Refractive index variations of glass microfragments by annealing – forensic applications

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Abstract
Annealing, as a possible glass investigating method was used for the first time in the Hungarian Institute for Forensic Sciences (HIFS). Glass is a frequently examined evidence type in forensic investigations. In the case of glass microfragments, the most common method for characterization is the measurement of the refractive index (RI). This characteristic value changes after heating up and cooling down the fragments according to the degree of the internal structural stress. The extent of this change (ΔRI) can be used in glass characterization and investigation. In the present study, first 25–25–25 toughened, nontoughened plate and container glasses were investigated. It has been found that, based on the ΔRI values, these glass types are distinguishable. Furthermore, the type of an unknown glass sample most likely can be determined. In real forensic cases it has been shown that a) more reliable information on the possible origin can be obtained if the refractive index measurements are supplemented with the examination of the RI values after annealing; b) if fragments have RI values very close to each other, the origin may be clarified using ΔRI, especially in those cases when the control samples with the same refractive indices are different types of glasses. The experiments were carried out on fragments in the range of ~100 μm.
Keywords: glass, crime evidence, delta RI, annealing, forensic discrimination
Kulcsszavak: üveg, bűncselekmény bizonyítéka, RI változás, felfűtés, forenzikus megkülönböztetés

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1. Introduction

Glass is a frequently examined material [1, 2], and it is also important – as crime evidence – in forensic laboratories [3]. In the case of criminal activities including glass breaking, small fragments may transfer to the clothing of the persons standing nearby, and glass fragments can also be found on the objects used for the breaking. To associate a person or an object with the crime scene, a comparison of the control sample(s) and the recovered fragments from the clothes or objects is required. A useful comparison method is the measurement of the refractive index, which has the advantage over other techniques, e.g. X-Ray Fluorescence Spectrometry (XRF), Inductively Coupled Plasma – Mass Spectrometry (ICP-MS) or Scanning Electron Microscopy with Energy Dispersive Spectroscopy (SEM/EDS), that it can be applied for glass fragments smaller than 100 μm in one dimension. However, the discrimination force of the refractive index (RI) is usually not as good as of the above-mentioned techniques. A possible way of improving the discrimination is the process of annealing. Glass has internal structural stress due to its thermal disequilibrium, which depends on the type of the glass. Because of the manufacturing process, toughened glass has greater internal structural stress compared to the non-toughened glasses. Annealing (heating up followed by a very slow cooling) minimizes this stress, which results in an increase in the RI values, so the difference between the post- and the preannealed RI values (ΔRI) is a characteristic feature of the stress level in the investigated glass [4]. Locke and Hayes have shown that the ΔRI value is in the range of 0.00173–0.00206 for toughened, 0.00086–0.00144 for non-toughened float, patterned or plate glass, while 0.00073 for container glass [5]. Work performed by Cassista and Sandercock [6], Locke and Rockett [7], Winstanley and Rydeard [8], and Marcouiller [9] has been also shown and confirmed that toughened glass can be classified by annealing. It was also shown that the increase of the heating time (6–12–24 hours) increases the measured ΔRI value [10].

The aim of our work was to carry out annealing experiments for the first time in the Hungarian Institute for Forensic Sciences, including the investigation of 25–25–25 toughened, nontoughened plate and container glasses, and testing the discrimination power of the annealing method in real cases.

2. Methods

Refractive indices were measured by the oil immersion method using the GRIM®3 system made by Foster&Freeman. A narrow-band pass filter (589 nm) was used to choose the appropriate wavelength and to block the other lines of the light source. All investigated glass microfragments were mounted onto separate microscope slides, covered with a few drops of silicone oil (Locke Scientific Oil B), crushed with a dissecting needle and covered with a thin glass cover plate. The fragments

Container glasses			Plate glasses			Toughened glasses		
RI_{before}	RI_{after}	ΔRI	RI_{before}	RI_{after}	ΔRI	RI_{before}	RI_{after}	ΔRI
1.51959	**1.51993**	**34·10⁻⁵**	1.51729	1.51842	113·10⁻⁵	1.51995	1.52186	191·10⁻⁵
1.52339	1.52375	36·10⁻⁵	1.52066	1.52143	77·10⁻⁵	**1.51961**	**1.52160**	**199·10⁻⁵**
1.52477	1.52509	32·10⁻⁵	1.51683	1.51751	68·10⁻⁵	1.52053	1.52255	202·10⁻⁵
1.52346	1.52377	31·10⁻⁵	1.51417	1.51515	98·10⁻⁵	1.52109	1.52311	202·10⁻⁵
1.52527	1.52547	20·10⁻⁵	1.51773	1.51881	108·10⁻⁵	1.52291	1.52471	180·10⁻⁵
1.52446	1.52470	24·10⁻⁵	1.51383	1.51485	102·10⁻⁵	1.52102	1.52309	207·10⁻⁵
1.52546	1.52576	30·10⁻⁵	1.51571	1.51681	110·10⁻⁵	1.52436	1.52619	183·10⁻⁵
1.52047	1.52081	34·10⁻⁵	1.51853	1.51944	91·10⁻⁵	1.52149	1.52355	206·10⁻⁵
1.52200	1.52235	35·10⁻⁵	1.51894	1.51993	99·10⁻⁵	1.51762	1.51951	189·10⁻⁵
1.52385	1.52423	38·10⁻⁵	1.51761	1.51855	94·10⁻⁵	1.52426	1.52621	195·10⁻⁵
1.52426	1.52448	22·10⁻⁵	1.51530	1.51633	103·10⁻⁵	1.52447	1.52632	185·10⁻⁵
1.52300	1.52331	31·10⁻⁵	1.52097	1.52182	85·10⁻⁵	1.52237	1.52435	198·10⁻⁵
1.52412	1.52440	28·10⁻⁵	1.52077	1.52140	63·10⁻⁵	1.52437	1.52632	195·10⁻⁵
1.52400	1.52435	35·10⁻⁵	1.51433	1.51524	91·10⁻⁵	1.52006	1.52201	195·10⁻⁵
1.52140	1.52180	40·10⁻⁵	1.52101	1.52172	71·10⁻⁵	1.51911	1.52110	199·10⁻⁵
1.52379	1.52407	28·10⁻⁵	1.52207	1.52278	71·10⁻⁵	1.51677	1.51843	166·10⁻⁵
1.52409	1.52435	26·10⁻⁵	1.51907	1.51970	63·10⁻⁵	1.52119	1.52288	169·10⁻⁵
1.52395	1.52430	35·10⁻⁵	1.52076	1.52152	76·10⁻⁵	1.52181	1.52360	179·10⁻⁵
1.52369	1.52394	25·10⁻⁵	1.52409	1.52469	60·10⁻⁵	1.51508	1.51686	178·10⁻⁵
1.52386	1.52424	38·10⁻⁵	1.51715	1.51798	83·10⁻⁵	1.51917	1.52098	181·10⁻⁵
1.52387	1.52417	30·10⁻⁵	1.51688	1.51765	77·10⁻⁵	1.51920	1.52109	189·10⁻⁵
1.52603	1.52625	22·10⁻⁵	1.52115	1.52184	69·10⁻⁵	1.52010	1.52206	196·10⁻⁵
1.52554	1.52583	29·10⁻⁵	1.51983	1.52048	65·10⁻⁵	1.52096	1.52299	203·10⁻⁵
1.52340	1.52374	34·10⁻⁵	**1.51946**	**1.52016**	**70·10⁻⁵**	1.51803	1.51984	181·10⁻⁵
1.52335	1.52358	23·10⁻⁵	1.51912	1.51979	67·10⁻⁵	1.52023	1.52206	183·10⁻⁵

Table 1 Average refractive indices of 25–25–25 container, plate and toughened glasses before (RI_{before}) and after (RI_{after}) annealing, and the difference of these values (ΔRI)
1. táblázat 25-25 db öblös-, sík- és biztonsági üveg átlagos optikai törésmutatója hőkezelés előtt (RI_{before}) és azt követően (RI_{after}), valamint ezen értékek különbségei (ΔRI)

were observed with a phase-contrast microscope as the temperature of the slides was varied at ramp-rate of 4 °C min⁻¹. The average RI value was determined from the measured matching temperature using a calibration curve determined by the measurements of 10 glass standards (Locke Scientific).

The annealing was carried out by using an OMSZÖV OH63 type furnace, which was heated up to the required temperature (see the exact values at the appropriate chapters in the Results section) during ~1 hour. The temperature was then held at the chosen degree for 4.0 hours in each experiment, and it was controlled by a GANZ DKT NiCr-Ni thermometer. As no installation existed for temperature programming, the rate of cooling was limited to that achieved by turning off the supply of the furnace. After an overnight cooling, the temperature was under 200 °C, and at this stage the furnace was opened. In the annealing experiments, a porcelain combustion boat or a homemade stainless steel sample holder with holes of 6 mm in diameter and 3 mm deep were used.

The investigated glasses, which were cleaned before the investigation using 96 V/V% ethanol, are mentioned in the appropriate chapters of the Results section.

3. Results

3.1 ΔRI of different glass types

Altogether 25 container glasses (5 colorless, 10 green, and 10 brown beer or wine bottles purchased in Hungary), 25 colorless plate glasses, and 25 toughened glasses (5 colorless, 10 pale green, and 10 green) were investigated. Each sample was crushed, and one bulk fragment was chosen from each glass. This fragment was broken into two equal parts, and the average RI value of one was measured (RI_{before}). The other part was annealed (up to 700 °C) followed by the RI determination (RI_{after}). These values are shown in *Table 1*.

According to the data shown in *Table 1*, the ΔRI values are in the range of 0.00020–0.00040 for container glasses, 0.00060–0.00113 for plate glasses, and 0.00166–0.00207 for toughened glasses. In the last case, the measured values are almost the same as observed by Locke and Hayes [5]. In contrast, the values for the plate and the container glasses are ~30·10⁻⁵ smaller compared to the results by Locke and Hayes. The observed ΔRI values form three distinct groups as it is shown in *Fig. 1*. Although different types of glasses could be almost the same RI_{before} values, according to the ΔRI values these glasses are distinguishable (see the bold values in *Table 1*). Moreover, the type of an unknown glass sample most likely can be determined.

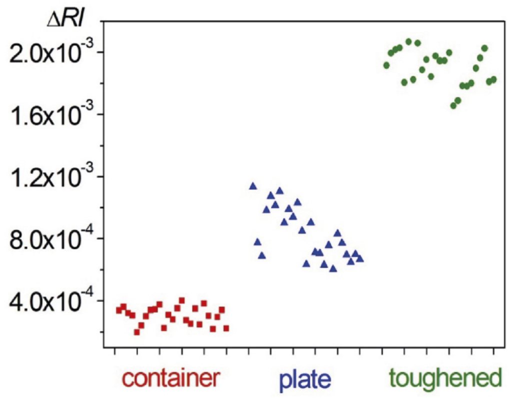

Fig. 1 The difference of the average refractive indices (ΔRI) of 25–25–25 container, plate, and toughened glasses after (RI_{after}) and before (RI_{before}) annealing
1. ábra 25-25 db öblös-, sík- és biztonsági üveg felfűtést követően (RI_{after}) és azt megelőzően (RI_{before}) mért átlagos optikai törésmutatójának különbségei (ΔRI)

3.2 ΔRI – an additional data for recovered microfragments

In the case of glass microfragments, the comparison of the recovered and the control samples in forensic laboratories usually based on only one information – the refractive index. In order to support the comparison from another side, the measurement of the ΔRI values could be important additional information. It is important to note that ΔRI can be measured also in the case of fragments smaller than 100 µm, which samples are not large enough for Micro X-Ray Fluorescence Spectrometry (µ-XRF) or Laser Ablation Inductively Coupled Plasma - Mass Spectrometry (LA-ICP-MS) investigations.

In one of our cases in 2019, first the refractive index of the control toughened glass sample was measured. It has been shown in many studies that the average RI value of the fragments from the surface can be a little bit different from the values of the bulk fragments [11, 12]. According to this, the average RI value of a bulk and a surface fragment was measured. As it is shown in *Table 2*, a ~$20 \cdot 10^{-5}$ difference was observed, which is consistent with the previous literature data. Based on the average refractive indices and on the range of RI values, our initial statement was that two of the recovered fragments could originate from the bulk, and two from the surface of the control sample. In the next step, both the bulk and the surface fragment of the control sample together with the four recovered fragments were annealed up to 450 °C. The lower temperature was chosen for two reasons: a) in the case of one glass fragment annealed up to 700 °C in the previous experiment a significant deformation was observed, which we would like to avoid in the experiments with recovered fragments; b) our aim was to examine how much the refractive index changes when lower temperature is applied. The measured average RI value after annealing together with the minimum and maximum values are also shown in *Table 2* and *Fig. 2*. It is clearly visible, that the data of the recovered fragments and the control sample is the same not just before, but after annealing. This makes it even more certain that each of the recovered four microscale glass fragments can originate from the control sample. Furthermore, it is interesting to note that the average ΔRI values are the same for the bulk and the surface fragments of the control sample ($85 \cdot 10^{-5}$), and – as it was expected – smaller than the observed RI changes for toughened glasses annealed up to 700 °C.

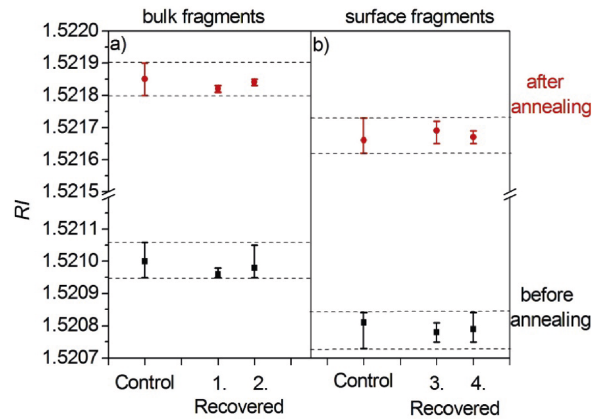

Fig. 2 The average RI values together with the range observed for a) bulk and b) surface fragments of the control sample and for four recovered microscale glass fragments before (black) and after (red) annealing in one of the examined cases in the HIFS
2. ábra Egy, az NSZKK-ban vizsgált ügyben érkezett összehasonlító minta a) tömbi és b) felszíni szemcséjének, valamint a rázalékban talált négy darab bűnjelszemcsének az átlagos optikai törésmutató értéke a mérési tartománnyal együtt felfűtés előtt (fekete) és azt követően (piros)

	Before annealing			After annealing			$\Delta RI_{average}$
	$RI_{average}$	$RI_{min.}$	$RI_{max.}$	$RI_{average}$	$RI_{min.}$	$RI_{max.}$	
Control (bulk)	1.52100	1.52095	1.52106	1.52185	1.52180	1.52190	$85 \cdot 10^{-5}$
Recovered (1.)	1.52096	1.52095	1.52098	1.52182	1.52181	1.52183	$86 \cdot 10^{-5}$
Recovered (2.)	1.52098	1.52095	1.52105	1.52184	1.52183	1.52185	$86 \cdot 10^{-5}$
Control (surface)	1.52081	1.52073	1.52084	1.52166	1.52162	1.52173	$85 \cdot 10^{-5}$
Recovered (3.)	1.52078	1.52075	1.52081	1.52169	1.52165	1.52172	$91 \cdot 10^{-5}$
Recovered (4.)	1.52079	1.52075	1.52084	1.52167	1.52165	1.52169	$88 \cdot 10^{-5}$

Table 2 The average, minimum, and maximum RI values observed for the bulk and surface fragments of the control sample and for four recovered microscale glass fragments before and after annealing together with the differences of the $RI_{average}$ values in one of the examined cases in the HIFS
2. táblázat Egy, a Nemzeti Szakértői és Kutató Központban (NSZKK) vizsgált ügyben érkezett összehasonlító minta tömbi és felszíni szemcséjének, valamint a rázalékban talált négy darab bűnjelszemcsének az átlagos optikai törésmutató értéke, az egyes szemcséknél mért legkisebb és legnagyobb optikai törésmutató értékek felfűtés előtt és azt követően, valamint a felfűtést követően és azt megelőzően mért átlagos RI értékek különbségei

3.3 Discrimination by ΔRI

It has been shown in earlier investigations [13], and also in our experiments (see *Table 1*) that different types of glasses from different sources can have very similar RI values. As it is shown in *Table 1* (see the values in bold) the average RI value measured before annealing of one of the container, plate, and toughened glasses (1.51959, 1.51946, 1.51961) may be so close to each other that the clear distinction based on the refractive indices is not possible. A similar situation was observed in one of our other cases, in which eight windows of a car were broken by a suspect. Four out of the eight control samples (indicated as 1.2., 1.5., 1.7., 1.8.) have similar average RI values, while the other four samples have different refractive indices as it is shown in *Table 3*. According to our measurements, two recovered fragments can originate from the control samples 1.2. and 1.5., and their average RI values are also close to the appropriate values of control samples 1.7. and 1.8. In order to clarify their origin, both the recovered fragments and the four control samples were annealed up to 650 °C. As it is shown in *Table 3*, the average RI changes in the case of 1.2., 1.7., and 1.8. samples are $187 \cdot 10^{-5}$, $179 \cdot 10^{-5}$, and $179 \cdot 10^{-5}$, respectively. These values are in accordance with our previous measurements for toughened glasses. However, in the case of 1.5., the ΔRI value is much smaller, $82 \cdot 10^{-5}$, which corresponds to the value measured for plate glasses. (As it is turned out later, the car was out of use, and one of its windows was replaced by a plate glass.) Additional result of this experiment was that annealing up to 650 °C or 700 °C causes similar changes in the refractive indices.

Examining the two recovered fragments, the first fragment has 1.52159 average RI value after annealing, which means $87 \cdot 10^{-5}$ change in the refractive index. Thus, it can be clearly seen that this fragment very likely originates from control sample 1.5., and cannot originate from any of the other control samples. The second examined recovered fragment was very small, so only one measurement could be carried out after annealing, which resulted in an RI value of 1.52169. This value is in the RI range (1.52153–1.52170) of control sample 1.5. after annealing, and significantly different from the other control samples examined by annealing. It means that the second investigated recovered fragment can also likely originate from sample 1.5. The results of this experiment are shown both in *Table 3* and *Fig. 3*.

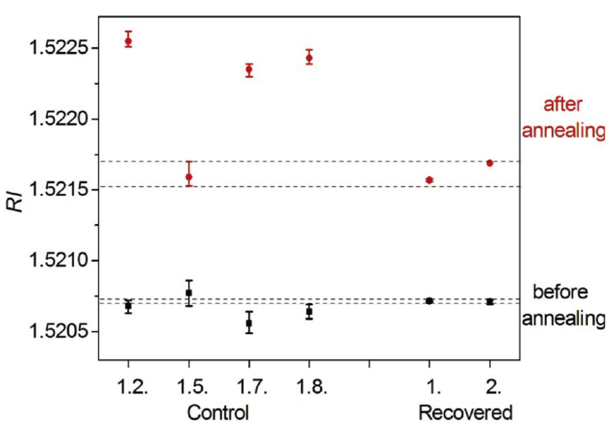

Fig. 3 *The average RI values together with the range observed for four control samples and for two recovered microscale glass fragments before (black) and after (red) annealing in one of the examined cases in the HIFS*

3. ábra *Egy, az NSZKK-ban vizsgált ügyben érkezett négy darab összehasonlító minta és két darab bűnjelszemcse átlagos optikai törésmutató értéke a mérési tartománnyal együtt felfűtés előtt (fekete) és azt követően (piros)*

		Before annealing			After annealing			
		$RI_{average}$	$RI_{min.}$	$RI_{max.}$	$RI_{average}$	$RI_{min.}$	$RI_{max.}$	$\Delta RI_{average}$
Control samples	**1.1.**	1.52301	1.52295	1.52304	not investigated	-	-	-
	1.2.	1.52068	1.52063	1.52072	1.52255	1.52251	1.52262	$187 \cdot 10^{-5}$
	1.3.	1.51795	1.51789	1.51799	not investigated	-	-	-
	1.4.	1.52357	1.52352	1.52364	not investigated	-	-	-
	1.5.	1.52077	1.52068	1.52086	1.52159	1.52153	1.52170	$82 \cdot 10^{-5}$
	1.6.	1.52146	1.52141	1.52153	not investigated	-	-	-
	1.7.	1.52056	1.52049	1.52064	1.52235	1.52230	1.52239	$179 \cdot 10^{-5}$
	1.8.	1.52064	1.52059	1.52069	1.52243	1.52239	1.52249	$179 \cdot 10^{-5}$
Recovered (1.)		1.52072	1.52071	1.52072	1.52159	1.52158	1.52160	$87 \cdot 10^{-5}$
Recovered (2.)		1.52071	1.52069	1.52072	1.52169	1.52169	1.52169	$98 \cdot 10^{-5}$

Table 3 *The average, minimum, and maximum RI values observed for eight control samples and two recovered fragments before annealing and in six cases after annealing together with the differences of the $RI_{average}$ values*

3. táblázat *Nyolc darab összehasonlító minta és két darab bűnjelszemcse átlagos optikai törésmutató értéke, valamint az egyes szemcséknél mért legkisebb és legnagyobb optikai törésmutató értékek felfűtés előtt és összesen hat esetben azt követően, valamint a felfűtést követően és azt megelőzően mért átlagos RI értékek különbségei*

4. Conclusions

The change in the refractive indices by annealing caused by the various structural stresses present in different types of glasses represents a possibility for glass origin examination which was not previously used in the Hungarian Institute for Forensic Sciences (HIFS). In accordance with previous investigations, our experiments have also shown that the type of an unknown glass fragment can be determined by annealing. So far, comparative analysis of microscale glass fragments in the HIFS has been performed only by refractive index measurements. By supplementing these investigations with the examination of the RI values after annealing, even more reliable information on the possible origin can be obtained. For those fragments which have more or less the same RI values the origin may be clarified, especially in those cases when the control samples with the same refractive indices are different types of glasses. A further advantage of the annealing experiments is that the ΔRI value can be measured on fragments in the range of ~100 μm, which are not large enough for comparison based on elemental analysis. In conclusion, annealing is a good additional opportunity for the examination of glass microfragments, which is – based on our experiments – applicable in the Hungarian Institute for Forensic Sciences.

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