





Leaching in Cold Recycled Materials

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Overview

WP5: Environmental Evaluation of Materials Used for Cold Recycled Mixes

Objective:

To determine the full extent of potential environmental benefits associated with the use of cold-mixed bitumen stabilized materials by cold-recycling techniques.

Overview

- Focus on leaching behaviour
- Identified tasks included:
 - Selection of appropriate test methods
 - Selection of appropriate materials
 - Testing
 - Evaluation

Experimental Testing

Key Issue: No clear test for characterising leaching behaviour of cold recycled materials

- Evaluation of existing tests required
- Assessment of suitability for cold recycled materials

Experimental Testing

Potential Tests were:

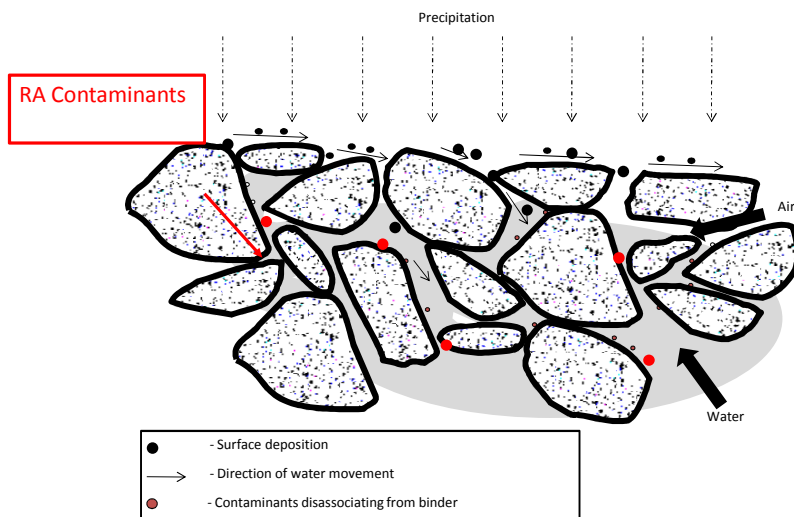
- Batch Test: EN 12457-4
- Batch Test: ISO/TS 21268-1
- Percolation Test:
 - Draft Method - CEN TC 351
 - CEN/TS 14405
- Tank Test
- EN 1744-3
- ER-H (Re-circulating column)

Experimental Testing

Evaluation Criteria

- Standardised EN Test
- Sample description (particle size)

Water Movement Through Asphalt



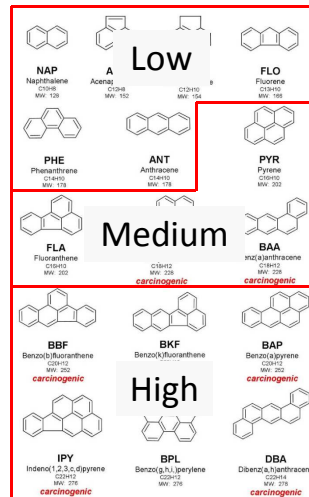
Experimental Testing

Evaluation Criteria

- Standardised EN Test
- Sample description (particle size)
- Test duration
- L/S Ratio
- No of Analyses required
- Suitability for PAH detection

Contaminants of Interest

- 16 EPA Priority PAHs:
 - Carcinogenic
 - Mutagenic
- Selected heavy metals:
Copper, Zinc , Nickel, Lead,
Manganese, Cobalt, Cadmium.



Source: Gregory J Sower, Spatial and temporal variations of bioavailable polycyclic aromatic hydrocarbons in the Lower Willamette River. Oregon State University. Doctoral Thesis / Dissertation, 2008, 80 Pages Doctor of Philosophy (Ph.D.)

Experimental Testing

Test Method	EN Test	Max Particle Size (mm)	Duration (days)	L/S Ratio	No of Analyses
Batch Test: EN 12457-4	Yes	10	1	10	1
Batch Test: ISO/TS21268	No	4	1	2	1
Percolation Test:	No	10	≈ 28-30	10	7
- CEN TC 351					
- CEN/TS 14405					
Tank Test	No	Monolith	64	-	8
EN 1744-3	Yes	32	1	10	1
ER-H	No	-	7	2	1

Experimental Testing

Preferred Tests

- Batch Test to EN 12457-4

Also of Interest

- Percolation Test
- Tank Test

Experimental Testing

Analytical Methods

- Gas Chromatography – Mass Spectrometry (GC-MS): used for analysis of PAH release
- Inductively Coupled Plasma – Mass Spectrometry (ICP-MS): used for analysis of heavy metals

Other Issues:

- Safety: Use of sodium azide biocide

Experimental Testing

Materials:

- Tar containing asphalt
 - Evaluate encapsulation effect
 - Site-sourced material
- Crumbed rubber modified asphalt

Experimental Testing

Materials

- Czech Republic
 - Reclaimed asphalt from Czech site
 - Foam-mix manufactured using this material
 - Emulsion mix from Czech job site
 - As above, with 10 % mechanically activated fly ash (Plzen) and 3.5 % bitumen emulsion
 - As above, with 10 % mechanically activated fly ash (Hodonin) and 3.5 % bitumen emulsion

Experimental Testing

Materials

- Germany
 - Tar material, cold recycled in 1991
 - Cores extracted for CoRePaSol
 - Cold recycled tar base course
 - Cold recycled tar base & binder course
 - Emulsion mix using cold recycled tar base

Experimental Testing

Materials

- Ireland
 - Reclaimed asphalt from Irish site
 - Emulsion mix manufactured using this material
- Portugal
 - Crumb-rubber modified asphalt
 - Emulsion mix containing this material

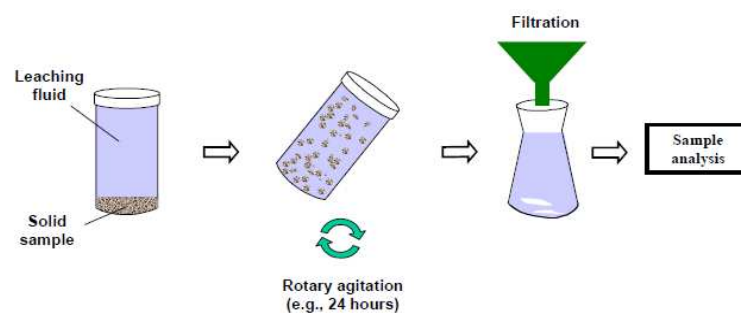
Experimental Testing

Material Preparation

- Based on Re-Road experience
- Large pieces gently heated ($\approx 80^{\circ}\text{C}$)
- Crumbled when warm
- Frozen at -20°C for 8 - 12hrs
- Passed through jaw crusher
 $\leq 10\text{mm}$ particle size



Batch Test Procedure



5 steps procedure:

1. Place test sample in the glass bottle
2. Add leachant (L/S) = 10 l/kg – 0.01% solution of CaCl_2 and NaN_3 in de-ionised water
3. Agitate for 24h, at about 10rpm
4. Filtration/sample extraction
5. Analysis.

Experimental Testing

Percolation Test

- 10 mm particles are packed into 3 glass columns in five separate layers; + 1 blank column
- Upflow test, continuous linear velocity of 15 ± 2 cm/day (measured through an empty column)
- Test duration is approximately 30 days, concluding once a terminal, cumulative liquid-to-solid (L/S) ratio of 10 l/kg was achieved.
- Samples taken at 7 discrete intervals

Percolation Test (TC 351)



Percolation Test: TS 14405



Results

- Batch Test
 - Czech RA
 - Irish RA
 - German cold recycled tar
 - Portuguese crumb rubber modified asphalt
- Percolation Test
 - German cold recycled tar
 - Portuguese crumb rubber modified asphalt
- Tank Test
 - Czech fly ash mixes

Batch Test: Czech RA

Compound	Detection Limit (µg/l)	Quantity Leached (µg/l)	
		B1: Czech RA	B6: Foam Mix with Czech RA
Naphthalene	<0.014	0.095	2.078
Acenaphthylene	<0.013	<0.013	<0.013
Acenaphthene	<0.013	0.025	0.106
Fluorene	<0.014	0.016	0.082
Phenanthrene	<0.011	0.029	0.234
Anthracene	<0.013	<0.014	<0.015
Fluoranthene	<0.012	0.037	0.110
Pyrene	<0.013	0.037	0.060
Benzo(a)anthracene	<0.015	0.008	0.018
Chrysene	<0.011	0.008	0.018
Benzo(bk)fluoranthene	<0.018	0.021	0.021
Benzo(a)pyrene	<0.016	0.013	0.007
Indeno(123cd)pyrene	<0.011	0.008	<0.011
Dibenzo(ah)anthracene	<0.01	<0.01	<0.01
Benzo(ghi)perylene	<0.011	0.013	<0.011
PAH 16 Total	<0.195	0.26	2.73

Batch Test: Irish RA

Compound	Detection Limit (µg/l)	Quantity Leached (µg/l)		
		B2: Irish RA	B7: Foam Mix with Irish RA	B8: Emulsion Mix with Irish RA
Naphthalene	<0.014	10.593	0.904	4.508
Acenaphthylene	<0.013	0.286	0.266	0.089
Acenaphthene	<0.013	18.387	5.419	5.507
Fluorene	<0.014	7.133	2.225	2.127
Phenanthrene	<0.011	18.950	8.535	3.975
Anthracene	<0.013	2.498	1.978	0.436
Fluoranthene	<0.012	11.605	9.557	0.793
Pyrene	<0.013	9.415	8.888	0.516
Benzo(a)anthracene	<0.015	3.289	4.130	0.047
Chrysene	<0.011	3.375	4.577	0.066
Benzo(bk)fluoranthene	<0.018	5.868	10.884	0.052
Benzo(a)pyrene	<0.016	4.203	7.414	0.038
Indeno(123cd)pyrene	<0.011	2.551	6.401	0.019
Dibenzo(ah)anthracene	<0.01	0.246	0.510	<0.01
Benzo(ghi)perylene	<0.011	2.881	5.987	0.028
PAH 16 Total	<0.195	101.26	77.68	18.20

Batch Test: German Cores

Compound	Detection Limit (µg/l)	Quantity Leached (µg/l)			
		B3: German Cold Recycled Tar base	B4: As B3, with Binder Course	B9: Foam Mix containing B3 as aggregate	B10: Emulsion Mix containing B3 as aggregate
Naphthalene	<0.014	3.811	0.999	0.066	<0.014
Acenaphthylene	<0.013	0.188	<0.013	0.078	<0.013
Acenaphthene	<0.013	5.115	<0.013	0.157	2.030
Fluorene	<0.014	2.923	<0.014	0.072	<0.014
Phenanthrene	<0.011	10.170	1.027	0.707	14.604
Anthracene	<0.013	0.321	0.263	0.353	2.371
Fluoranthene	<0.012	4.927	2.046	3.923	19.498
Pyrene	<0.013	2.786	1.829	4.322	13.088
Benzo(a)anthracene	<0.015	1.328	1.535	2.624	10.990
Chrysene	<0.011	1.302	1.529	3.045	12.248
Benzo(bk)fluoranthene	<0.018	1.972	2.854	6.855	18.916
Benzo(a)pyrene	<0.016	1.086	1.524	3.811	9.864
Indeno(123cd)pyrene	<0.011	0.582	1.115	2.506	6.295
Dibenzo(ah)anthracene	<0.01	0.129	<0.01	0.640	0.793
Benzo(ghi)perylene	<0.011	0.534	0.576	1.536	0.650
PAH 16 Total	<0.195	37.03	17.04	31.02	116.75

Comparison: German Cores

Parameter	Quantity Leached (µg/l)					
	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Average
Original Coal Tar Asphalt						
PAH 16	98.7	119.4	194.9	112.0	102.8	125.6
PAH 6	66.3	70.0	110.0	64.0	58.3	73.7
Benzo(a)pyrene	18.0	20.0	29.0	17.0	17.0	20.2
Cold Recycled Tar (conducted in 1991)						
PAH 16	14.2	23.6	37.2	45.7	17.1	27.6
PAH 6	7.9	11.8	18.6	23.6	9.2	14.2
Benzo(a)pyrene	2.0	2.8	4.0	4.6	1.8	3.0
Cold Recycled Tar (conducted in 2014)						
PAH 16	38.71	35.18	37.21			37.03
PAH 6	9.50	8.32	9.06			8.96
Benzo(a)pyrene	1.18	0.97	1.11			1.09

Batch Test: Portuguese CRMA

Polycyclic Aromatics Hydrocarbons	QL (GC-MS) (µg/l)	B5 (RAR) leachate (µg/l)	B11 (RAR-BE) leachate (µg/l)
Naphthalene	0.100	<0.100	<0.100
Acenaphthylene	0.010	<0.010	<0.010
Acenaphthene	0.010	<0.010	<0.010
Fluorene	0.020	<0.020	<0.020
Phenanthrene	0.030	<0.030	0.032 ^(a)
Anthracene	0.020	<0.020	<0.020
Fluoranthene	0.030	<0.030	<0.030
Pyrene	0.060	<0.060	<0.060
Benz(a)anthracene	0.010	<0.010	<0.010
Chrysene	0.010	<0.010	<0.010
Benzo(b)fluoranthene	0.010	<0.010	<0.010
Benzo(k)fluoranthene	0.010	<0.010	<0.010
Benzo(a)pyrene	0.020	<0.020	<0.020
Indeno(1,2,3-cd)pyrene	0.010	<0.010	<0.010
Benzo(g,h,i)perylene	0.010	<0.010	<0.010
Dibenz(a,h)anthracene	0.010	<0.010	<0.010
Sum of 16 PAH	0.370	<0.370	<0.370

Batch Test: Portuguese CRMA

Metals	Limit values for waste acceptable at landfills for inert waste (L/S=10 l/kg) (mg/kg dry substance)	B5 (RAR) leachate		B11 (RAR-BE) leachate	
		Analysed by ICP-AES (mg/kg)	Evaluation	Analysed by ICP-AES (mg/kg)	Evaluation
Cadmium, Cd	0.04	<0.02	✓	<0.02	✓
Cobalt, Co	-	<0.20	(n/a)	<0.20	(n/a)
Copper, Cu	2	<0.13	✓	<0.13	✓
Lead, Pb	0.5	<0.09	✓	<0.09	✓
Manganese, Mn	-	<0.13	(n/a)	<0.13	(n/a)
Nickel, Ni	0.4	<0.06	✓	<0.06	✓
Zinc, Zn	4	<0.06	✓	0.26	✓

Percolation Test: German Cores

Sample Data:

Compound	Quantity Leached (µg/kg)						
	1st Extraction	2nd Extraction	3rd Extraction	4th Extraction	5th Extraction	6th Extraction	7th Extraction
Naphthalene	0.52	0.96	6.77	2.97	10.24	23.46	59.40
Acenaphthylene	0.04	0.06	0.33	0.13	0.23	0.50	0.60
Phenanthrene	0.26	0.64	4.03	2.08	6.39	14.72	40.70
Pyrene	0.05	0.09	0.59	0.33	0.94	1.79	4.52
Benzo(a)anthracene	0.01	0.01	0.06	0.04	0.12	0.18	0.40
Chrysene	0.01	0.01	0.07	0.04	0.12	0.21	0.47
Benzo(k)fluoranthene	0.01	0.00	0.01	0.01	0.04	0.05	0.15
Benzo(a)pyrene	0.00	0.00	0.01	0.01	0.03	0.03	0.09

Percolation Test: Portuguese CRMA

Polycyclic Aromatics Hydrocarbons	QL by GC-MS	P2 (RAR) leachate	P3 (RAR-BE) leachate
	(µg/l)	1 st fraction volume (on a total of seven); (L/S=0.1 l/kg) (µg/l)	1 st fraction volume (on a total of seven); (L/S=0.1 l/kg) (µg/l)
Naphthalene	0.100	<0.100	<0.100
Acenaphthylene	0.010	<0.010	<0.010
Acenaphthene	0.010	<0.010	<0.010
Fluorene	0.020	<0.020	<0.020
Phenanthrene	0.030	<0.030	<0.030
Anthracene	0.020	<0.020	<0.020
Fluoranthene	0.030	<0.030	<0.030
Pyrene	0.060	<0.060	<0.060
Benz(a)anthracene	0.010	<0.010	<0.010
Chrysene	0.010	<0.010	<0.010
Benzo(b)fluoranthene	0.010	<0.010	<0.010
Benzo(k)fluoranthene	0.010	<0.010	<0.010
Benzo(a)pyrene	0.020	<0.020	<0.020
Indeno(1,2,3-cd)pyrene	0.010	<0.010	<0.010
Benzo(g,h,i)perylene	0.010	<0.010	<0.010
Dibenz(a,h)anthracene	0.010	<0.010	<0.010
Sum of 16 PAH	0.370	<0.370	<0.370

Tank Test: Czech Fly Ash Mixes

Leaching of analysed elements (mg/m ²)	Sample T1	Sample T2	Sample T3	BMD 64 days	Leaching from Landfilled waste(*) (64 days)
Chloride	2 810	2 553	2 516	-	10 000
Sulfate	2 390	71 397	126 308	27 000	10 000
Arsenic	0.45	9	2.48	41	1.3
Cadmium	0.09	0.086	0.085	1.1	0.2
Chromium	0.6	0.6	0.6	140	5
Copper	4.3	4	3.6	51	45
Mercury	0.03	0.03	0.02	0.4	0.1
Nickel	0.9	0.9	0.8	50	6
Lead	0.2	0.31	0.3	120	6
Zinc	8.3	5.2	6.1	200	30

Conclusions

Materials

- Large variation in PAH levels detected
 - Issue with Legacy Roads
- Importance of preliminary screening
 - PAK marker
- Crumb rubber – low PAH, some inorganics

Conclusions

Encapsulation effect

- Present, but not proven
 - German tar cores (1991 vs 2014)
 - Portuguese CRMA
- Sample preparation an issue

Conclusions

Application Area

- Majority of water flows off pavement surface
 - Limited percolation
- Location of tar in pavement structure an important consideration
 - Related to L/S ratio
- Continuous contact with water the key driver for leaching

Conclusions

Test Methods

- No ideal method for assessing PAH release
 - TC 351 – equilibrium not reached
 - Tank test – good for inorganics
 - Batch test
- Sample preparation/test set-ups available do not reflect in-service application of materials