ESRF	**Experiment title:** A study of crack propagation in non- and rubber-toughened epoxy adhesives with the use of scanning micro beam synchrotron radiation	**Experiment number**: SC-770
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Epoxy adhesives have found widespread applications in the last years, since they are easy to use and can promote adhesive contact with many materials. However, epoxy adhesives are not totally free of problems. More specifically, epoxies are very brittle and this characteristic creates particular problems in load bearing applications. Thus epoxy adhesives show low fracture toughness, which is one of the main reasons, which limits their range of application. A promising way to increase the fracture toughness of these materials is the blending with softer materials, e.g. rubber particles. In this case the presence of the rubber particles helps to prevent crack propagation through a mechanism where defects can be localised.

The present study aims at the understanding of the local processes in the crack zone and focuses on the investigation of the crack propagation of two grades of engineering epoxy materials:

- Non-toughened epoxy (Araldite),
- Rubber-toughened epoxy (XD4236-2 with rubber particles included).

The samples consist of two aluminium plates bonded together with the different epoxy adhesives. For crack formation an on-line wedge test is carried out *in-situ* with synchrotron radiation small angle scattering with a scanning micro-focused beam (diameter about 6 µm). The samples were mounted on a particularly designed Double Cantilever Beam Test set-up (DCB) [1-4]. A sharp razor blade was inserted at the adhesive layer of the samples for a length of approximately 5 mm. The crack propagated close to the interface between the aluminium plate and the adhesive. Then the cell with the sample was fixed in the beam line and it was tried to identify optically (through the use of a light microscope) the end of the crack. A series of 2D-SAXS measurements were performed in the y-z plane normal to the beam direction to identify the extent of the crack (scanning x-ray microscopy).

Fig. 1 shows a micrograph of the non-toughened epoxy material after crack initiation. A y-z scan was performed around the visual end of the crack. The series of the SAXS patterns reveal (fig. 2) that the crack travels straight through the Al-epoxy interface and that the end of the crack is far away from the visually identified spot. Thus, the crack width is about 40µm and the crack length extends far beyond the scanned area. The SAXS pattern of the measured lines of z = –0.02 mm

and z = –0.04 mm shows a (nearly symmetrical) void scattering around the crack. This behaviour is due to the fact that Araldite epoxy is very brittle and that the crack propagates fast after initiation.
On the other hand**,** the behaviour of rubber-toughened material was very different. The SAXS patterns prior to and after crack initiation are essentially the same (fig. 3a/b). In this case the visual inspection of the specimens is also not consistent with the y-ray results. In this case even at a distance approximately 10mm away from the tip of the razor blade no crack was detected. This essentially indicates that the crack has been stopped or deviated very close to the tip of the razor blade. Therefore a more detailed study at different locations along the interface is necessary.

The SAXS patterns of the rubber toughened epoxies provide also additional interesting information. It is seen in fig. 3 that in the bulk of the adhesive the scattering patterns reveal isotropic scattering characteristic of spherical particles. One can try to estimate the size of these (rubber) particles and ultimately obtain details of the distribution of the particle sizes. The SAXS patterns at the interface are clearly different from those of the bulk. In particular the SAXS patterns are characteristic for fibre structures and their shapes reveal irregular or random orientation of these fibrils. It is known from the literature that inside the rubber particles cavitations may occur even without the application of any load. These cavitations have been shown not to be totally empty, but to contain a number of very thin fibrils. These fibrils along with the existence of cavitations are believed to be responsible for the localisation of the crack by energy dissipation mechanisms. The existence of this structural morphology close to the interface (and not in the bulk of the specimens) is not understood at the moment. Further investigations on the samples will be performed close to the tip of the wedge in an attempt to study the vicinity of the crack.

In addition to those experiments a test was performed with respect to the quite different debonding behaviour of soft polymer materials. If the adhesive polymer is above the glass transition temperature, it is not brittle and tough, but soft and viscous leading to formation of cavities besides interfacial failure and crack propagation. With poly-n-butylacrylate as an adhesive for silicon the first debonding step during mechanical loading was detected by the in-situ scanning of the x-ray micro beam over the sample in the vicinity of the defect region. The defect region is larger than in the case of the epoxy adhesive and voids are identified. More systematic studies are planned in further experiments.

Reference:

[1] C. Creton, E. J. Kramer, C.-Y. Hui, H. R. Brown, *Macromolecules*, 25 (1992) 3075
[2] C. Creton, H. R. Brown, K. R. Shull, *Macromolecules*, 27 (1994) 3174
[3] E. Bocher, J. P. Kolkers, H. Hervet, C. Creton, *Macromolecules*, 29 (1996) 774
[4] R. Schnell, M. Stamm, C. Creton, *Macromolecules*, 32 (1999) 3420

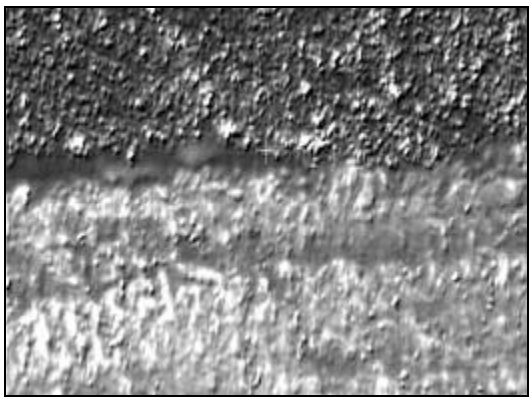

Fig. 1: Microscope picture from measuring point Aral02 of a non-toughened sample after crack initiation

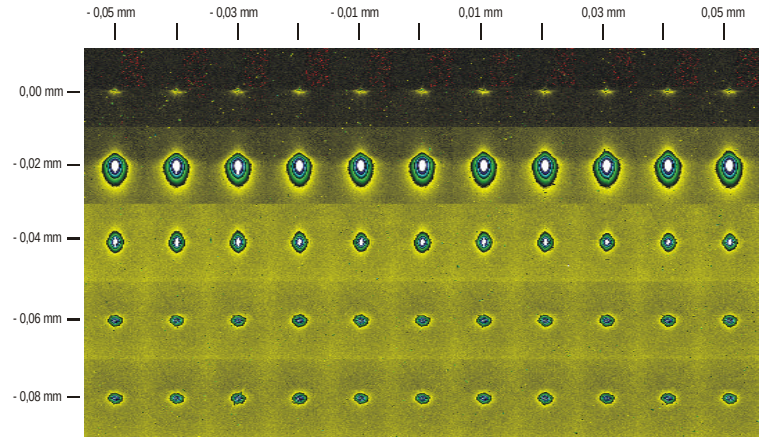

Fig. 2: SAXS pattern scan around measuring point toughened sample after crack initiation

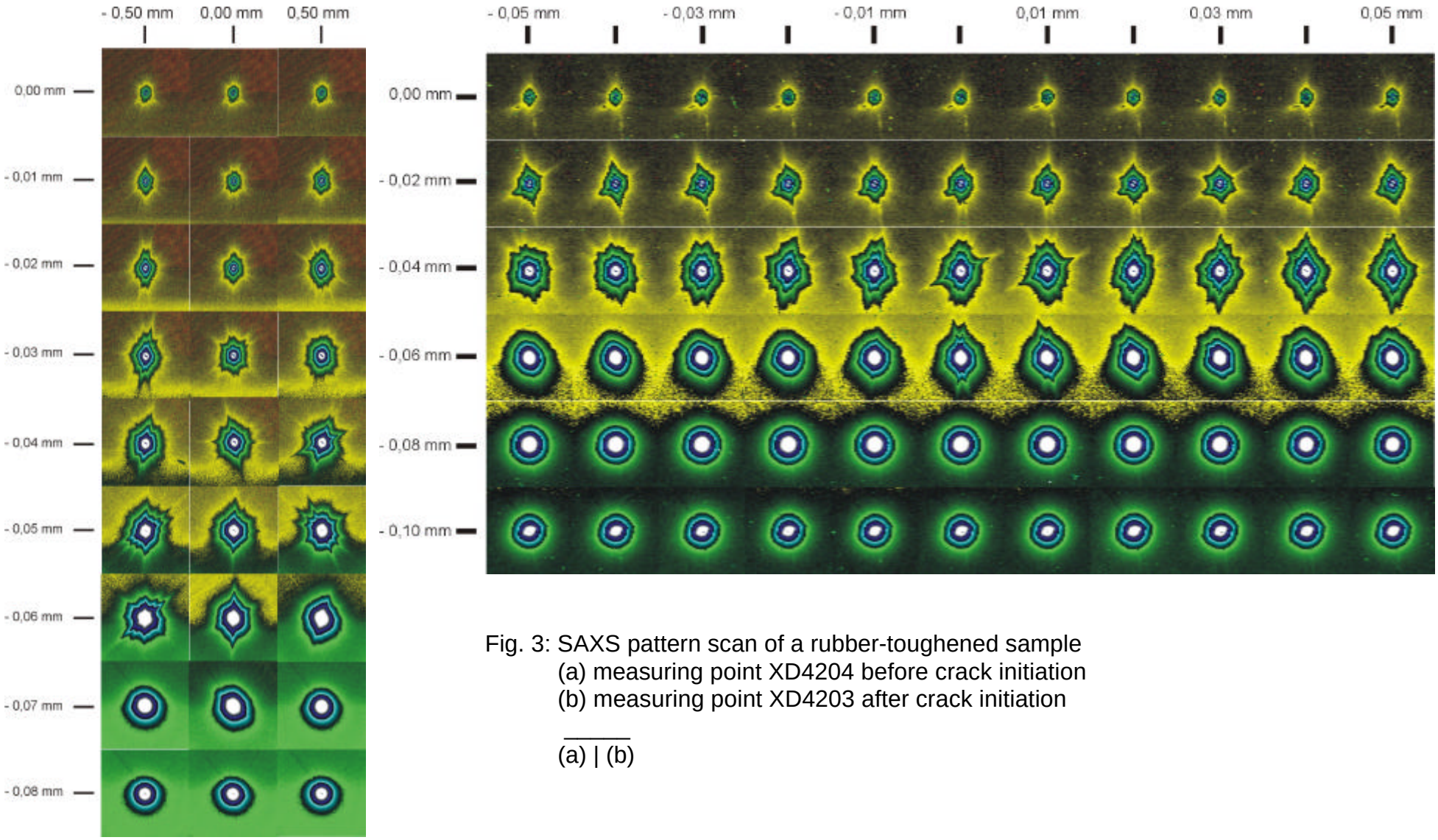

Fig. 3: SAXS pattern scan of a rubber-toughened sample
(a) measuring point XD4204 before crack initiation
(b) measuring point XD4203 after crack initiation

(a) | (b)