture surface. Therefore, under the load micro-cracks of a slightly larger size than in other materials were formed in Charisma Classic. The increase in the number of micro-cracks in Filtek Z250 was accompanied by AE signals of the highest energy, which confirms its greater embrittlement.

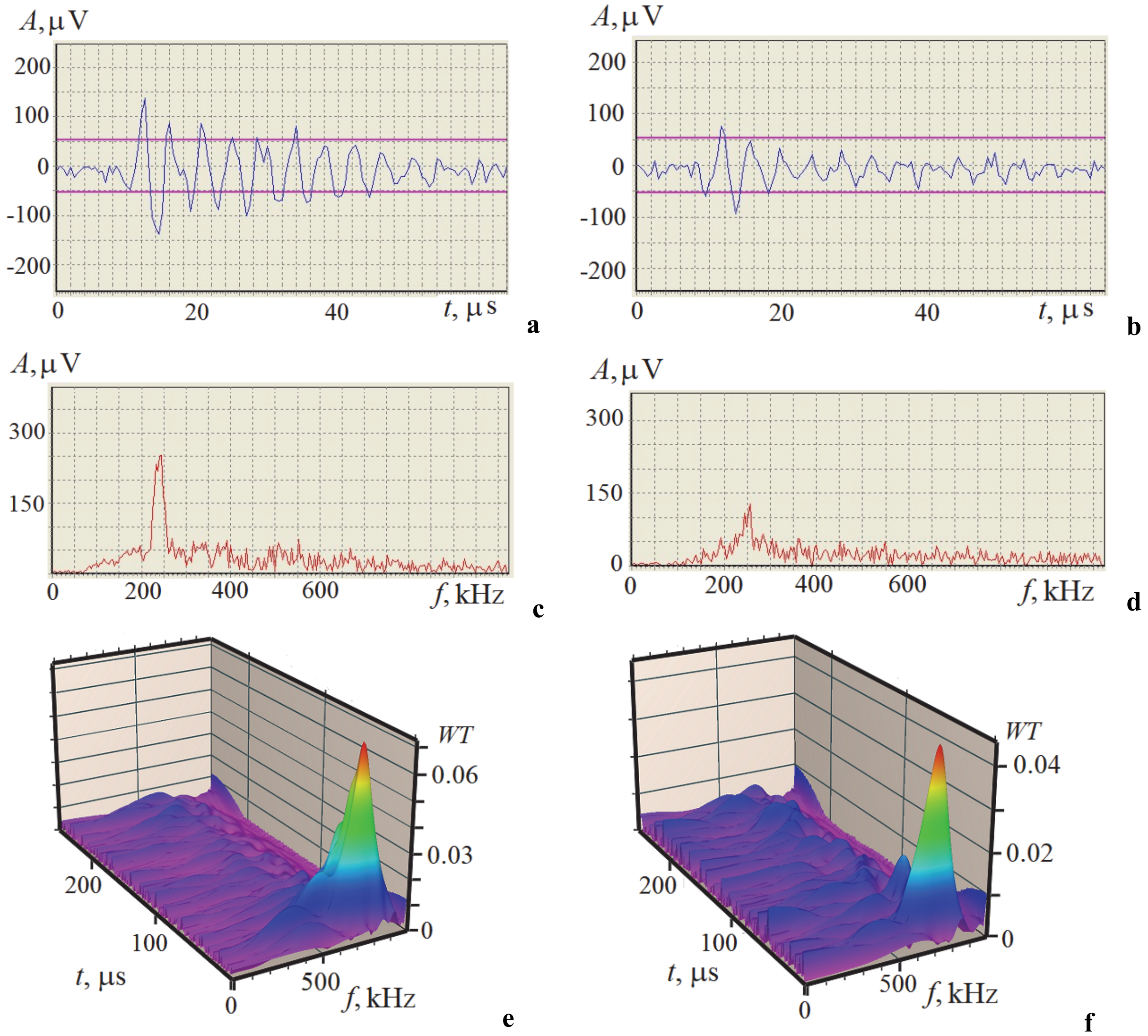

Fig. 3. Waveform (*a*, *b*), spectral distribution (*c*, *d*), and CWT (*e*, *f*) of typical AE signals that were generated during the propagation of micro-cracks in the following materials: Latelux (*a*, *c*, *e*) and Charisma Classic (*b*, *d*, *f*).

Fig. 3 shows the waveforms, spectral distribution, and CWT of typical AE signals that accompanied the fracture of composites at different stages.

It was determined that during the propagation of micro-cracks, the CWT of AE signals has one local maximum of the same frequency range, which indicates the dominance of the exact fracture mechanism. The values of the maximum wavelet coefficient in the CWT differ for different materials, which is associated with forming of new defects of larger or smaller sizes in different materials.

The dominance of a local maximum of the lower frequency range CWT is the peculiarity of the AE signals that were generated during the fracture of Filtek Z250 (fig. 4).

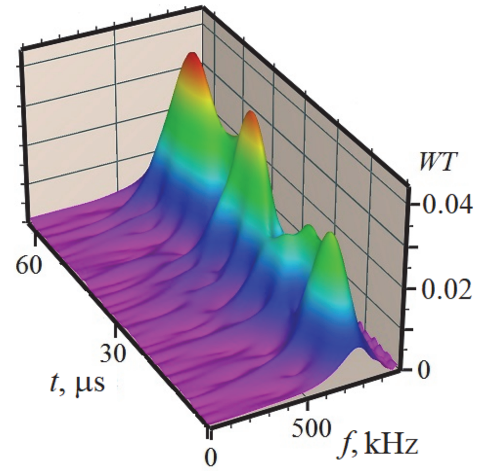

Fig. 4. The CWT of typical AE signals that were generated at the initial stages of Filtek Z250 fracture.

Considering the mechanical parameters of this material and the results of AE analysis, Filtek Z250 is the most brittle among the examined composites, which shall be considered when choosing a suitable material for restorative works.

According to the spectral analysis of AE signals, it was established that the formation of micro-cracks in polymers was accompanied by AE signals with a frequency range of 220–260 kHz, whereas propagation of macro-cracks was accompanied by AE signals with a frequency range of 120–150 kHz.

The nature of the fracture of Latelux, Tetric N-Ceram, and Charisma Classic materials under bending load is elastic-plastic at the initial stage, with a transition to brittle as the load increases. On the contrary, the fracture of the Filtek Z250 was brittle.

Composite resins are widely used to restore both front and back teeth due to the development of their aesthetic and mechanical properties. Any restorative dental material, as well as natural teeth, must have sufficient mechanical integrity to function in the oral cavity for a long period of time. Considering this, studying the mechanical properties of these materials is an extremely important clinical task.

Our study aims to compare four nanocomposites in terms of their mechanical behavior. For this purpose, the flexural strength and modulus of elasticity of the composites were evaluated, and the dynamics of material fracture under quasi-static loading were studied using the AE method [3, 5].

According to the results of the performed research on flexural strength, dental composites were arranged in the following order (from the largest to the smallest): Tetric N-Ceram (164.44 MPa) > Charisma Classic (141.19 MPa) > Filtek Z250 (129.38 MPa) > Latelux (60.75 MPa).

A somewhat different sequence was obtained for the value of the modulus of elasticity of the studied materials (from the largest to the smallest): Filtek Z250 (107.46 GPa) > Charisma Classic (60.10 GPa) > Tetric N-Ceram (57.59 GPa) > Latelux (19.80 GPa).

The obtained results are consistent with those known in the literature [1]. In terms of flexural strength and modulus of elasticity, the studied materials meet modern requirements for the strength of hybrid materials, and Tetric N-Ceram has the best indicators. It can be recommended for the restoration of teeth that are subjected to the greatest chewing loads.

The character of the fracture under the bending load of Latelux, Tetric N-Ceram, and Charisma Classic is elastic-plastic at the initial stage, with a transition to brittle as the load increases. Instead, Filtek Z250 was brittle. This must be considered when choosing a material for tooth restoration in different jaw areas since the restorations are subjected to different loads. Filtek Z250 may be recommended for the restoration of teeth that are subjected to the smallest chewing loads.

The analysis of AE signal parameters has shown that the number of micro- and macro-cracks in dental composites grows under bending load. Signals that accompanied the formation of micro-cracks had the highest amplitude and energy in the Charisma Classic whereas the lowest amplitude and energy were observed in the Latelux, which indicates the formation of slightly larger micro-cracks in the Charisma Classic than in other materials. Tetric N-Ceram, in which the micro-cracks propagation started already on the verge of fracture strength, showed the highest fracture resistance. Propagation of micro-cracks in the Filtek Z250 material was accompanied by AE signals of the highest energy, which confirms its greater embrittlement.

Conclusions

1. This study made it possible to rank nanocomposites by flexural strength and modulus of elasticity. Tetric N-Ceram nanocomposite has the highest flexural strength (164.44±20.42 MPa), and Latelux has the lowest one (60.75±9.24 MPa). Filtek Z250 nanocomposite has the highest modulus of elasticity (107.46±2.95 GPa), and Latelux (19.80±5.26) has the lowest modulus.

2. Tetric N-Ceram has the highest resistance to fracture, and Filtek Z250 was brittle.

3. The performed studies allow us to recommend the use of Tetric N-Ceram nanocomposite for the restoration of teeth that are subjected to the greatest chewing load and Filtek Z250 – to the smallest chewing load. The results of studies will help to make an effective choice of filling materials to ensure the long-term operation of the corresponding restorations.

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Стаття надійшла 17.10.2021 р.