

The aim of this PhD study was to develop, apply, and validate the Diffusive Gradients in Thin Films (DGT) technique for aqueous U speciation in the vicinity of the former uranium mining sites. Despite the advancement in understanding of the metal speciation concept and the tremendous progress in the development of various speciation methods, the biogeochemical cycling of trace metals, such as uranium, has not yet been fully described and understood. This research addressed the requirement of the development of an *in situ* speciation and monitoring tool, which yields information not only about uranium speciation, but it can be also used as a predictor of uranium bioavailability. For the first time, the DGT technique was engaged in the investigation on the U speciation in the vicinity of the former uranium mining sites. Therefore, this study successfully attempted filling several knowledge gaps about determination of uranium using the DGT technique and its general application in mining influenced environments.

In the first step, a comprehensive laboratory DGT investigation using Chelex[®]-100, Metsorb[™], Diphonix[®] and two experimental resins was carried out. The influence of parameters such as pH, ionic strength, effective capacity, presence of interferences on the quantitative U accumulation by the DGT technique was explored. Subsequently, the DGT technique was validated by several *in situ* DGT field trials which were carried out in both, natural and mining influenced waters. Over the course of the study, a multi-technique approach, that employed filtration, ultrafiltration coupled with the geochemical modelling speciation and DGT technique, was adopted in order to obtain a comprehensive knowledge about the speciation of U in mining impacted environments. The feasibility of the DGT technique as a predictor of potentially bioavailable U fraction was explored as well.

The combination of the laboratory experiments, field studies and speciation modelling allows obtaining a better understanding of the applicability of the DGT technique as an environmental monitoring and speciation tool in mining environments.

Nederlandstalige abstract

In deze thesis werd de beoogde Diffusieve Gradients in Dunne Lagen Techniek (DGT) te ontwikkelen, toe te passen en te valideren voor de analyse van opgelost uranium species in de omgeving van vroegere uranium mijnen. Ondanks de vooruitgang die reeds geboekt werd in de ontwikkeling van speciatietechnieken is de biogeochemische cyclus van metalen zoals uranium weinig gekend. Er is nood aan de ontwikkeling van *in situ* monitoringstechnieken die informatie verschaffen over de speciatie en biobeschikbaarheid van uranium. In deze thesis werd de DGT techniek voor het eerst toegepast in de omgeving van uranium mijnen. Laboratorium experimenten hebben de invloed van de specifieke condities van de uranium mijnen op de DGT respons onderzocht. Verschillende nieuwe harsen werden onderzocht op hun toepasbaarheid voor uranium metingen met DGT. De DGT speciatie werd vergeleken met filtratie en ultrafiltratietechnieken alsook met geochemische modelisatie. De gecombineerde aanpak laat ons toe een beter inzicht te bekomen in de geschiktheid van DGT als een monitorings- en speciatieinstrument in de omgeving van uranium mijnen.