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Research article

METHODICAL APPROACHES TO RAISING THE RELIABILITY OF HEALTH RISK ASSESSMENT WHEN USING POLYMER MATERIALS IN DRINKING WATER SUPPLY

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Plastic pipes and coatings may contain additives, including metal stabilizers and antioxidants, designed to protect the material during manufacture and use. Some chemical compounds can be released from these plastic pipes and affect quality of drinking water. The article focuses on analyzing various approaches to examining polymer materials with the aim to assess migration of chemicals into drinking water. These approaches usually underlie methodologies of hygienic assessment developed for polymers.

Migration was assessed under the same conditions as per two types of migration processes, a continuous and a sequential one. These two types of migration processes emulate conditions typical for different flows in drinking water pipelines: situations of continuous stagnation in the system and a standard flow when water is renewed regularly in water supply networks. More than 20 organic compounds were identified in tested water samples. Most of them occurred in small concentrations (excluding benzenesulfonic acid butyl amide). Moreover, many of these chemicals are not regulated in drinking water, there no standards or reference concentrations fixed for them or a relevant toxicological assessment. Given that, it is practically impossible to assess health risks caused by exposure to these chemicals according to conventional assessment procedures.

It was also shown that release of chemicals differed considerably under different experimental designs. The results produced by successive migration tests indicated that intensity of migration from polymer materials the pipes were made from tended to change over time whereas the results of continuous migration tests showed that in case of stagnation quality of drinking water could deteriorate rather rapidly due to migration of organic compounds.

Keywords: *water supply, drinking water, hygienic assessment of polymer materials, polymers, migration, polyurethane coatings, chromato-mass spectrometric studies, water risk.*

Polymer pipes and polymer coatings protect inner surfaces of pipelines; they are often used to repair and reconstruct the existing drinking water supply systems, including main and distribution water supply networks since they are cheaper and easier to install in comparison with metallic alternatives. Unfortunately, plastic pipes and coatings can contain various additives such as metal stabilizers, plasticizers and other antioxidants that protect the materials they are made of during manufacturing and utilization. Since a plasticizer molecule or a molecule of any other additive is usually not chemically bound to a polymer chain, it can be released when a polymer is being manufactured or later during its routine use. Migration of plasticizers during a polymer product life span can contaminate the environment and do damage to human health [1–7].

In time, contamination of drinking water with organic compounds is going to become a more pressing issue due to wider use of up-to-date polymer coatings in pipelines. At present, such toxic environmental contaminants as semi-volatile organic compounds

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(SVOC) already occur everywhere [8–10]. The study [11] describes an established considerable correlation between SVOC contents in drinking water and a scale of economy and population in cities in China, which means that the problem is aggravated by the scientific and technical progress in general and developing chemical industry in particular. However, occurrence of semi-volatile chemical compounds in tap water and associated human health risks have not been studied enough [12], despite the fact that these compounds can produce negative effects on human health even in low concentrations. To assess this exposure and calculate associated risks, relevant toxicity tests are required. However, data on toxicity are not available for many compounds and even if they are, it takes specific searching in literature sources to find them.

Field observations can give an opportunity to trace migration of organic compounds into drinking water [13–20]. Migration of pollutants into tap water is potentially influenced by various housing types since they tend to vary in terms of water supply and distribution system, water consumption choices and other hydraulic factors. Contents of organic compounds in tap water turned out to be greatly dependent, among other things, on whether housing was high-rise or landed [18]. Migration of plastic organic compounds falls considerably during the first several months of water supply exploitation. However, a period during which drinking water is stagnated in pipes also has great influence on contents of organic compounds. Organic compounds migrate from materials into drinking water in higher quantities as a stagnation period becomes longer. The authors of the work [19] considered it necessary to create an instruction for users stating the necessity to drain certain amount of water prior to drinking it.

Although, a conventional way to reduce pollutant concentration in drinking water is to replace it with fresh one, there are still disputes on the issue and the issue itself has not been given proper attention in scientific literature. All this makes it difficult to predict what volume of water will be necessary to reduce the secondary organic pollution.

Results described in the work [20] indicate that it is likely to take several days of continuous flushing to remove pollutants in quantities sufficient to reduce their levels below the hygienic standards.

At present, mathematical molecular dynamic simulation of diffusion can be used to calculate theoretical migration of additives for various surface-to-volume ratios and concentrations of compounds introduced into a polymer on the surface of the material. Precise data on coefficients showing diffusion of additives in a polymer make it possible to predict migration into drinking water over a certain period [21–23]. It was shown in several previous domestic studies that migration decreased over time as per an exponential curve[1]. It was established that chemical compounds were washed out most intensively from polymer materials during the first days of a contact with water; later on, levels of toxicant release decreased.

Studies on chemical migration and calculation of diffusion coefficients for additives in a polymer give a possibility to predict probable secondary pollution in drinking water. Analysis of the results gives grounds for developing various approaches to hygienic assessment of polymer materials. It is a rather serious methodical issue since correct assessment of exposure to a hazardous matter in water is important for interpreting its results and for further health risk assessment as a vital method of hygienic assessment. The matter is that multiple different factors can change composition, concentration, and, consequently, toxicity of a mixture of migrating compounds and can ultimately influence the precision of prediction techniques.

[1] Sheftel V.O., Dyshinevich N.E., Sova R.E. Toksikologiya polimernykh materialov [Toxicology of polymer materials]. Kiev, Zdorov'ya, 1988, 210 p. (in Russian).

These factors complicate assessment of health and environmental risks caused by these compounds. Besides, any studies on assessment of risks created by a mixture of compounds will unavoidably face insufficient toxicological data and gaps in knowledge about synergic effects produced by a mixture of pollutants [24].

Similar conclusions were made by the authors who assessed water risks [25]. They concluded that most compounds never occur in concentrations that individually create an appreciable human health risk. However, they noted that several detected substances might still impose certain health hazards, for example, vinyl chloride, trichloroethylene, bromodichloromethane, phenol, 2-chlorobenzylamine, and some others. For part of the selected substances, toxicological risk assessment for drinking water could not be performed since no data on their toxicity were available. In case the necessary data were absent, the authors suggested using "Threshold of Toxicological Concern" (TTC) to assess screening level risks.

Turning back to correct assessment of exposure to a matter in water, we should note that in such countries as Great Britain, Germany and some others, hygienic assessment of polymer materials is performed after water change. This assessment involves analyzing water extracts from two- or three-day contacts between a material and drinking water. That is, the assessment is based on analyzing a subsequent migration process that simulates a normal (standard) water flow with regular water renewal in water supply networks.

In Russia, according to the methodical guidelines MU 2.1.4.2898-11 "Sanitary-epidemiological examinations (tests) of materials, reagents and equipment used for water treatment and preparation"[2], hygienic assessment on materials used in drinking water supply is based on examining a continuous migration process that simulates a continuous stagnation of water in a water supply network.

Therefore, **our research goal** was to examine and compare different approaches to hygienic assessment of polymer materials with its focus on chemical migration into drinking water. Migration was estimated under similar conditions as per two types of migration processes, a continuous and subsequent one. These two types emulate conditions typical for different flows in drinking water pipelines: continuous stagnation in the system and a normal flow when water is regularly renewed in drinking water supply networks.

Materials and methods. The study involved examining a two-component polyurethane coating manufactured in a European country. According to the covering technical documentation, this material is allowed for use in drinking water supply in a country of origin. According to a manufacturer's manual, this coating is used in main pipelines with a pipe diameter varying from 100 to 610 mm.

The samples prepared for analysis were smooth and solid but still elastic grey plates, 0.3 cm thick, without any smell.

We ran two series of tests to confirm our previous assumptions [26, 27]. The first series concentrated on a continuous migration process. The two-component polyurethane coating was assessed bearing in mind the Unified Requirements (The EAEU Unified sanitary-epidemiological and hygienic requirements to

[2] MU 2.1.4.2898-11. Pit'evaya voda i vodosnabzhenie naselennykh mest. Sanitarno-epidemiologicheskie issledovaniya (ispytaniya) materialov, reagentov i oborudovaniya, ispol'zuemykh dlya vodoochistki i vodopodgotovki / utv. Rukovoditelem Federal'noi sluzhby po nadzoru v sfere zashchity prav potrebitelei i blagopoluchiya cheloveka, Glavnym gosudarstvennym sanitarnym vrachom Rossiiskoi Federatsii; vved. v deistvie 12.07.2011 (vzamen MU 2.1.4.783-99) [Drinking water and drinking water supply in settlements. Sanitary-epidemiological examinations (tests) of materials, reagents and equipment used for water treatment and preparation, approved by the Head of the Federal Service for Surveillance over Consumer Rights Protection and Human Well-being, the RF Chief Sanitary Inspector; introduced on July 12, 2011 (as a replacement to the MU 2.1.4.783-99)]. *KODEKS: electronic fund for legal and reference documentation*. Available at: https://docs.cntd.ru/document/1200089967 (February 11, 2022) (in Russian).

goods that are subject to sanitary-epidemiological surveillance (control)"[3] (2010)). We also examined certain indicators that were not mandatory in the assessment of polymer materials used in drinking water supply. The samples were preliminarily prepared in accordance with the methodical guidelines MU 2.1.4.2898-11 "Sanitary-epidemiological examinations (tests) of materials, reagents and equipment used for water treatment and preparation"[2].

The second series involved using a procedure accepted by the DWI (Drinking Water Inspectorate, Great Britain) as a model one to examine a continuous migration process. The samples were washed with running water for one hour and then were submerged into water for testing. The following periods of contacts with water (ST) were applied: ST1, 1 hour; ST2, 23 hours; ST3, 24 hours; ST4, 24 hours; ST5, 72 hours; ST6, 72 hours. After each period of a contact with water (ST) was over, a sample was taken out and placed into fresh water (distillate) for the next period. The samples from ST2, ST3 and ST6 were analyzed.

A ratio between volume of a material and volume of contacting water was 1 cm^2 to 1 cm^3. Tap water from a distribution network in Moscow and distilled water were used as initial water to prepare water extracts (test samples). The water extracts were kept under 20 ^oC. The same water types were applied as reference samples in order to perform proper hygienic assessment. Test samples (water extracts) and reference samples used in the first series of the study were taken on the 1^{st}, 3^{rd}, 5^{th}, 15^{th} and 30^{th} day of the experiment.

The water extracts were analyzed to determine their organoleptic properties and to examine migration of organic compounds and metals. Organoleptic properties of water extracts (smell, taste, color and turbidity) were examined with physical and chemical methods; we also determined water pH and permanganate oxidation.

Tests aimed at identifying and quantifying hardly volatile organic compounds in water were performed by using chromatography-mass-spectrometry. This method allows reliable identification and quantification of a wide range of organic compounds C1-C40 in water with unknown pollution structure with the sensitivity at the level of hygienic standards or even below them. The examination was performed on Focus GC chromatography-mass-spectrometer with DSQ II (USA) in conformity with the existing methodical documents on control over organic compounds. We also identified several substances that were mandatory within this hygienic assessment according to the Unified sanitary requirements. These substances were formaldehyde, acetaldehyde, methanol and ethylene glycol.

Non-organic compounds were analyzed in the water extracts according to the State Standard GOST 31870-2012[4] by using atomic spectroscopy.

Results and discussion. In the first series of tests, the examined material practically did not make any changes into smell or taste of a water extract. Turbidity and color of Moscow tap water did not grow considerably after contacts with it. We also did not detect any differences between water pH of the test and reference samples.

Having analyzed data on probable migration of organic compounds from the examined material, we revealed that permanganate oxidation grew in the test water extract practically in the same way as in the reference samples and did not exceed hygienic standards.

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[3] Edinye sanitarno-epidemiologicheskie i gigienicheskie trebovaniya k produktsii (tovaram), podlezhashchei sanitarno-epidemiologicheskomu nadzoru (kontrolyu) (s izmeneniyami na 22.02.2022) / utv. Resheniem Komissii tamozhennogo soyuza ot 28.05.2010 № 299 [The EAEU Unified sanitary-epidemiological and hygienic requirements to goods that are subject to sanitary-epidemiological surveillance (control) (last edited on February 22, 2022), approved by the Decision of the Customs Union Commission on May 28, 2010 No. 299]. *KODEKS: electronic fund for legal and reference documentation.* Available at: https://docs.cntd.ru/document/902249109 (March 13, 2022) (in Russian).

[4] GOST 31870-2012. Drinking water. Determination of elements content by atomic spectrometry methods: inter-state standard, approved by the Inter-state Council on Standardization, metrology and certification (the meeting report issued on November 15, 2012 No. 42); became valid on January 01, 2014. *KODEKS: electronic fund for legal and reference documentation*. Available at: https://docs.cntd.ru/document/1200097409 (March 13, 2022) (in Russian).

To analyze migration of organic compounds, distilled water under 20 °C was used as initial water to prepare water samples. Tests aimed at identifying and quantifying organic compounds in water were performed by using chromatography-mass-spectrometry on the 1st, 2rd and 5th day of the experiment. We identified up to 27 organic compounds in the water extracts from the samples of the examined two-component polyurethane coating (Table 1).

Table 1

Organic compounds identified in water extracts from the analyzed polyurethane-based material (under 20 °C) when a continuous migration process was examined

№	Compound	Gross formula	CAS	Concentration, mg/dm^3		
				1st day	3rd day	5th day
1	Tetradecane	$C_{14}H_{30}$		0.003	-	0.004
2	Hexadecane	$C_{16}H_{34}$		0.002	0.004	0.004
	spirits					
3	Hexadecanol	$C_{16}H_{34}O$	36653-82-4	0.003	-	0.003
4	Heptadecanol	$C_{17}H_{36}O$		0.006	-	-
	phenols					
5	4,4-isopropylidene diphenol	$C_{15}H_{16}O_2$	80-05-7	0.003	0.006	-
6	2,6-Di-tert-butyl-4-ethylphenol	$C_{16}H_{26}O$	4130-42-1	0.004	-	-
7	Para-tert-butylphenol	$C_{10}H_{14}O$	98-54-4	0.005	0.002	
8	2,4-Di-tert-butylphenol	$C_{14}H_{22}O$	96-76-4	-	0.003	
	simple ethers					
9	2,5,8,11- Tetraoxadodecane *	$C_8H_{18}O_4$	112-49-2	0.056	0.016	-
10	Pentaoxapentadecane	$C_{10}H_{22}O_5$	-			0.023
	ketones					
11	Benzophenone	$C_{13}H_{10}O$	119-61-9	0.004	0.004	0.008
	carboxylic acids					
12	Tetradecane	$C_{14}H_{28}O$	544-63-8	-	0.005	0.004
13	Hexadecane	$C_{16}H_{32}O$	57-10-3	0.011	0.019	0.016
	complex ethers					
14	Dimethylethyl methyl propanol ether of 2-methylpropanoic acid	$C_{16}H_{30}O_4$	74381-40-1	0.005	0.014	0.011
15	Diisobutyl ether of adipic acid	$C_{14}H_{26}O_4$	141-04-8	0.030	0.025	0.014
16	Dimetoxy tetraethylene glycol*	$C_{10}H_{22}O_5$	143-24-8	0.018	0.64	**0.149**
17	Methyl ether of tetrapropylene glycol *	$C_{13}H_{28}O_5$	20324-34-9	0.012	-	-
18	Dimethyl ether of hexaethylene glycol*	$C_{14}H_{30}O_7$	1072-40-8	0.035	0.050	**0.227**
19	Acetyl tributyl citrate	$C_{20}H_{34}O_8$	77-90-7	-	-	0.022
	phthalates					
20	Dimethyl phthalate	$C_{10}H_{10}O_4$	131-11-3	0.001	0.003	-
21	Diisobutyl phthalate	$C_{14}H_{26}O_4$	84-69-5	0.032	0.026	0.033
22	Dibutyl phthalate	$C_{14}H_{26}O_4$	84-69-2	0.021	0.019	0.078
23	Monobutyl phthalate	$C_{12}H_{14}O_4$	131-70-4	0.063	0.023	0.084
	Nitrogen-containing compounds					
24	Acridine	$C_{13}H_9N$	260-94-6	0.006	0.003	0.014
25	1,2,3,4,5,6,7,8-Octahydroacridine	$C_{13}H_{17}N$	1658-088-8	0.003	0.003	0.003
26	4-Piperidinyl-1-methyl-4(2-methyl-2-propenyl)*	$C_{10}H_{19}NO$	-	0.034	-	-
	Nitrogen-, sulfur containing compounds					
27	N-Butylbenzenesulfonamide	$C_{10}H_{15}NO_2S$	3622-84-2	0.044	0.075	**0.123**

Note: * means identification is conventional.

Some of the identified compounds were detected in insignificant concentrations; most of them do not have maximum permissible levels in drinking water established for them by regulatory documents. These compounds are mostly oxygen-containing ones, including ketones, ethers and phthalates. The following compounds were detected in the highest concentrations: dimethyl ether of hexaethylene glycol, 0.227 mg/l, and dimetoxy tetraethylene glycol, 0.149 mg/l. These compounds were identified conventionally and there are no established MPCs for them in drinking water (ethylene glycol MPC is 1 mg/l). Besides, nitrogen-containing compounds were identified in insignificant concentrations; we also identified a nitrogen- and sulfur-containing compound, namely N-Butylbenzenesulfonamide (CAS No. 3622-84-2) in a concentration higher than its maximum permissible one, 0.123 mg/l (4.1 MPC). N-Butylbenzenesulfonamide is standardized as per a sanitary-toxicological harmfulness factor and assigned into the 2nd hazard category.

Non-organic compounds were analyzed in a water extract on the 30th day in the experiment (an extract from the analyzed material on distilled water under 20 °C). The analysis revealed that toxic elements belonging to I and II hazard category such as lead, nickel, cobalt and lithium practically did not migrate into water.

We also analyzed such organic pollutants as acetaldehyde and formaldehyde in water extracts. Acetaldehyde migrated in quantities equal to 0.75 MPC and the other identified pollutants occurred in water in even smaller quantities.

As for metals, only chromium and cadmium were detected in water extracts and their concentrations were insignificant, 0.0005 mg/l (0.005 MPC) and 0.00003 mg/l (0.03 MPC) accordingly.

We identified 21 organic compounds (Table 2) in water extracts from the samples of the analyzed polyurethane coating in the second series of tests (when a subsequent migration process was analyzed). All the compounds were detected in insignificant quantities. Their migration was estimated as stable or declining. N-Butylbenzenesulfonamide was detected in concentrations equal to 0.023 mg/l, 0.006 mg/l, and 0.005 mg/l, that is, lower than MPC fixed for this compound and this indicated the migration was declining.

We also determined several substances that were mandatory within this hygienic assessment according to the Unified sanitary requirements. These substances were formaldehyde, acetaldehyde, methanol and ethylene glycol. Acetaldehyde migrated in quantities equal to 0.25 MPC (in ST2 and ST3 periods), all the other analyzed compounds were detected in even lower concentrations.

The analysis of non-organic compounds in water extracts showed that toxic elements belonging to the I and II hazard category and metals that could affect organoleptic properties of water practically did not migrate from the analyzed material.

Conclusion. A potential increase in human health risks caused by drinking tap water that is polluted with organic compounds migrating from plastic is a major challenge in hygienic assessment of polymer materials.

On one hand, a trend to replace metal pipelines with polymer ones in quite justified since the latter are cheaper, easier to install, not prone to corrosion, etc.

On the other hand, it is practically impossible to avoid migration of organic compounds from plastics into drinking water given the contemporary development of chemical industry. There are ongoing studies on developing new plasticizers, searching for new compounds that can secure good mechanic properties of a material with simultaneous limited or even zero migration, resistance to extraction and low volatility[5]. Still, at present, as it is also obviously confirmed by our research results, it

[5] R 1.2.3156-13. Otsenka toksichnosti i opasnosti khimicheskikh veshchestv i ikh smesei dlya zdorov'ya cheloveka: rukovodstvo / utv. vrio Glavnogo gosudarstvennogo sanitarnogo vracha Rossiiskoi Federatsii ot 27 dekabrya 2013 g. [Assessment of toxicity and hazards of chemicals and their mixtures for human health: guide, approved by the (acting as) RF Chief Sanitary Inspector on December 27, 2013]. Moscow, The Federal Center for Hygiene and Epidemiology, 2014, 639 p. (in Russian).

Table 2

Organic compounds identified in water extracts from the analyzed polyurethane-based material (under 20 °C) when a subsequent migration process was examined

№	Compound	Gross formula	CAS	Concentration, mg/dm^3		
				ST2	ST3	ST6
1	Tetradecane	$C_{14}H_{30}$		0.003	-	0.002
2	Hexadecane	$C_{16}H_{34}$		0.002	0.004	0.002
	spirits					
3	Hexadecanol	$C_{16}H_{34}O$	36653-82-4	0.003	-	0.003
4	Heptadecanol	$C_{17}H_{36}O$		0.006	-	-
	phenols					
5	4,4-isopropylidene diphenol	$C_{15}H_{16}O_2$	80-05-7	0.003	0.006	-
6	2,6-Di-tert-butyl-4-ethylphenol	$C_{16}H_{26}O$	4130-42-1	0.004	-	-
	simple ethers					
7	2,5,8,11- Tetraoxadodecane *	$C_8H_{18}O_4$	112-49-2	0.056	0.016	-
	ketones					
8	Benzophenone	$C_{13}H_{10}O$	119-61-9	0.004	0.004	-
	carboxylic acids					
9	Tetradecane	$C_{14}H_{28}O$	544-63-8	-	0.005	0.004
10	Hexadecane	$C_{16}H_{32}O$	57-10-3	0.011	0.019	0.016
	complex ethers					
11	Dimethylethyl methyl propanol ether of 2-methylpropanoic acid	$C_{16}H_{30}O_4$	74381-40-1	0.005	0.014	0.011
12	Diisobutyl ether of adipic acid	$C_{14}H_{26}O_4$	141-04-8	0.030	0.025	0.014
13	Dimetoxy tetraethylene glycol *	$C_{10}H_{22}O_5$	143-24-8	0.018	0.016	0.018
14	Methyl ether of tetrapropylene glycol *	$C_{13}H_{28}O_5$	20324-34-9	0.012	-	-
	phthalates					
15	Dimethyl phthalate	$C_{10}H_{10}O_4$	131-11-3	0.001	0.003	-
16	Diisobutyl phthalate	$C_{14}H_{26}O_4$	84-69-5	0.032	0.026	0.033
17	Dibutyl phthalate	$C_{14}H_{26}O_4$	84-69-2	0.021	0.019	0.016
18	Monobutyl phthalate	$C_{12}H_{14}O_4$	131-70-4	0.063	0.023	-
	nitrogen-containing compounds					
19	1,2,3,4,5,6,7,8-Octahydroacridine	$C_{13}H_{17}N$	1658-088-8	0.003	0.003	0.003
20	4-Piperidinyl-1-methyl-4(2-methyl-2-propenyl)*	$C_{10}H_{19}NO$	-	0.034	-	-
	Nitrogen-, sulfur containing compounds					
21	N-Butylbenzenesulfonamide	$C_{10}H_{15}NO_2S$	3622-84-2	0.023	0.006	0.005

is necessary to pay attention to health effects produced by migrating organic compounds.

We assessed a two-component polyurethane coating that was already allowed for use in drinking water supply in the country of its origin. The assessment results indicate that chemical concentrations in the tested water differ considerably in different experimental designs. When a subsequent migration process was analyzed, contents of organic compounds indicated that intensity of their migration tended to decrease over time whereas the results produced by analyzing a continuous migration process according to the methodical guidelines MU 2.1.4.2898-11[2] indicated that quality of drinking water could deteriorate rapidly due to ongoing migration during long-term stagnation.

More than 20 organic compounds were identified in the analyzed water samples. Most of them occurred in insignificant concentra-

tions (excluding N-Butylbenzenesulfonamide). We should note that many of them do not have hygienic standards or reference concentrations in drinking water established for them in regulatory documents and a relevant toxicological assessment is also absent [28]. Given that, it is practically impossible to perform conventional assessment of human health risks caused by exposure to these compounds. Further research is necessary to specify risk parameters and toxicity for specific pollutants as well as to examine how these detected chemicals interact with each other.

Our research confirms the necessity to perform hygienic assessment of materials in conditions close to their actual use in drinking water supply. It concerns estimating an area of contact between a material and test water, analyzing a continuous or subsequent migration process, and applying a set of indicators that are subject to mandatory control. All this again highlights the necessity to actualize and test all the assessment procedures applied to materials that are planned for use in drinking water supply, depending, among other things, on a type of a material and its functional purpose.

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Competing interests. The authors declare no competing interests.

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