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High-cycle fatigue monitoring in $AgCl_xBr_{1-x}$ fibres using absorption spectroscopy

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The spectral transmittance of mixed silver-halide polycrystalline fibres was measured while they were undergoing repeated bending leading to mechanical fatigue. Microscopic mechanical defects were detected through their influence on the optical losses, without interfering with the deformation. Optical and mechanical lifetimes of the fibres were found to depend on the composition and to be larger for highly-mixed compositions. Scanning electron microscopy observations of fatigued fibres revealed a strong influence of the fibre composition on the fatigue damage. This dependence on the composition was explained using a theoretical model of solid-solution strengthening.

1. Introduction

The characterization of the mechanical and optical properties of materials are normally performed using two independent measurements. In this work the measurement of both properties has been combined into a single experiment in which the infrared transmission spectrum of silver-halide fibres was measured while they were undergoing mechanical stress. It was found that mechanical defects can be detected by such optical measurements even when the defects are microscopic, and the optical monitoring does not interfere with the mechanical processes.

Silver-halide single crystals have been grown with various $AgCl_xBr_{1-x}$ compositions ($0 < x < 1$) in our laboratory [1], and extruded to form polycrystalline fibres in lengths of several metres and diameters of around 1 mm. During extrusion the crystals are plastically deformed, and the resulting fibres consist of polycrystalline solid-solution materials. The optical and mechanical properties of these crystals and fibres have been previously reported [2, 3]. These flexible optical fibres, which transmit in the mid-infrared (IR) spectral region of 2–20 μm, are useful [1] for IR spectroscopy, radiometry, and high-power laser transmission. The basic mechanical behaviour of the silver halides resembles soft materials such as polycrystalline metals, but because of their high transparency they are also suitable for optical characterization during the mechanical experiment.

High-cycle fatigue of mixed silver halide fibres using a CO_2 laser (at the 10.6 μm wavelength) have been previously reported [4, 5], and the spectral measurements reported here are a useful extension of such studies. Spectral measurements under fatigue bending of silver-halide fibres in the plastic strain-range were reported for the first time by Barkay *et al.* [6, 7]. The spectral measurements reported here are an extension of these measurements to the elastic strain-range, where the number of bending cycles may reach 10^7 (fatigue limit). The spectral characterization, as opposed to investigation at a single wavelength, provides insight into the growth and dynamics of the defects in the material, as the defect growth is expected to be related to the ratio between the defect size and wavelength.

2. Experimental procedures

An experiment has been designed to measure the spectral transmission of infrared fibres during forced bending vibrations. The experimental setup, shown schematically in Fig. 1, is based on a combination of a Fourier transform infrared (FTIR) spectrometer (Nicolet Model 5PC) and a specially built mechanical device whose details have been described elsewhere [5].

The input end of the fibre is held in a holder which moves up and down with an amplitude $h = 2.8$ mm and a frequency of 45 Hz. The fibre is stopped in the "down" position, coinciding with the lens focus, so that the spectral transmission can be taken while the fibre is maximally bent. This assures that the position of the fibre input end relative to the FTIR beam is maintained during the actual measurement. We used a ZnSe lens of 2.5 cm diameter and 2.0 cm focal length. This optical coupling has a large numerical aperture and rays propagate in the fibre at large angles (high modes). Such coupling is advantageous for sensing defects in the fibre, especially for surface defects [8].

The fixed output end of the fibre is placed very close to an external pyroelectric detector (DTGS type). This detector is identical to the one that is normally used in the FTIR system. The output of the detector is fed into the electronics of the FTIR and the signal recorded

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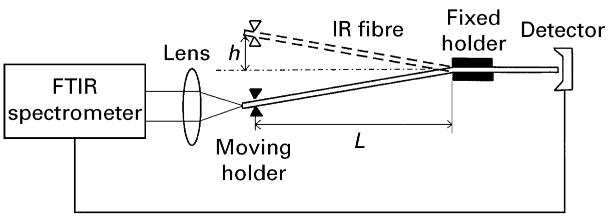

Figure 1 The experimental setup.

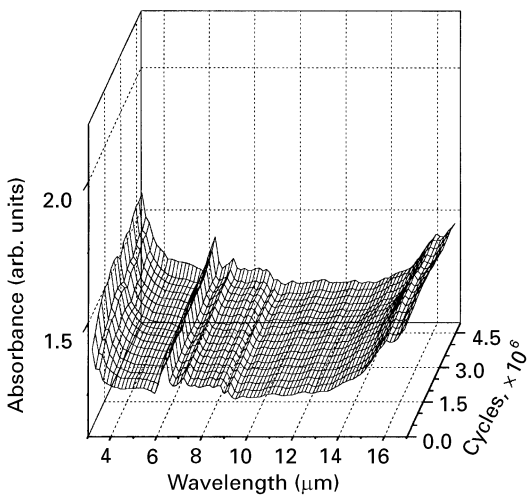

Figure 2 Total spectral attenuation of a fatigued $AgCl_{0.3}Br_{0.7}$ fibre in the bent position as a function of the number of bending cycles ($R = 24$ cm, $\varepsilon_m = 0.19\%$).

and processed using the standard procedures of the FTIR system.

The bending radius, R, of the fibre is not uniform along its length. It is minimal at the fixed holder and can be calculated according to the expression $R = L^2/3h$ [4], where L is the distance between the holders. Multiple bending of a fibre section in the elastic regime of deformation is continued until fracture of the fibre, while the spectrum is measured periodically.

3. Results and discussion

Fibres of three representative compositions ($AgCl_{0.3}Br_{0.7}$, $AgCl_{0.77}Br_{0.23}$ and $AgCl_{0.89}Br_{0.11}$) were flexed in a strain range (ε) close to their fatigue limit ($R = 24$ cm, $\varepsilon = 0.19\%$), where the influence of the composition on mechanical lifetime is more pronounced [5].

The results of the experiment for a $AgCl_{0.3}Br_{0.7}$ fibre are shown in Fig. 2 in which the infrared spectral attenuation of the fibre in the bent position is shown as a function of the number of bending cycles. Each spectrum indicates the sum of all the optical losses, including absorption, scattering and coupling losses into the fibre. Transmittance decreases in the shorter wavelength region ($\lambda < 5$ μm) due to scattering [9] and at the longer wavelength edge ($\lambda > 13$ μm) due to multiphonon absorption [3]. The small peaks in the "transparent" region correspond to the absorption of material impurities, including ambient water and CO_2. The attenuation grows slightly at short wavelengths as bending is repeated, until finally the fibre breaks.

In order to distinguish the mechanical fatigue induced losses from other optical losses the transmittance results were normalized to the initial values for each wavelength. The net degradation of transmittance as a function of number of cycles is plotted in Fig. 3(a–c) for "soft" $AgCl_{0.89}Br_{0.11}$, "medium" $AgCl_{0.77}Br_{0.23}$ and "hard" $AgCl_{0.3}Br_{0.7}$ fibres. In the "soft" fibre optical deterioration starts at the early stage of bending and propagates rapidly at all wavelengths. In the "medium" fibre there is a decrease of transmittance at all wavelengths, as the experiment proceeds, while the effect at long wavelengths is smaller. In the "hard" fibre only the transmittance at 4–5 μm decreases, while at $\lambda > 5$ μm it is constant, until the final break. The fibres were bent at the same strain conditions ($R = 24$ cm, $\varepsilon = 0.19\%$), and the

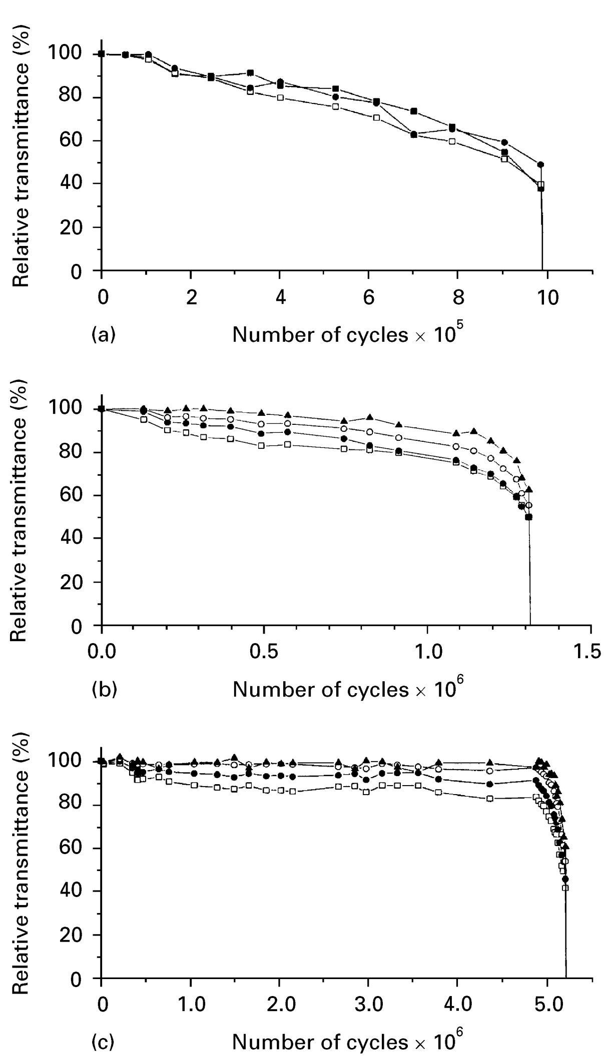

Figure 3 Fatigue induced degradation ($\varepsilon_m = 0.19\%$) of the optical transmittance in the following fibres: (a) $AgCl_{0.89}Br_{0.11}$ – "soft" fibre at (■) 4 μm, (□) 10 μm and (●) 16 μm, (b) $AgCl_{0.77}Br_{0.23}$ – "medium" fibre at (□) 4 μm, (●) 10 μm, (○) 14 μm and (▲) 18 μm and (c) $AgCl_{0.3}Br_{0.7}$ – "hard" fibre at (□) 4 μm, (●) 5 μm, (○) 10 μm and (▲) 14 μm. Measurements were performed for fibres in the bent position.

number of cycles to failure was found to be higher for the harder fibre. This is in agreement with our previous measurements of the dependence of fatigue limit on the $AgCl_xBr_{1-x}$ composition [5].

In order to emphasize the contrast in degradation of the fibres of different compositions during repeated bending we plotted in Fig. 4 their losses in the bent position at $\lambda = 10$ μm as a function of the number of bending cycles. It is useful to introduce qualitative periods in the "life" of repeatedly bent fibres. We define the "mechanical lifetime" of the fibre as being the total number of cycles until mechanical fracture, at which transmission drops to zero. We also define the "optical lifetime" as being the period (number of bending cycles) before there is significant change in the optical transmission. Using these definitions we can compare the behaviour of fibres of different compositions during repeated bending. One can see from Fig. 4 that there is a remarkable difference in the "mechanical lifetime" of the fibres: the "hard" fibre transmits to five times more bending cycles. The "optical lifetime" of this fibre is also significantly longer than that observed in fibres of softer compositions. Optical transmittance of the "hard" fibre is kept at its initial value (at 10 μm) until close to the fibre break. The "optical lifetime" in softer fibres is significantly shorter: optical degradation starts at the early stages of bending and decreases with a constant slope. The rate of degradation is higher for the "soft" fibre than that for the "medium" fibre.

The optical behaviour of the fibres is closely related to the known mechanical phases in a fatigue phenomenon [10]. At the beginning of bending vibration there is the nucleation phase of the defects. In this phase there is no observable mechanical damage and it corresponds to the "optical lifetime" period. The length of this period depends on the fibre composition: it is longer for the "hard" fibre (about 90% of total lifetime). In the "soft" and "medium" fibres the nucleation phase of fatigue defects is short ($\sim 10^5$ cycles), relative to the total lifetime ($\sim 10^6$ cycles).

The next period in the fibre lifetime corresponds to the defects growth phase. This growth progresses at an accelerated rate until fracture, followed by the decrease of the fibre transmission. This period is longer in soft fibres and is very short in hard and brittle fibres.

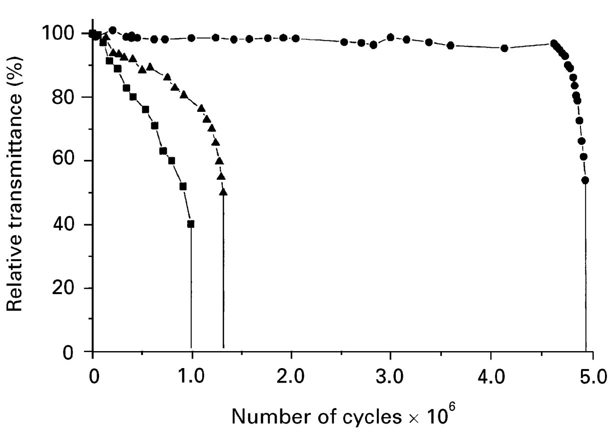

Figure 4 Fatigue induced degradation of the optical transmittance in the bent position at 10 μm for (■) $AgCl_{0.89}Br_{0.11}$, (▲) $AgCl_{0.77}Br_{0.23}$ and (●) $AgCl_{0.3}Br_{0.7}$.

Transmittance losses in the fibres are probably the result of scattering from defects (mainly surface defects), which are formed in the fatigued sections. Infrared spectral monitoring allows an estimate of the fatigue defects size, because the wavelength dependence of the mechanical fatigue induced optical deterioration stems from the ratio of the wavelength to the defects size. Small defects are "sensed" by the short wavelength radiation, while they only slightly affect the long wavelength transmittance. The number and size of defects increase as the experiment proceeds, and the dynamics of this process depends on the fibre composition. It should be mentioned that the defect size should be compared to the wavelength inside the material, which is about half of the free-space value, due to the refractive index of the material.

Comparing the FTIR spectral measurements with our CO_2 laser transmission results at $\lambda = 10.6$ μm [5], it is seen that the spectral ones are more sensitive. We observe an optical degradation in most of the fibres at the early stage of fatigue process even at a wavelength of 10 μm. One reason is that the IR spectral measurements were performed in the bent position. The fatigue defects are expected to be on the surface of the fibre and the bent position particularly enhances those effects specifically related to the fatigue process. A second reason is that the optical setup in the spectral experiments used a high numerical aperture (NA) value and the guided rays propagated at large angles to the axis (high modes). In this setup the optical losses were higher and the sensitivity to any surface defects increased due to the larger number of surface reflections [8]. This is in contrast to the laser measurements, where the NAs is small and many of the guided rays propagate at small angles to the fibre axis.

Fig. 5(a–c) shows photographs of direct observations of typical surface defects using a scanning electron microscope (SEM). Cracks appear on the surface, only in the bent section of the fibres and only on the outer sectors which experience the most strain. Their general direction is perpendicular to the fibre's axis, follow grain boundaries and cause inter-granular separation. It must be mentioned that the spectral measurements are completed when the fibre is maximally bent and the cracks on its surface are "opened". Microscopical observations are carried out after the fibre fracture, so that the applied elastic strain is completely released and the cracks "close".

In the broken "hard" $AgCl_{0.3}Br_{0.7}$ fibre we notice just a few narrow (less than 0.2 μm) cracks of length 20–50 μm, one of which is shown in Fig. 5a. In the "medium" $AgCl_{0.77}Br_{0.23}$ fibre (Fig. 5b) the number of long perpendicular cracks is larger and the fatigued area of the fibre is much longer: up to 5 mm from the broken end. The width of these cracks is about 1–2 μm, they are not straight and have junctions with more narrow cracks, where one can see partial recrystallization and protrusions of the material. Observation of the "soft" $AgCl_{0.89}Br_{0.11}$ fibre using the SEM also revealed several long deep cracks (width about 1–2 μm), one of which is shown in Fig. 5c. One can see

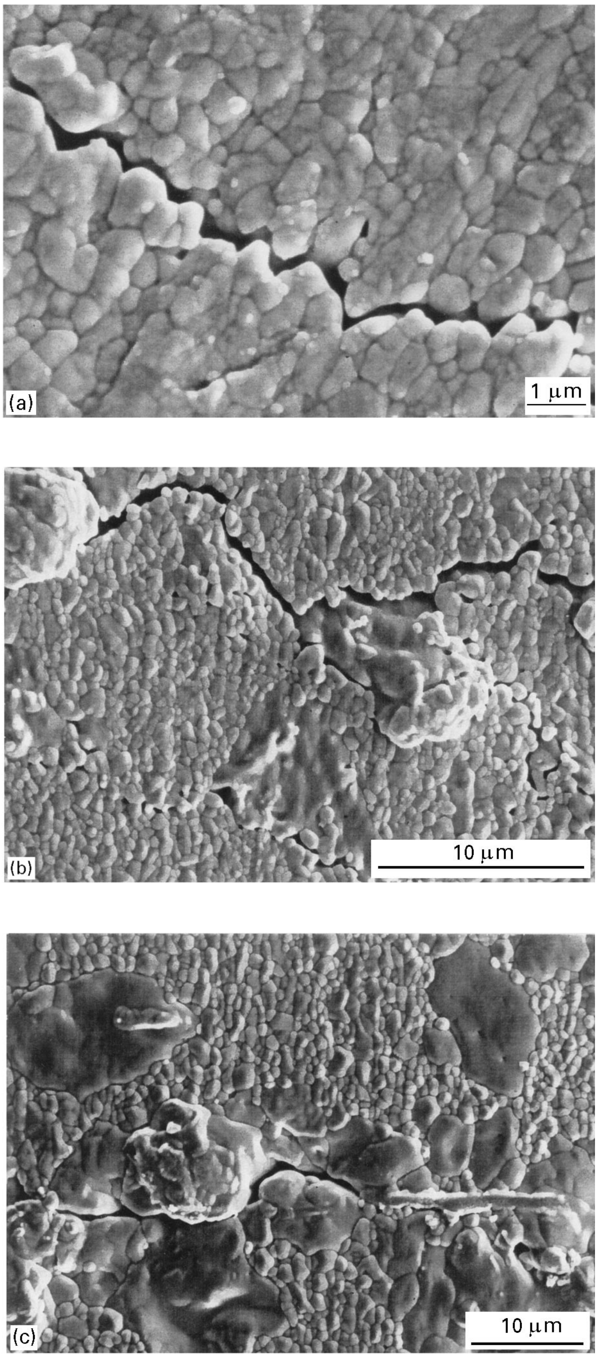

Figure 5 Scanning electron micrographs of the fibres after break: (a) $AgCl_{0.3}Br_{0.7}$ – "hard" fibre, (b) $AgCl_{0.77}Br_{0.23}$ – "medium" fibre, (c) $AgCl_{0.89}Br_{0.11}$ – "soft" fibre.

that in this case recrystallization along the crack, and independently of it, is more pronounced than in the "medium" fibre. The average diameter of these new grains is 10–15 μm. Thus the defects type, size and density strongly depends on the fibre composition. The "hard" fibre is more stable in its deformed-by-extrusion microstructure and defects appears in fewer points. The microstructure of the "soft" fibre is more granular due to the composition, which is close to one of the components (AgCl). It is easily separated along grain boundaries to produce narrow defects.

In ductile material such as silver-halides dislocations and their slip dictate the fatigue behaviour of the material [11]. The dependence of the fatigue defects size on the fibre composition may be explained by a theoretical model, which we have already used to explain the dependence of the fatigue limit of $AgCl_xBr_{1-x}$ fibres on the composition [5]. The model takes into account the additional internal stress field in mixed crystals – an outcome of the random distribution of different sized anions in the solid solution. Dislocations in a mixed crystal require extra work to move against the additional fluctuating stress field, and a higher external stress is thus needed. Thus, when a similar strain is applied to our fibres, dislocations in $AgCl_{0.89}Br_{0.11}$ (close to the pure AgCl composition) slip more easily and produce larger defects which propagate at accelerated rates, leading to lower mechanical and optical lives of this fibre. In our fibres microcracks nucleate along grain boundaries at the fibre surface due to the stress concentration caused by the interaction between persistent slipbands and grain boundaries.

During bending vibration a periodic shear stress acts on the surface of the fibre in the direction of 45° with respect to its axis. The value of shear stress is small relative to the normal stress and leads to formation of fatigue defects only in "medium" and "soft" fibres. In the "soft" fibre we clearly observe (Fig. 5c) shear strain-induced coalescence of grain boundaries. It was shown by Jones *et al.* [12] that under periodic stress the sub-boundary dislocations may link continuously with dislocations in an adjacent high-angle grain boundary. The disappearance of the sub-boundary entails a detectable rearrangement of the dislocations in the grain boundary, and indeed, we found that grain boundaries act more efficiently as sinks than as sources of dislocations. This is the precondition of coalescence – the "evaporation" of sub-boundaries and the formation of new large grains of dimensions $\approx$ 10–12 μm on the fatigued surface. Interaction of dislocations with each other, leading to coalescence of sub-boundaries and formation of the recrystallized structure, decreases the energy of the fibre. The experiment was carried out at room temperature and the macroscopical temperature rise on the surface of the fibre due to internal friction was measured to be less than 0.5 °C, but locally on the edges of microcracks the temperature rise may be significantly higher and activate the recrystallization process. Recrystallization at relatively low temperatures is a very slow process. Our experiment continued for 6 h in the "soft" fibre and recrystallized grains are clearly observed. In the "hard" fibre recrystallization did not start even after 20 h of high-cycle bending vibration.

4. Summary and conclusions

In summary, we have monitored the progress of the formation of mechanical fatigue defects using optical (infrared) spectral measurements. Such optical monitoring does not interfere with the mechanical process itself. Combined mechanical and optical characterization is possible in the same experiment due to the properties of polycrystalline silver-halide fibres. They are transparent in the 2–20 μm regime, while their mechanical behaviour resembles that of other crystalline materials (such as polycrystalline metals).

Detailed experimental results were shown for three representative types of silver-halide fibres: a "hard" $AgCl_{0.3}Br_{0.7}$, a "medium" $AgCl_{0.77}Br_{0.23}$ and a "soft" $AgCl_{0.89}Br_{0.11}$ fibre, that were flexed, in the high-cycle regime close to the fatigue limit. The results showed the degradation of optical transmittance with the number of bending cycles. Thus mechanical defects were observed even when they were microscopic, through their influence on the optical spectrum.

Plotting the attenuation at several wavelength values, it was found that there were differences between the fibre types. Optical and mechanical lifetimes of the "hard" fibre were the largest. This fibre showed optical losses only in short wavelengths ($\lambda \approx 4$–$5\ \mu m$) which grew slightly with the number of bending cycles, but losses at longer λ were not observed until close to final break. The "medium" fibre showed optical losses right from the beginning of the experiment which increased with the same slope at all λ, while the effect at short wavelengths was more pronounced. The "soft" fibre also showed significant losses at the early stage of bending which rapidly increased at similar rates for all λ.

SEM examination was performed after the fibres broke, but only an analysis of the spectral attenuation can indicate which of the observed fatigue defects were really the main cause of optical losses.

The density of fatigue cracks in the "hard" $AgCl_{0.3}Br_{0.7}$ fibre was low and their size was small (width $\sim 0.2\ \mu m$), so they influenced only the short wavelength part of the spectrum. These cracks did not propagate and the spectral transmittance was kept nearly constant until the final break of the fibre. In the "medium" $AgCl_{0.77}Br_{0.23}$ fibre there were several wide (~ 4–$6\ \mu m$ when opened) cracks which grew during the fatigue process, leading to a decrease of transmittance at all wavelengths, while the effect at short wavelengths was more pronounced. In the "soft" $AgCl_{0.89}Br_{0.11}$ fibre fatigue cracks were the largest ($>6\ \mu m$ when opened) and propagated at an accelerated rate during the experiment, leading to a significant decrease of the transmittance at all wavelengths. The dependence of the size and the density of fatigue defects on the $AgCl_xBr_{1-x}$ composition was explained by dislocation slip in the ductile silver-halide material using a theoretical model of solid-solution strengthening and extra strain needed to move dislocations against the additional fluctuating stress field in mixed crystals.

An interesting phenomenon was observed in the "medium" and more pronounced in the "soft" fibre dynamic recrystallization under high-cycle straining: sub-boundary dislocations may link continuously with dislocations in an adjacent high-angle grain boundary, leading to coalescence ("evaporation") of grain boundaries and the formation of new large grains with dimensions of ~ 10–$12\ \mu m$ on the fatigued surface. The recrystallization process dominates in the direction of maximal shear stress on the surface of the fibre and is probably activated by heat formation during friction on the edges of small superficial cracks on the surface of the fibre. We suppose that these new grains only slightly increase scattering in the fibre because of their small phase perturbation that they introduce in comparison with cracks.

From the practical point of view highly-mixed ("hard") compositions, close to $AgCl_{0.5}Br_{0.5}$ composition, have shown the best results. These fibres are the most stable under repeated bending both optically and mechanically. Thus, for applications involving multiple bending of fibres, highly-mixed silver-halide compositions are desirable.

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